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Effect of fuel composition on jet flames in a heated and diluted oxidant stream

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Abstract

The role of hydrogen addition on the structure of the Moderate or Intense Low oxygen Dilution (MILD) combustion regime is examined using a combination of experimental techniques and laminar flame calculations. Laser diagnostic imaging is used to simultaneously reveal the in-situ distribution of the hydroxyl radical (OH), formaldehyde (H₂CO), and temperature using the Jet in Hot Coflow (JHC) burner. The fuels considered are natural gas, ethylene, and LPG (each diluted with hydrogen 1:1 by volume). Hydrogen addition to the primary fuel was found necessary to stabilise the flames. Further to the role of hydrogen in the stabilisation of the flames, hydrogen addition also leads to the reaction zone exhibiting similar structure for different primary fuel types. The independence of the reaction zone structure with hydrogen addition suggests that a wide variety of fuels may be usable for achieving MILD combustion.

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1 1 Introduction

The combustion of fuel in a heated and diluted oxidant stream results in a 2 unique combustion regime referred to as Moderate or Intense Low oxygen Di-3 lution (MILD) combustion [1]. The strong recirculation of exhaust gases back into the reaction zone results in a localised reduction in O_2 level, leading to 5 a distributed reaction zone. The result of MILD combustion is a reduction 6 in pollutant emissions (notably NO_x) and an increase in net radiation flux [2, 3]. Application of MILD combustion has been successfully applied in numerous applications, additionally incorporating the use of biomass fuel [4]. MILD combustion is typified by low (near unity) Damköhler numbers [5], and 10 has the potential for cost effective low NO_x gas turbine power generation [6]. 11

The depleted O_2 oxidant at elevated temperatures, necessary for MILD com-12 bustion, is typically realised by the recirculation of hot exhaust gases. Recir-13 culation may be achieved either internally or externally with regard to the 14 combustor. The complex interactions within such a system make it unsuit-15 able for a fundamental study of the reaction zone. Instead, in this study an 16 experimental burner is used to emulate MILD combustion under controlled 17 conditions. The Jet in Hot Coflow (JHC) burner [7] enables a range of com-18 bustion parameters to be varied independently, and decouples the flow from 19 the chemical kinetics. The JHC burner has been used for both experiments [7– 20 10] and modelling [11-19]. A similar burner has also been used for combined 21 experiments [20, 21] and modelling [22]. 22

²³ Despite much progress in the application of MILD combustion to practical
²⁴ systems, there remain unresolved issues on the stabilisation, auto-ignition,

and structure of the reaction zone near the jet exit under the hot and diluted 25 conditions [10, 23, 24]. In particular, the influence of fuel type on the funda-26 mental aspects of the combustion remain poorly understood. From a global 27 perspective, MILD combustion features have been shown to be insensitive to 28 fuel type, including liquid fuels [25]. Similarly, in a furnace environment it has 29 been observed that the combustion of light fuel oil is very similar to natural 30 gas, though differences were observed for heavy fuel oil and coal [26]. Further-31 more, MILD combustion is particularly well-suited to using low calorific value 32 (LCV) fuels [27]. Notably, the operation of a furnace using natural gas and bio-33 gas $(60\% \text{ CH}_4 / 40\% \text{ CO}_2)$ has shown similar performance and emissions [28], 34 as shown elsewhere [29]. MILD combustion has been demonstrated for both 35 LCV fuels and industrial waste in the presence of hydrogen [30], with coke 36 oven gas [31, 32], and in gas turbine combustors with LCV fuels [33]. There is 37 also a move toward using coal with MILD combustion [34, 35]. Nonetheless, 38 detailed fundamental understanding of MILD combustion of non-conventional 39 fuels has yet to be widely achieved [31]. While other studies have used both 40 methane and propane for MILD combustion [36], the present paper aims to 41 provide a dedicated detailed fundamental-level investigation on the effect of 42 fuel composition. 43

Hydrogen has been proposed as a "clean fuel" alternative for the future, in particular for use as a supplemental fuel additive [37, 38]. Whilst the use of pure hydrogen eliminates CO₂ emissions, it requires a redesign of existing burners [18]. Avoiding the need for purity, hydrogen may also be used for enrichment, which can be obtained using methods (such as gasification of biomass) that have both economical and environmental benefits [38]. The addition of hydrogen to other fuels is of particular interest to MILD combustion as it can

⁵¹ be well-controlled, improves the range of operating conditions and is also im⁵² portant for the incorporation of low calorific fuels [30]. Furthermore, although
⁵³ MILD combustion is effective at eliminating soot, the addition of hydrogen
⁵⁴ will further increase the ability to use fuels that have sooting tendencies [31].

