Global reaction mechanism for ethylene flames with preferential diffusion

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1 Introduction

The tremendous progress in combustion science and technology in the past two decades would have been impossible without the simultaneous advances in computational resources. Hence we are now able to simulate with fair confidence simplified combustion phenomena to great detail, using simple fuels for fundamental studies, while engine manufacture industries routinely employ tailored combustion codes for their development work. However, often very complex geometries in 3D CFD simulations require high levels of computational memory, and the need for compact chemical mechanisms capturing the most important physical and chemical features is still as relevant as ever.

New challenges for predictive simulations are related to the yet unknown behaviour of alternative clean fuels, such as bio-fuels. Hence, further development of reliable and practical models that can be implemented into modern computational tools are strongly needed from an industrial point of view. The recent focus on the use of alternative fuels has furthermore increased the timeliness of developing reliable combustion models capable of capturing not only physical parameters such as flame speeds and ignition timing, but also emission levels. Although overall green-house gas emissions are typically low for biomass fuels (carbon neutral fuels), concern need to be addressed regarding high concentrations of heavy metals and significant particle formation associated with such fuels. They are responsible for deposits and harmful products that may hinder the efficient, clean and durable run of these devices.

The formation of particles is located in fuel rich regions during non-premixed combustion such as in diesel engines, or in small fuel rich pockets due to inhomogeneities during combustion e.g. in petrol engines, in particular when running in direct injection mode. In the flamelet [1] or the conditional moment closure (CMC) [2] models this interaction is described by the mean scalar dissipation rate obtained from the flow field. However, the scalar dissipation rate is influenced by the mixing of the reactive scalars; species mass fractions, particle number densities, etc. This is further influenced by the Lewis number which is the ratio of species diffusion and heat flux. This is often assumed to be unity for turbulent diffusion flames and it is argued that the species diffusion is controlled by the turbulent

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mixing. However, in the case of particle formation such as soot, the transport of radicals from the main reaction zone, the inner reaction layer, to the soot formation layer is highly affecting the amount of particle formation and growth. These layers are close to each other and it is expected that, depending on the level of scalar dissipation rate, the mixing characteristics changes when going from turbulent to laminar flows, and preferential diffusion effects become important. The predicted levels of emission in this regime are highly sensitive to this effect, and the details of the transition have yet not been investigated.

Ethylene is a fuel of interest when studying soot formation through the oxidation to acetylene, which can be considered one of the most important particle pre-cursors. Comprehensive and detailed chemical models for C2 hydrocarbons are available and contain over hundreds of chemical elements and several hundred reactions [3, 4]. They are validated for a wide range of combustion targets including flame speeds and ignition. However, often very simplified short mechanisms are sufficient to predict important combustion parameters for given conditions, such as the 10 step ethylene short mechanism by Singh and Jachimowski [5]. Montgomery et al. [6] investigated the performance of a series of reduced n-haptane and ethylene mechanisms compiled from various sources (please see [6] and further references therein). The smallest ethylene mechanism studied was a 6 step mechanism containing 10 species.

We will first demonstrate the effect of preferential diffusion effects on important species for particle formation for flame combustion. Thereafter will we propose a reduced global mechanism for ethylene combustion with 8 reaction steps, including important species such as H, capable of capturing this particular effect for implementation into computational flow simulation software.

2 The flamelet model with preferential diffusion.

We have studied the effect of preferential diffusion using the unsteady flamelet approach [1]. The fast chemistry assumption fails whenever non-equilibrium effects are important. Furthermore, at the flame front there are rapid changes of the scalar dissipation profiles. Therefore the unsteady flamelet model is used to account for these transient effects.

The unsteady flamelet equations can be written as:

$$\rho\frac{\partial Y_i}{\partial \tau} = \frac{\rho\chi}{2Le_i}\frac{\partial^2 Y_i}{\partial Z^2} + \omega_i,$$

$$\rho\frac{\partial T}{\partial \tau} = \frac{\rho\chi}{2Le_i}\frac{\partial^2 T}{\partial Z^2} - \frac{1}{c_p}\sum_{i=1}^{N} h_i\omega_i + \frac{Q_R}{c_p}$$

where the Lewis number, *Le*, is constant but not unity for species *i*, and the scalar dissipation rate is unsteady: $\chi = \chi(t)$. The source term, ω_i, is computed from employing an 81 species ethylene mechanism with 801 reactions [7]. Thermal radiation is included in the energy equation.

