

# Dependence of the Laminar Burning Velocity of Methane, Propane and Ethylene on Initial Temperature and Inert Diluent Concentration

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## Abstract

Through its influence on flamelet combustion, the laminar burning velocity is a crucial parameter in describing turbulent combustion. Calculations with a single chemical kinetics mechanism have determined the effect of equivalence ratio and initial temperature on the burning velocities of methane, ethylene and propane in air at atmospheric pressure. These results have been checked against available data in the literature, and then used to extrapolate to the much higher temperatures that are generally of interest in recirculating flows. Simple correlations have been developed to describe them to within 10% over the temperature range from 300K to 1100K in a convenient form for modeling purposes. Dilution of fuel by carbon dioxide is of interest in the context of synthetic and biogas fuels, and so its effect has also been computed. Comparison with dilution by nitrogen indicates the extent of CO<sub>2</sub>'s role as a reactive species.

## Introduction

The unstretched laminar burning velocity  $S_L$  is of significance not only for laminar flames, but through flamelet concepts, for turbulent combustion and extinction. Many researchers have determined experimentally the burning velocity of the common gaseous fuels, methane, propane and ethylene in air, when the reactants are initially at room temperature. There are fewer data for elevated initial temperatures, particularly for non-stoichiometric mixtures, even though in practical systems there is usually either internal or external preheat. Additionally, there is increasing interest in synthetic or biogas fuels, where a predominantly methane product is diluted by carbon dioxide. The extent to which this is an inert diluent can be tested by comparison with nitrogen dilution.

There are therefore several incentives to generate a comprehensive and consistent data bank for the burning of CH<sub>4</sub> at elevated temperatures and with CO<sub>2</sub> or N<sub>2</sub> dilution. At least for laboratory purposes, similar data for C<sub>3</sub>H<sub>8</sub> and C<sub>2</sub>H<sub>4</sub> are desirable. The approach in this paper is to compute the variations of  $S_L$  with a chemical kinetics model, adapted from that coded by Wang [1], checking against experimental data where these are available.

## Specific Objectives

- To compute the burning velocity of methane, ethylene and propane at atmospheric pressure over the full range of lean equivalence ratios, as a function of initial temperatures from 300K upwards.
- To compute the effect of CO<sub>2</sub> and N<sub>2</sub> addition to these fuels and to examine the extent to which CO<sub>2</sub> may be regarded as an inert diluent.
- To correlate the computed results so as to provide simple expressions of adequate accuracy for incorporation in flame models.

## Calculation Models

Detailed chemical kinetic calculations were performed using an extended version of the PREMIX laminar 1-D flame code and the Chemkin-II suite of programmes [2]. The specific extension used in this work involved the facility to increase the initial temperature and utilise a previous converged solution as the initial solution to reduce the CPU time required. The flame simulation covered a 20cm domain and in excess of 150 grid nodes was used to ensure that any grid dependence of the solution was reduced. Mixture averaged transport properties were used from the existing Chemkin database. Initially the well-characterised and tested GRI Mech 3.0 [3] kinetic model was used. Whilst this mechanism gave

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an excellent simulation for methane there were significant deviations for both propane and ethylene. GRI Mech 3.0 overpredicts the burning velocity for these fuels.

Recently a mechanism has been published, by Wang and co-workers, that builds upon GRI Mech 3.0 but extends it with specific focus on C3 chemistry [4]. This mechanism was adapted for use in this study by removing C4 chemistry to reduce the overall size of the model to 323 reactions and 46 species. Transport and thermodynamic data were obtained from web download of the Wang model [1].

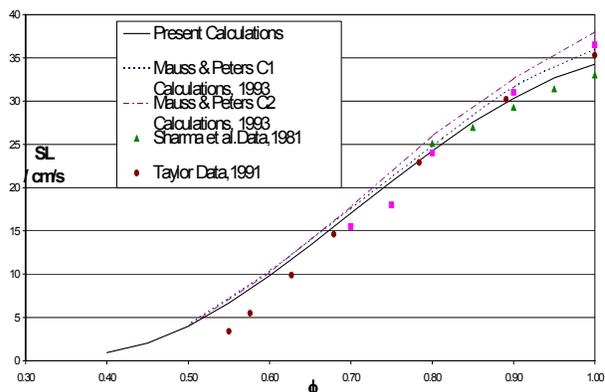
A key aspect of this paper is the modelling of burning velocity at elevated temperature. As expected, burning velocity increases with increasing temperature but the simulation results tend towards those for a plug flow reactor (PFR). This shows that transport properties and thermal diffusion become less important as the temperature increases. Indeed, the reaction front moves closer to the boundary as the temperature increases and this has a significant effect on the production of a stable converged solution and the time taken to reach this solution. For this reason it is appropriate to place an upper temperature limit on the results.