Hydrogen is commonly used as a fuel enhancer to improve system reactiv-55 ity, such as to promote ignition of low calorific value fuels [30]. The role of 56 different hydrogen concentration with hydrocarbon (methane) fuel has been 57 studied numerically by Mardani *et al.* [14], and shown to increase the reaction 58 intensity. Hydrogen addition is known to improve laminar flame speed [39], 59 and increase the maximum strain rate before extinction [40]. These features 60 may enable operation over a wider range of conditions [30], but may require 61 the redesign of conventional burners [38]. The addition of hydrogen increases 62 the flame speed, such that flames are more likely to attach to the burner 63 nozzle [38, 41, 42], which may negatively impact the operation of MILD com-64 bustion burners which require significant mixing of the fuel prior to reaction 65 [31]. As expected, increased hydrogen concentration in the fuel also leads to 66 increased OH concentration in the reaction zone [18, 38]. As a consequence 67 of the reaction occurring closer to the exit, in conjunction with the higher 68 reaction rates, there is less opportunity for the mixing that is required to 69 achieve low- NO_x emissions [43]. Nonetheless, this effect will be dependent on 70 burner configuration (especially burners that rely on entrainment due to high 71 velocity). It has also been reported that NO_x emissions are not sensitive to 72 the hydrogen concentration under MILD conditions [44], though this observa-73 tion requires accurate understanding of NO formation via the NNH route [45]. 74 Further details on NO_x formation are detailed in the literature |46-51|. It is 75 important to note that relatively small quantities of hydrogen are required to 76

have significant changes on the combustion: further increasing the hydrogen
concentration does not lead to proportional changes in the behaviour, which
is attributed to the role of hydrogen in affecting the chain-branching [31], and
highlights the importance of detailed chemistry in modelling such combustion
[52]. The influential role of hydrogen addition has been observed to eliminate
the region of "no ignition" in the jet stirred flow reactor of Sabia *et al.* [30].

In this paper the influence of combustion chemistry on the flame behaviour 83 and reaction zone structure is examined by systematically increasing the fuel 84 complexity under MILD combustion conditions. Temperature, the hydroxyl 85 radical and formaldehyde are measured instantaneously and simultaneously 86 using planar laser imaging techniques to reveal details of the structure of the 87 reaction zone. The hydroxyl radical (OH) is used as a flame marker while the 88 formaldehyde (H_2CO) intermediate species is predominant at low tempera-89 tures typical of those found in MILD combustion. The product of [OH] and 90 $[H_2CO]$ has also been suggested as an indicator of the formyl (HCO) radical, 91 which is closely related to the heat release rate [53]. Three different fuels are 92 considered, namely; natural gas (predominately methane), ethylene, and LPG 93 (predominately propane). The difference in the chemical path for these fuel 94 mixtures provides a way of assessing the sensitivity of the MILD combustion 95 regime to fuel type (for gaseous hydrocarbon fuels). Each primary fuel is di-96 luted with hydrogen (H_2) in an equal volumetric ratio to reduce the levels 97 of soot and to stabilise the flame. The role of hydrogen addition is addition-98 ally investigated through laminar flame calculations. For each fuel type in 99 the jet, differences in the fluid properties necessitates a change in the veloc-100 ity to maintain the Reynolds number constant (at $Re_{jet}=10,000$) in order to 101 maintain similar turbulence levels. 102

103 2 Experimental Details

The MILD combustion burner used in this study is the jet in hot coflow 104 (JHC) burner used previously [8, 9], and shown in Figure 1. It consists of 105 a central insulated fuel jet (\emptyset 4.6mm) within an annular coflow (\emptyset 82mm) of 106 hot exhaust products from a premixed secondary burner mounted upstream 107 of the jet exit plane. The coflow O_2 level is either 3% or 9% (volumetric), with 108 an exit temperature of 1100K. Various hydrocarbon fuels are used in the jet; 109 natural gas (NG), ethylene (C_2H_4) and liquefied petroleum gas (LPG). Each 110 of these primary fuels is diluted with hydrogen (H_2) in an equal volumetric 111 ratio. The addition of H_2 is found to be necessary to stabilise the flames. 112 Hydrogen addition also lowers the level of soot, which is advantageous for 113 laser measurements. Addition of H_2 also has implications for the potential use 114 of hydrogen as a supplemental fuel additive. Noteworthy is that although the 115 fuel stream is 50% H_2 by volume, it is less than 11% on a mass basis, and 116 the heat release from the hydrogen is <25%. The jet Reynolds number for the 117 experimental data presented in this paper is 10,000. 118

