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Effects of hydrogen and nitrogen on soot volume fraction, primary particle diameter and temperature in laminar ethylene/air diffusion flames

Zhiwei Sun[a,b], Bassam Dally[a,b,*], Graham Nathan[a,b], Zeyad Alwahabi[b,c]

[a] *School of Mechanical Engineering, The University of Adelaide, SA 5005, Australia*
[b] *Centre for Energy Technology, The University of Adelaide, SA 5005, Australia*
[c] *School of Chemical Engineering, The University of Adelaide, SA 5005, Australia*

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ABSTRACT

This study reports the effects of hydrogen (H_2) and nitrogen (N_2) blended into the fuel on soot evolution and flame temperature in axisymmetric ethylene/air diffusion flames at atmospheric pressure. Two series of laminar ethylene (C_2H_4) diffusion flames blended with H_2 and N_2 were experimentally investigated, in which soot volume fraction (f_v), primary particle diameter (d_p) and flame temperature (T) were measured using planar laser-based techniques. These twelve flames are grouped into two sets. In the first one, H_2 or N_2 are added into a constant volumetric flow of C_2H_4 to separate their complementary effects. In the second set, the total volumetric flow rate of the mixture of $C_2H_4/H_2/N_2$ was kept constant for the same ratios of dilution to isolate the influence of exit velocity. For both sets, a reference flame corresponding to a mixture of $C_2H_4/H_2/N_2$ = 40%/40%/20%, by volume, was included to match the fuel composition of a turbulent 'target' sooting flame (termed the "Adelaide simple jet flame" in the International Sooting Flames Workshop). Planar laser-induced incandescence (LII), time-resolved LII (TiRe-LII) and two-line atomic fluorescence (TLAF) thermometry (using atomic indium as tracer) were applied to measure f_v, d_p and T. Radial profiles of the flame temperature were also measured using a thermocouple positioned 3 mm above the burner lip, which is just upstream from the sooting region. It is found that dilution either with H_2 or N_2 causes a significant reduction in f_v and d_p, while the influence on the peak flame temperature is weaker. Flame structure (length and spatial profiles) is also influenced by the blending with H_2 and N_2. This database can serve as benchmark for model development due to its systematic nature and combination of measured scalars.

1. Introduction

There is on-going need for high-fidelity experimental data of sooting flames to advance understanding of the mechanisms that govern soot formation and oxidation, while also enabling reliable model validation. This holds true for both laminar and turbulent flames. More data sets are now available for steady laminar flames than for turbulent flames, which may be due to the challenges in conducting the measurements in turbulent flames. A number of data sets for steady laminar sooting flames are accessible via the International Sooting Flames Workshop website [1], while ethylene (C_2H_4) has most often been chosen as the fuel for previous investigations, which may be due to its high soot yields and relatively simple chemistry in soot formation. Parallel with laminar flames, turbulent sooting flames have also been studied to understand the complicated interactions between soot chemistry and turbulence, e.g. the piloted flames burning C_2H_4 [2,3] and natural gas (~81% methane and 14% N_2) [4], the lifted C_2H_4/air diffusion jet flame [5,6] and the attached jet flame using a mixture of $C_2H_4/H_2/N_2$ with a ratio of 40%/40%/20% by volume as fuel [7]. These investigations both in laminar and turbulent flames have provided valuable experimental data for soot modelling, however, one drawback of these previous investigations is that they have mostly been undertaken in isolation, which makes it difficult to link the results and complicates the interpretation of modelling results in turbulent flames.

Some high-quality measurements in laminar C_2H_4/air diffusion flames, as well as those diluted with N_2, are available in the literature [8–14]. However, highly turbulent flames lift off from the

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[*] Corresponding author at: School of Mechanical Engineering, The University of Adelaide, SA 5005, Australia. Fax: +61 883134367.
E-mail address: bassam.dally@adelaide.edu.au (B. Dally).

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burner when flame dimensions are suitable for investigation in most laboratories with the use of pure C_2H_4 the fuel [5,6], which introduces challenges to model validation. Although it is possible to stabilise the flame using a pilot [2–4], this also generates complexity for model validation. Another approach to achieve an attached turbulent flame is to employ a fuel blend, such as with the use of $C_2H_4/H_2/N_2$ [7]. However, despite some advantages in this approach, insufficient data are available from the literature with which to isolate the separate influences of blending into the fuel the reacting, but non-sooting H_2 from the inert N_2. To meet this need, a systematic investigation of the influences of H_2 and N_2 on laminar, sooting flames with different blends of $C_2H_4/H_2/N_2$ is required. This provides the main motivation for the present work.