### Results for Pure Fuels

To establish confidence in the validity of the calculations, they were first compared with experimental data for the pure fuels at an initial temperature of 300K. While many data sets are available in the literature, those of two schools were chosen because they both cover more than one fuel and hence have an internal consistency. Taylor [5] used spherical bomb apparatus at a British Gas research laboratory (now at the University of Leeds), while Vagelopoulos and Egolfopoulos at the University of Southern California have made measurements in impinging jet flames [6]. As seen in Figs 1-3, the two sets are generally in good agreement. For  $C_2H_4$  these are supplemented by an earlier study by Egolfopoulos et al. [7] on counter-flowing jets, from which results down to  $\phi=0.5$  have recently been quoted by Hirasawa et al. [8]. This last investigation confirms the earlier data for  $\phi>0.7$ , where they also agree with Taylor [5], but they are lower than his for the leaner mixtures (Fig. 2).

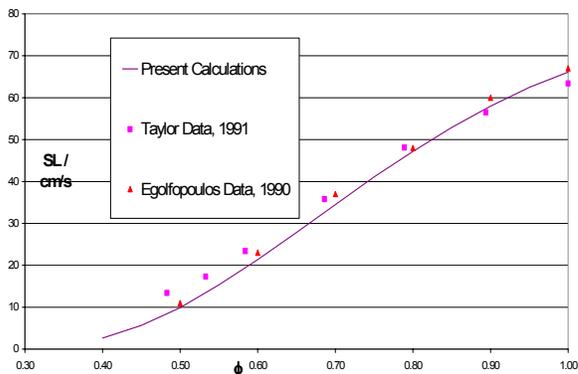
In Fig. 1 for  $CH_4$  the experimental data of Sharma et al [9] acquired in a spherical bomb are also plotted. They measured slightly lower burning velocity under

Fig. 1: Methane/Air Mixtures: Dependence on Equivalence Ratio for 300K



stoichiometric conditions than the other two groups. Also shown are the calculated results of Mauss and Peters [10], who used C1 and C2 schemes to compute the dependence on equivalence ratio. Our own calculations with a C1 scheme were almost in precise agreement, but with the Wang scheme for C1, C2 and C3 (which proved much more satisfactory for  $C_2H_4$  and  $C_3H_8$ ),  $S_L$  at stoichiometric conditions was some 6% lower than the Mauss and Peters calculations with the C2 mechanism. The agreement in the three calculations at lean equivalence ratios is almost perfect, but as seen in Fig. 1, all three predictions are higher than the measured  $S_L$  of Taylor [5] at  $\phi=0.6$ . This may be due to difficulties in the experiments close to lean extinction. No stable laminar  $CH_4$  flames without preheat, pilot flame or recirculation have been reported for  $\phi<0.5$ , but in the numerical calculations, extinction does not occur until  $\phi=0.4$ .

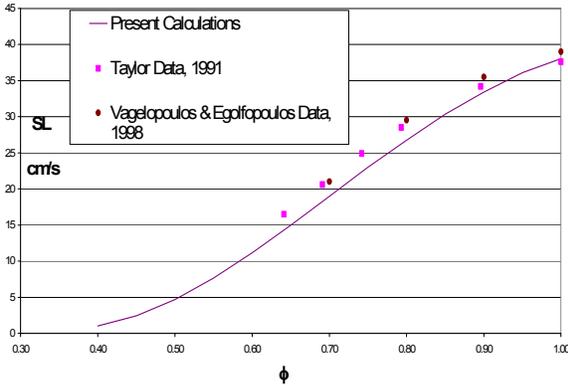
Fig. 2: Ethylene/Air Mixtures: Dependence on Equivalence Ratio for 300K



The experimental data for  $C_2H_4$  (Fig. 2) are well-predicted down to  $\phi=0.5$ . However, the data for  $C_3H_8$  (Fig. 3) suggest somewhat faster burning than the predictions down to the leanest  $\phi$  at which

measurements were made (0.6). Results were also generated for those fuels with initial temperatures up to 1100K.