Laser induced fluorescence (LIF) is used to image OH and H_2CO , and tem-119 perature is inferred from Rayleigh scattering measurements. The laser pulses 120 are fired sequentially to reduce interferences on the other systems, with the 121 entire sequence occurring in 300ns to ensure the flow field is effectively frozen 122 with respect to the fluid time scales. The in-plane resolution of all three ICCD 123 cameras is 160µm, after spatial matching. The laser sheet heights were all 124 ~ 12 mm, of which the central 8mm portion is presented herein. All images 125 are corrected for laser power and profile variations shot-to-shot based on the 126 signal from a laminar slot burner. Description of the experimental details is 127

described in-depth in a previous publication by the authors [8].

129 3 Laminar Flame Calculations

Laminar flame calculations have been performed to extend the study beyond 130 the available measurements, and shed more light on the structure of the reac-131 tion zone and molecular transport. The OPPDIF routine of the Chemkin pack-132 age is used to compute temperature and species concentration for opposed-133 flow diffusion flames. The opposed-flow laminar diffusion flame configuration 134 represents a one-dimensional flame, analogous to the traverse across the well-135 defined reaction zone from fuel to oxidant. By increasing the velocity of the 136 flow from two facing nozzles, the strain rate imposed on the flame front can 137 be varied. The strain rate quoted throughout most of this paper is the average 138 normal strain rate reported in the OPPDIF post-processor output. The use 139 of a laminar, one-dimensional, configuration provided by the OPPDIF mod-140 els is a well established methodology to enable the role of strain alone to be 141 de-coupled from the more complex turbulent interactions that are observed 142 experimentally [10]. Furthermore, it has previously been demonstrated that 143 one-dimensional laminar diffusion flame calculations give excellent agreement 144 with detailed single-point measurements in the JHC burner [7]. Other studies 145 on the fundamental aspects of MILD combustion also have used this configu-146 ration [54, 55]. 147

For all calculations the GRI-Mech 3.0 mechanism is used. Previous studies have shown the GRI mechanism to provide agreement with experimental measurements of the JHC burner used in this study under similar conditions [7, 12, 56]. Both thermal diffusion and multi-component diffusion models are ¹⁵² used for the calculations.

Since the coflow oxidant stream consists of combustion products (H₂O and CO₂), the standard definition of mixture fraction is not appropriately defined for calculations based on the mass fraction of H & C (hydrogen & carbon) atoms. A normalised mixture fraction, $\xi^* = (\xi - \xi_{oxi}) / (\xi_{fuel} - \xi_{oxi})$ is used instead, where ξ_{fuel} & ξ_{oxi} refer to the standard definition of mixture fraction at the fuel and oxidant stream boundaries, respectively.

159 4 Results

160 4.1 Visual Observations

Figure 2 shows photographs of the flames presented in this paper. The hori-161 zontal dashed lines at 35 & 125mm downstream of the jet exit plane indicate 162 the locations of the laser diagnostic measurements. Apparent from Figure 2 is 163 that the natural gas flames show significantly less soot than the ethylene and 164 LPG flames. While soot is seen at both coflow O_2 levels for the ethylene and 165 LPG flames, at the 3% O_2 level soot does not appear until around $\sim 200 \text{mm}$ 166 downstream, whereas for the 9% O₂ case soot appears much closer to the jet 167 exit (~100mm). For either fuel type, at the higher (9%) O_2 level, close to 168 the jet exit the flame luminosity is significantly greater as compared to the 169 lower (3%) O₂ case. The low luminosity of the 3% O₂ flames almost makes 170 them appear invisible for the first ~ 100 mm. While not clearly apparent from 171 the photographs, a faint reaction is indeed apparent in this region. Further 172 downstream, once the effects of the coflow are diminished by the entrainment 173 of surrounding air (viz. ~ 100 mm), soot does begin to appear for either coflow 174

 O_2 level. The presence of soot around the 125mm downstream location in the 9% O_2 flames could potentially interfere with laser diagnostic measurements, and so data is collected at this location for the 3% O_2 flames only.