Figure 1 shows species profiles and temperature for a given strain rate value based on assuming unity Lewis numbers compared to those where preferential diffusion effects are taken into account. It is not surprising that species such as *H* are significantly influenced by the non unity Lewis number. However, from the figure it is also evident that the particle pre-cursor, acetylene, is significantly influenced by preferential diffusion through the diffusion of *H* from the main reaction zone. Also, the flame temperature is affected through the shift in reaction zone. This will be important when including preferential diffusion in turbulent combustion modelling, particularly in the regime of transition between mixing dominated and laminar conditions.

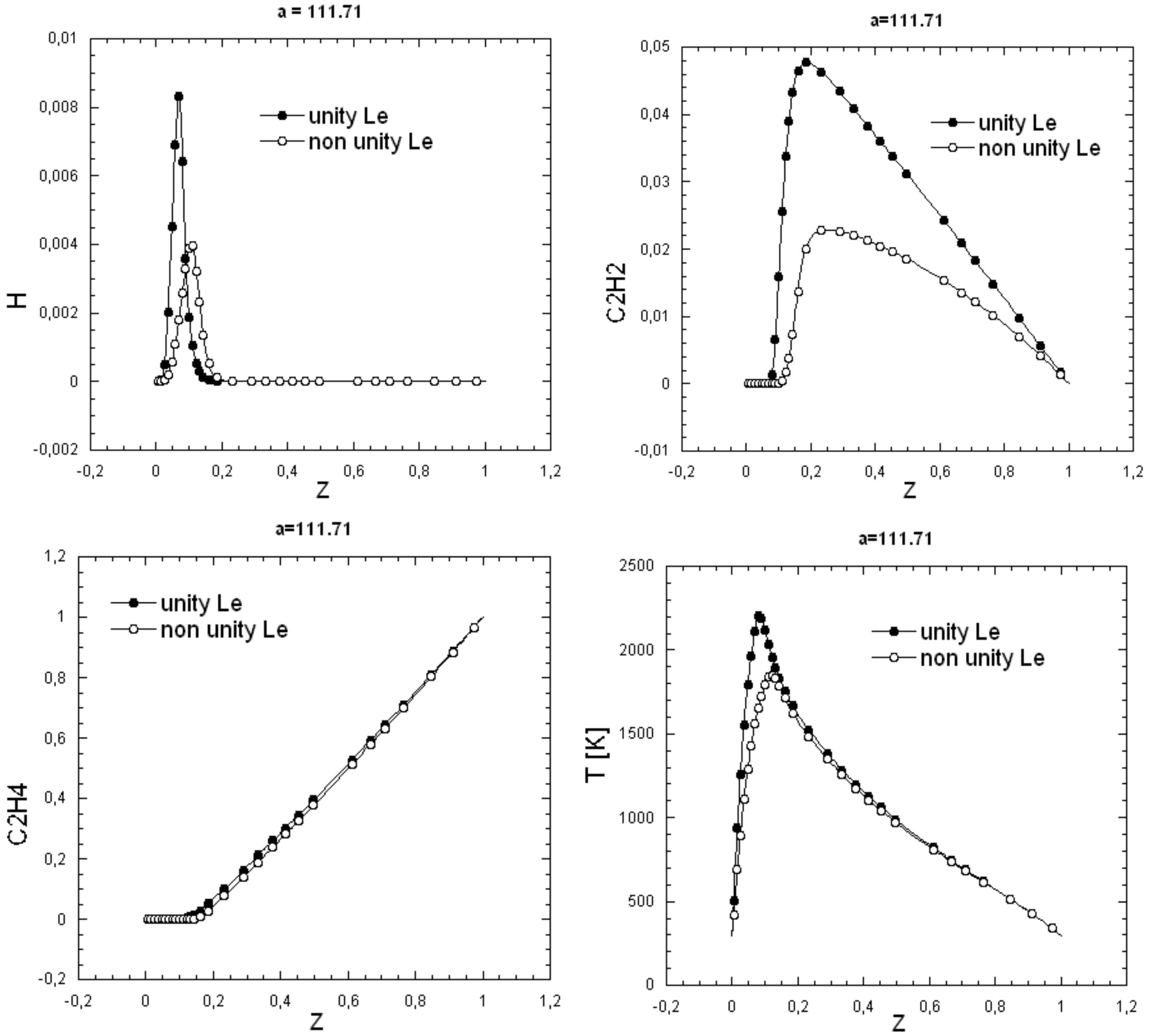

Figure 1: Concentration profiles for hydrogen, acetylene, fuel and temperature as a function of conserved scalar mixture fraction. Solid circles results from employing unity Lewis number for all species, whereas open circles results from non unity Lewis numbers.