**Fig. 3: Propane/Air Mixtures : Dependence on Equivalence Ratio for 300K**

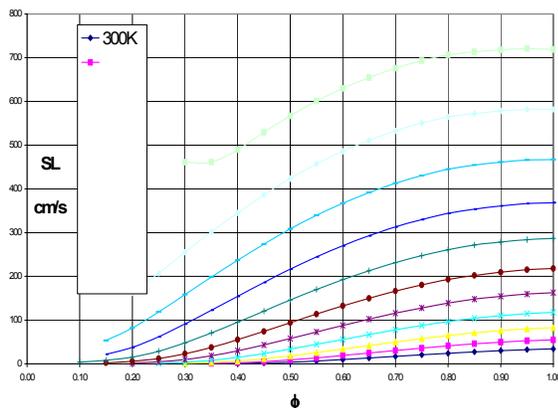


Overall, the performance of the present calculation scheme over the range of the data for  $T_i=300K$  must be regarded as very satisfactory, but corroboration of behaviour close to lean extinction is lacking.

Confidence is enhanced, however, when the dependence on the initial temperature is compared with one of the few data sets available, that of Sharma et al. [9] for  $CH_4$ . The predicted increase in burning velocity with temperature is shown in Fig. 4 up to 1300K, even though there are substantial uncertainties in the calculations at the higher temperatures, as already discussed. Sharma et al. chose to correlate their data for  $T_i$  up to 600 K in the form

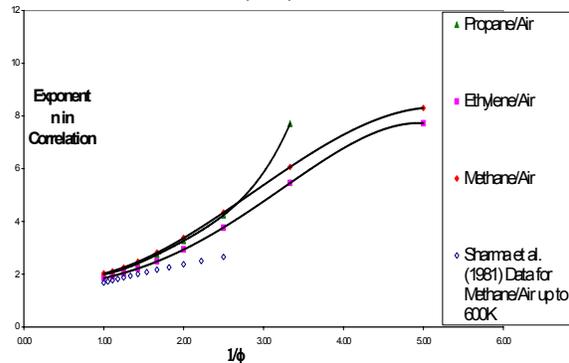
$$S_L = C(\phi) \left( \frac{T_i}{300} \right)^{n(\phi)}$$

**Fig. 4: Predicted Burning Velocity of Methane/Air Mixtures as a Function of Initial Temperature**



This simple form was sometimes inadequate to describe our calculations beyond  $T_i=1100 K$  at  $\phi$  near unity, so the correlations have been limited to the range  $300K < T_i < 1100K$ , within which they represent the calculated results to within 10%, with much smaller discrepancies arising at the lower equivalence ratios. This form of correlation was adopted here, in spite of the loss of accuracy, because of its simplicity. For a representation of the predictions to within 2% in the limited range  $300K < T_i < 600K$ , another set of coefficients for the same equation was produced.

**Fig. 5: Exponent of Temperature in Burning Velocity Correlation for  $300K < T_i < 1100K$**   
 $SL = C(T/300)^n$



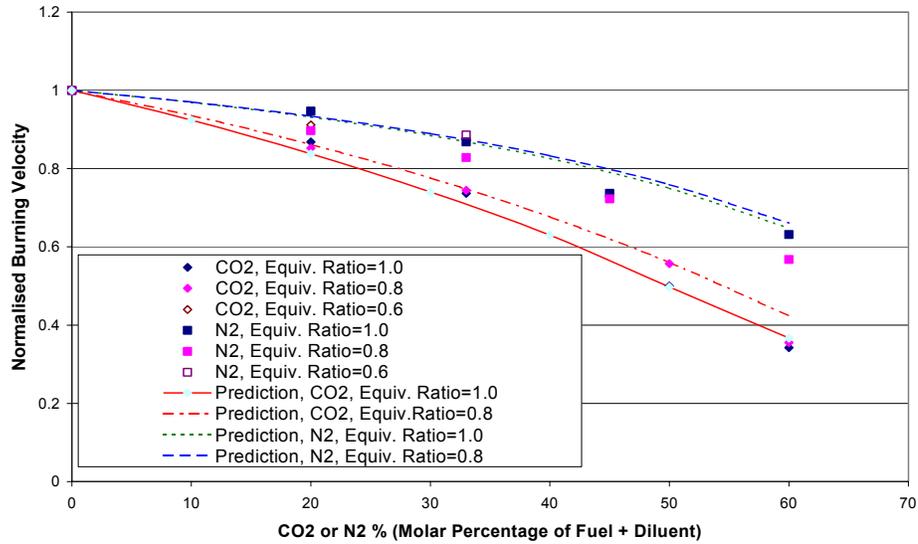
Comparison with the experimental data of Sharma et al. [9] for the exponent  $n(\phi)$  is made in Fig. 5, where the predictions are seen to have similar dependence on equivalence ratio. Using the correlation for the restricted temperature range (corresponding to the experiments but not shown on Fig. 5),  $n(\phi)$  is only 6% greater than the experimental data for  $0.8 < \phi < 1.0$ . Fig. 5 also shows that the predicted temperature exponents for  $C_2H_4$  and  $C_3H_8$  are similar in level and have similar dependence on equivalence ratio to that of  $CH_4$ .