178 4.2 Typical Features

Figure 3 presents typical image triplets of OH, H_2CO and temperature for 179 each flame condition (three fuel types, each at two coflow O_2 levels). The 180 measurements are centred at 35mm downstream of the jet exit plane. The 181 corresponding size of each image is 8mm in height and 35mm wide. The jet 182 centreline is marked by the vertical dashed line. These images are for a jet 183 Reynolds number of 10,000 and are typical of other Reynolds number flames 184 as well. Despite the flow being nominally turbulent, the majority of the images 185 show no sign of large-scale turbulent structure. 186

In each of the images presented in Figure 3 the OH appears as an unconvoluted layer which is quite uniform in intensity along the length of the sheet. For each fuel type, the OH concentration is considerably less at 3% O_2 as compared to the 9% O_2 case. The suppression of OH with a reduction in O_2 level is consistent with previous work (*e.g.* [8, 57]) and is directly related to the reduced temperature of the reaction zone. At either O_2 level the OH concentration does not significantly vary with the fuel type.

The temperature in the coflow is seen to be uniform. With the 9% O_2 coflow the temperature is seen to increase in the region corresponding to OH. For all of the 3% O_2 cases there is no obvious sign of a temperature increase across the reaction zone, although a reaction is clearly taking place as identified by the presence of OH. Similar observations regarding the very low, almost indistinguishable temperature rise across the reaction zone has been seen in a MILD combustion furnace [58]. The low measured temperatures are believed to be genuine, and not because of interferences or problems with the Rayleigh to temperature conversion process. Noteworthy is that the laser-based measurements of the jet and coflow temperatures agree well with those expected, and were also confirmed with thermocouple measurements.

The H₂CO concentration varies with both the O₂ level and even more dramatically with the type of fuel. Most notably, the 3% O₂ C₂H₄/H₂ flame has significantly higher H₂CO than any other flame. In all cases the H₂CO appears quite uniformly distributed and always exists on the fuel-rich side of the OH layer. The broad radial distribution of H₂CO is also seen in strained laminar flame calculations [9].

211 4.3 Radial Profiles

Figure 4 shows the mean and RMS radial profiles of OH, H_2CO , and temperature for both 3% and 9% O_2 for the various fuel types, and at an axial location 35mm above the jet exit plane. Each plot is generated only from the central 3mm strip of the images, and not from the entire sheet height, so as to avoid potentially over-corrected values towards the edge of the images where the low laser energy possibly makes sheet corrections less reliable.

The instantaneous images presented (Figure 3) suggest that each of the various fuel types have a very similar structure. This is also seen in the radial plots of Figure 4. At the 3% O₂ coflow the mean OH profiles seem quite coincident,

with each fuel having a similar peak mean value and similar spatial shape. 221 These observations are consistent with the similarity of global features ob-222 served in a MILD combustion reactor [25]. Nonetheless, in both the mean and 223 RMS, there is a slight shift of the OH profiles inward towards the centreline as 224 the fuel complexity is increased. This radial shift corresponds to a drop in jet 225 velocity required to maintain the jet Reynolds number. At the 9% O_2 coflow 226 the radial shift of the OH peak with the fuel type is more noticeable. Also 227 more noticeable for the 9% case is a variation in the mean OH peak, although 228 the changes are still relatively small. Worth noting is that despite an almost 229 three-fold difference in the jet exit velocity for the various fuels, because the 230 Reynolds number is constant in all cases, the OH RMS is comparable for either 231 O_2 level. 232

At both coflow O_2 levels, in the H_2CO profiles of Figure 4 a very significant 233 increase is noted for the C_2H_4/H_2 flame. The mean H_2CO is distributed widely 234 across the radial profiles, but does show evidence of a dip along the jet cen-235 treline. The broad radial distribution of H_2CO has already been noted in the 236 instantaneous images, and is consistent with strained laminar flame calcula-237 tions. The H_2CO levels in the C_2H_4/H_2 flame are significantly higher with the 238 $3\%~O_2$ coflow as compared to $9\%~O_2,$ but for the other fuels (natural gas/H_2 239 & LPG/H_2) the H_2CO is similar at either O_2 level. 240