The effect of H_2 addition on suppressing soot formation had been investigated many times previously, e.g., [15–18]. It is generally accepted that addition of H_2/dilution by H_2 can influence soot formation through three mechanisms: (1) direct chemical interactions, (2) a dilution effect and (3) a thermal effect due to the change of flame temperature [19,20]. However, the details of these mechanisms are still incompletely understood, with some inconsistency in previous investigations, so that more work is required. Gülder et al [20], showed that the addition of H_2 to axisymmetric co-flow C_2H_4/air diffusion flame can decrease soot yield through both the dilution and chemistry effects. However, H_2 is more effective than the inert gas helium in inhibiting soot formation, revealing the presence of the chemical effect. Moreover, the changes in the temperature fields are negligible when either H_2 or helium added into the fuel side, suggesting that the thermal effect is also negligible. These results were then confirmed in a subsequent numerical study [21], which identified a reduction in atomic hydrogen concentration and an increase in molecular H_2 concentration. The computational work [21] also revealed that hydrogen addition plays a role in reducing the active surface site density, which reduces the proportion of larger soot particles. The chemical inhibition effect can be summarized as decreasing the hydrogen atom concentration in regions of soot surface growth and increasing the concentration of molecular hydrogen in the lower regions of the flame. Being different from C_2H_4 flames, no influence of H_2 addition on soot formation was found in propane or butane flames apart from the dilution effect [20], revealing that the significance of the inhibiting effect of H_2 depends on the properties of the fuel. A different chemical effect was also found in a co-flow methane/air diffusion flame, where both numerical and experimental studies [22,23] showed that adding H_2 chemically enhances soot formation, while also introducing the dilution effect. The numerical work showed that H_2 addition increases the concentrations of propargyl, benzene and pyrene, which then promotes soot nucleation process. Noteworthy is that this increase was only assessed for methane/air flames, not for ethylene/air flames which are the focus of the present study. Pandey et al. experimentally studied the influence of H_2 addition in co-flow acetylene/air diffusion flame [19]. They found that both f_v and d_p decrease, while the flame temperature moderately increases (<100 K) and the fractal dimension of soot aggregates ($\sim$1.7–1.8) does not change. In addition to the chemical effect, in which atomic and molecular hydrogen influence soot formation through the hydrogen-abstraction/acetylene-addition (HACA) mechanism [24], the authors also proposed that soot oxidation is enhanced by increasing the concentration of OH radicals. Zhao et al. experimentally investigated the influence of adding/diluting H_2 and adding helium in C_2H_4/air diffusion flames using an optical diagnostic technique [25]. They found that replacing C_2H_4 with H_2 (which is termed H_2 dilution in the present work) is much more effective at reducing soot formation than adding helium. They also found that either adding H_2 or helium does not significantly affect d_p but does reduce f_v. Adding H_2 can moderately increase the flame temperature (<100 K), while adding helium has negligible influence on the temperature in the reaction region but increases it elsewhere. In counter-flow diffusion flames, Du et al. compared the effects of the additives H_2, He, Ar, N_2 and CO introduced into the fuel C_2H_4, C_3H_8 and C_4H_{10} [26]. They found that addition of H_2 increases flame temperature, while helium is the most effective gas for inhibiting soot formation.

Compared with H_2, the influence of N_2 blending on sooting laminar flames has been studied relatively less, for which the reason may be that N_2, as a non-reacting species, was simply regarded as a diluted gas that reduces soot yield and flame temperature. Following the work of Tesner et al. in the sixties [17], a number of early work reported soot concentration decreased due to N_2 dilution into the fuel, and attributed this soot reduction to the changes in flame temperature, i.e. via the thermal effect [10,27,28]. The dilution effect, or the change in species concentration, was not taken into account. While Kent and Wagner argued that the reduction in flame temperature is not sufficient to explain the amount of soot reduction [29], the thermal effect and the dilution effect were then separately investigated [30–33]. It was found that the dilution effect typically plays a major role in soot reduction when moderate N_2 was added into the fuel with a constant flow rate [30,31], also observing the changes in flame height and diameter. Boedeker and Dobbs found that with addition of N_2 into ethylene, the temperature of the co-flow diffusion flame does not change homogeneously, flame temperature decreasing in the lower half of the flame, while it increases in the upper half, meanwhile the luminous zone becomes short [34]. While these early studies provided high-quality experimental data, these results were generally limited to point-wise measurements, with limited spatial information. The recent most significant investigation may be that of Smooke et al. [1,11,12,35], who investigated a series of laminar co-flow diffusion flames both experimentally and numerically as a function of the fuel fraction (32%, 40%, 60% and 80% C_2H_4) mixed with inert N_2. The effect of four diluents (argon, helium, nitrogen, and carbon dioxide) on soot precursor formation and temperature in ethylene laminar diffusion flames has also been studied individually by Abhinavam et al. [36], but their work mainly focused on the effect of pressure on soot evolution. Another work is from Liu et al. [37], in which the effects of N_2 and carbon dioxide on soot formation were numerically and experimentally investigated at pressures between 5 and 20 atmospheric pressure.

Despite the aforementioned studies, two gaps remain to be filled to enable a detailed understanding of the separate chemical and dilution mechanisms in multi-component blends of $C_2H_4/H_2/N_2$ mixtures in turbulent jet flames. The first relates to the change in the exit velocity of the fuel mixture when H_2 was added into the fuel, resulting in changes to the exit strain rate of the flames, defined as (u/D), where u is the exit flow velocity and D is the diameter of fuel pipe. Decroix et al. showed that soot formation is sensitive to the initial strain rate in steady counter-flow diffusion flames [38]. Hence, it may be premature to assess the potential effects (i.e. chemical, thermal and dilution effects) of H_2 addition on soot formation without isolating the effect of the change in the exit flow velocity, and hence the exit strain rate. To the best of our knowledge, the influence of strain rate has not been isolated in 2-D diffusion sooting flames. Further investigations are therefore required to assess both flames with a constant exit flow rate, which are here termed “dilution flames”, and those at constant ethylene flow-rate.

The second gap is that the influence of H_2 addition (as well as other gaseous additives) has mostly been studied for only two-component mixtures. The influence of a second blended gas in a multi-component mixture, as in the mixture of $C_2H_4/H_2/N_2$, has not been investigated. Furthermore, the comparison of the different effects with identical methods is also not available, which is highly desirable to avoid lab-to-lab differences. The influence of