Quartic and cubic polynomial curve fits were then made to  $C(\phi)$  and  $n(1/\phi)$ , respectively. The results for the range  $300K < T_i < 1100K$  are given in Table 1, and those for  $300K < T_i < 600K$ , in Table 2.

### Results for Diluted Fuels

Computations have also been made for the effect of dilution of all three fuels by  $CO_2$ , and for methane by  $N_2$ . Normalising by the undiluted predictions, the results for methane with  $T_i=300K$  are shown in Fig. 6 where they are compared with the experimental data of Clarke et al. [11]. These researchers carried out

**Fig. 6: Predicted Effect of Dilution by CO<sub>2</sub> or N<sub>2</sub> on the Burning Velocity of Methane in Air at Ti=300K compared with the Experimental Data of Clarke et al. (1995)**



free-fall experiments on the combustion of natural gas in a spherical bomb. Up to 60% dilution, the predictions of the reduction in burning velocity are again within 10% of the data for both CO<sub>2</sub> and N<sub>2</sub>.

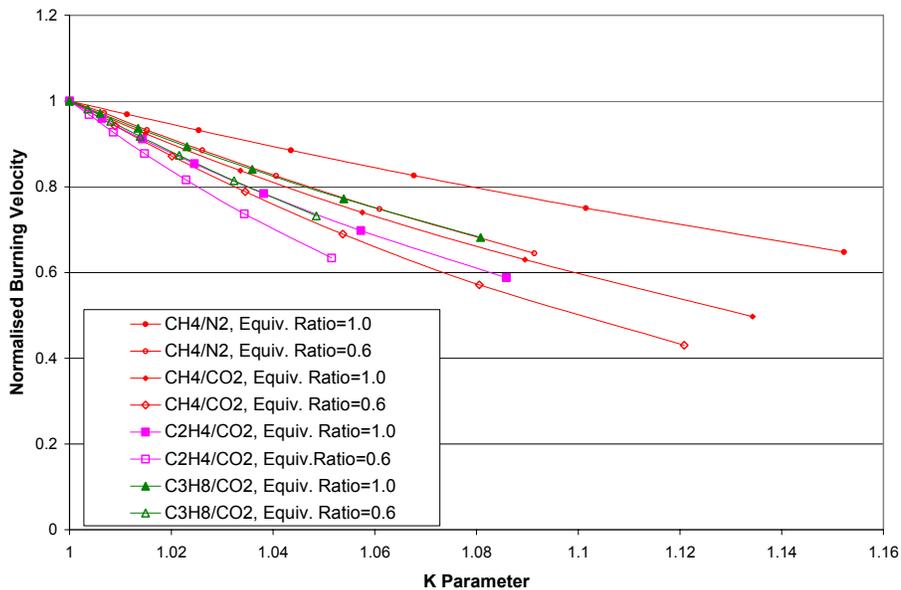
As expected, the dilution effect of CO<sub>2</sub> and N<sub>2</sub> is not the same on a molar basis. If the diluent were inert, a parameter that correlated the effect on the adiabatic flame temperature might also correlate the

effect on the burning velocity, even though other thermo-physical properties apart from the heat capacity come into play in its determination. To a first approximation, it can be shown that such a parameter is

$$K \equiv 1 + \frac{Y_d}{Y_f} \frac{Y_{st0}}{(1 - Y_{st0})} \phi \frac{C_{pd}}{C_{pa}} = \frac{T_{ad0} - T_i}{T_{ad} - T_i}$$