3 The reduced global mechanism for ethylene combustion

Accounting for preferential diffusion effects in numerical simulations results in a computational overhead, which may be significant close to extinction and re-ignition limits. We have compiled an 8 step global reaction scheme containing 10 species including inert nitrogen (H, O, OH, CO, CO_2, H_2, H_2O, O_2, C_2H_4 and N_2). The first reaction is the modelled fuel breaking initiating reaction, followed by several elementary reaction steps involving radicals. Reactions 3, 6 and 7 are chain terminating reactions, forming the main products. The rate constants are tuned to give the best reproduction of the important variables such as temperature and the radical H.

Table 1: 8 step global reaction mechanism. Third body efficiencies for H_2=2.5, and for H_2O= 16.0

Reaction	A	n	E_a (cal)
O2+C2H4<=>2CO+2H2	**7.800E+25**	**.000**	**25500.00**
O+CO(+M)<=>CO2(+M)	**5.300E+13**	**.000**	**-4540.00**
OH+CO<=>H+CO2	**4.400E+06**	**1.500**	**-740.00**
O2+H2<=>OH+OH	**1.700E+20**	**.000**	**48000.00**
O2+H<=>OH+O	**2.600E+14**	**.000**	**16800.00**
OH+H2<=>H+H2O	**2.200E+17**	**.000**	**5150.00**
O+H2+M<=>H2O+M	**1.100E+20**	**-2.000**	**0.00**
2H+M<=>H2+M	**2.400E+19**	**-1.000**	**.00**

The performance of the global mechanism is illustrated in Figure 2, for hydrogen and temperature. The results for H are given for both employing unity Lewis numbers and constant Lewis. Furthermore, for comparison the H profile resulting from the 10 step global mechanism of Singh et al. [5] is included. The peak values are very good predicted considering the strong reduction level, for both excluding and including preferential diffusion effects. The temperature profiles are also very well predicted with the 8 step global mechanism compared to the full chemical description. The flame appears a little broader. However the peak temperature value is predicted well. Also here the profile resulting from the 10 step mechanism from Singh et al is shown. The broadening of the flame is also visible here, but in this case also the maximum is under predicted. The remaining profile shapes are reproduced equally well, together with the consumption rate of the fuel (not shown).

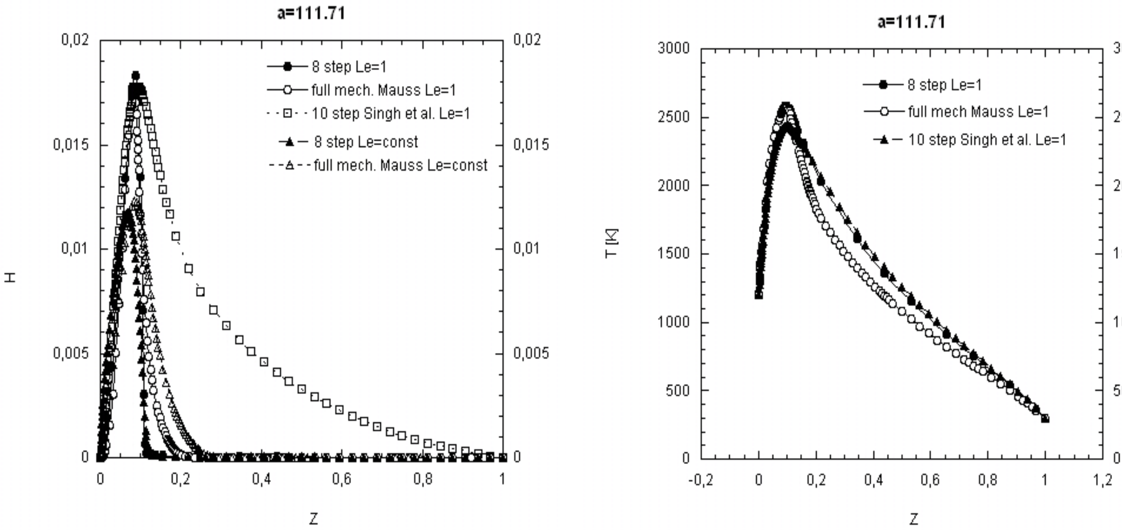

Figure 2. Concentration profiles of hydrogen and acetylene as a function of the conserved scalar mixture fraction. Solid circles represent the detailed mechanism and open circles represent the 5 step mechanism.

4 Conclusion

We have presented a study on the effect of preferential diffusion and its impact on emission levels of important species for particle formation in flames. The effect is significant both on small species such as H, but is also expected on particle pre-cursors such as C_2H_2. We have furthermore proposed an 8 step global mechanism for inclusion in flow simulations to capture the effect of preferential diffusion. Future work will include validations of the effect of preferential diffusion using direct numerical simulations in order to eliminate the impact of modelling and to further establish the effect of preferential diffusion in transition from turbulent to laminar conditions.

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