where  $Y_f$  and  $Y_d$  are the mass fractions of fuel and

**Fig. 7: Predicted Effect of Dilution by CO<sub>2</sub> or N<sub>2</sub> on the Burning Velocity of Methane, Ethylene and Propane in Air at Ti=300K**



diluent in the mixture,  $Y_{st0}$  is the stoichiometric mass fraction in the absence of diluent, and  $C_{pa}$  and  $C_{pd}$  are the specific heat capacities of air and diluent. The predicted effect on the non-dimensional temperature rise (where  $T_{ad0}$  is the adiabatic temperature in the absence of diluent) generated in the course of the burning velocity calculations was compared with the dilution parameter and found to be a good approximation for methane and propane under the leaner conditions for which  $T_{ad0}$  was below about 1700K: however, it was only indicative of the calculated effects for stoichiometric conditions, and for ethylene over the full range of equivalence ratios.

With the above observations, it is perhaps not surprising that the parameter is only partially successful in correlating the results for burning velocity (Fig. 7). The equivalence ratio remains a parameter of independent importance, as does the nature of the diluent, but the fuel dependence is almost eliminated. The reduction in burning velocity with  $N_2$  dilution is less than with  $CO_2$  for the same value of  $K$ . Clearly  $CO_2$  is not operating as an inert diluent with the high adiabatic temperatures (up to 2200K) at stoichiometric conditions. For a given equivalence ratio, its effect on the burning velocity was found to be greatly diminished as the initial temperature increased, even though its effect on the adiabatic temperature ratio remained almost constant.

### Conclusion

Simple correlations have been developed to describe the results of these calculations to within 10% over the initial temperature range from 300K to 1100K, or alternatively, to within 2% over the limited range from 300K to 600K. Comparison with experimental data has confirmed the general validity of the calculations, but their accuracy cannot be rigorously assessed.

### Acknowledgement

Dr D.B.Smith of the University of Leeds provided useful input in the formulation of this study.

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**Table 1:Correlations for the Calculated Results for Pure Fuels over the Full Range of Initial Temperature**

$$S_L = C(\phi) \left( \frac{T_i}{300} \right)^{n(\phi)} \text{ cm/s} \quad 300\text{K} < T_i < 1100\text{K}, \phi \leq 1$$

	$C=c_0+c_1\phi+c_2\phi^2+c_3\phi^3+c_4\phi^4$					$n=n_0+n_1(1/\phi)+n_2(1/\phi^2)+n_3(1/\phi^3)$			
	$c_0$	$c_1$	$c_2$	$c_3$	$c_4$	$n_0$	$n_1$	$n_2$	$n_3$
CH <sub>4</sub>	-0.2	+21.3	-168.7	+390.0	-211.9	2.109	1.101	-1.056	-0.128
C <sub>2</sub> H <sub>4</sub>	+9.7	-63.4	+28.4	+301.8	-217.3	2.340	-1.481	+1.145	-0.127
C <sub>3</sub> H <sub>8</sub>	+2.3	-2.2	-101.4	+322.3	-187.1	+0.051	+2.972	-1.438	+0.371

**Table 2:Correlations for the Calculated Results for Pure Fuels over a Limited Range of Initial Temperature**

$$S_L = C(\phi) \left( \frac{T_i}{300} \right)^{n(\phi)} \text{ cm/s} \quad 300\text{K} < T_i < 600\text{K}, \phi \leq 1$$

	$C=c_0+c_1\phi+c_2\phi^2+c_3\phi^3+c_4\phi^4$					$n=n_0+n_1(1/\phi)+n_2(1/\phi^2)+n_3(1/\phi^3)$			
	$c_0$	$c_1$	$c_2$	$c_3$	$c_4$	$n_0$	$n_1$	$n_2$	$n_3$
CH <sub>4</sub>	+17.9	-107.1	+146.9	+73.1	-97.2	+2.003	-1.237	+1.144	-0.131
C <sub>2</sub> H <sub>4</sub>	+83.7	-558.4	+1203.1	-866.1	+202.6	+1.528	-0.406	+0.507	-0.012
C <sub>3</sub> H <sub>8</sub>	+47.2	-298.0	+590.3	-356.3	+54.1	+1.534	-0.221	+0.386	+0.043