As was noted in the instantaneous images, the temperature rise across the reaction zone in the 3% O_2 coflow is barely discernible. For the 3% O_2 LPG/H₂ flame the temperature rise is not resolved in the mean profile. The lack of temperature rise has already been discussed in the typical instantaneous images. It is also important to note that the fluctuations in the radial location cause the peak in the mean radial profile to be significantly lower than instantaneous temperature peak. The low temperature rise in the mean radial profiles is consistent with previous single-point data measured in the JHC burner under MILD conditions [7]. At the 9% O₂ coflow, again the LPG/H₂ flame shows the lowest reaction zone temperature. The trends relating to the location of the peak temperature follows the same trend as seen in the OH profiles, *viz.* the peak shifts towards the centreline as the fuel complexity increases.

253 4.4 Laminar Flame Calculations

254 4.4.1 3% O2 oxidant

The trends noted in the flame measurements can be compared to strained 255 laminar flame calculations. It should be noted that due to the turbulent na-256 ture of the experimental measurements, the fluctuations in the reaction zone 257 location will cause the mean OH and temperature measurements to be lower 258 than those from laminar flame calculations. Figure 5 shows selected species 259 concentrations in mixture fraction space for a strain rate of $\sim 100 \text{ s}^{-1}$. The 260 temperature and OH, H₂CO & HCO mole fractions were obtained from OP-261 PDIF calculations of the Chemkin package using GRI-Mech 3.0 mechanism. 262 Previous studies have shown that the reaction structure is relatively insensi-263 tive to strain under MILD conditions, and that a strain rate of this order gives 264 the best agreement with experimental measurements [7, 8]. A normalised mix-265 ture fraction (ξ^*) is used instead of the standard definition obtained from the 266 calculations because of the non-standard oxidant stream composition [9]. The 267 normalised mixture fraction ensures that ξ^* ranges from zero in the oxidant 268 stream to one in the fuel stream. To observe features more clearly, the mixture 269 fraction is only shown to 0.25, beyond which there are no features of interest. 270

Consistent with the measurements, the OH profiles in Figure 5 for the various 271 fuels virtually overlap, both in location and peak value. There are some minor 272 differences in the width of the OH profiles in Figure 5, but when normalised, 273 the FWHM (full-width half-maximum) width varies by only around $\sim 5\%$. 274 Similarly, the temperature profiles in Figure 5 across the reaction zone are 275 also very similar, and virtually overlap for the different fuel types. At higher 276 mixture fractions, the temperature for the CH_4/H_2 flame does roll-off a little 277 faster than the others, but the differences are relatively minor. The general 278 observations regarding OH and temperature confirm the experimental result 279 that the fuel type has only minor effects on the reaction zone structure. 280

While the OH and temperature do not seem to vary significantly with the type 281 of fuel, from Figure 5, H₂CO does show very different behaviour depending 282 on the fuel type. In all cases, H_2CO is found almost over the entire range of 283 mixture fraction, extending well beyond the presented range. This explains the 284 very wide radial distribution of H₂CO in the images and reinforces that PAH 285 or Raman interference is not responsible for the distribution seen in the mea-286 surements. It is therefore more appropriate to make comparisons away from 287 the stoichiometric conditions. It is only for the CH_4/H_2 flame that the H_2CO 288 peak is located near the stoichiometric mixture fraction. Both the C_2H_4/H_2 289 and C_3H_8/H_2 flames reach a local maximum around stoichiometry, but fur-290 ther toward the fuel rich side the H_2CO concentration dips slightly and then 291 increases again. Analysis of the H_2CO rate of production indicates that the 292 $C_2H_3+O_2 \rightleftharpoons HCO+H_2CO$ reaction is primarily responsible for the presence 293 of of H_2CO in the fuel rich region. In this region, the concentration of C_2H_3 is 294 higher in the C_2H_4/H_2 and C_3H_8/H_2 flames, leading to the higher concentra-295 tion of H_2CO . The localised dip in H_2CO between stoichiometry and the fuel 296

rich region is a result of the increased H_2CO consumption via the $H+H_2CO$ $\Rightarrow HCO+H_2$ reaction.

Also plotted in Figure 5 is the HCO mole fraction. As expected, HCO lies in between the OH and H_2CO profiles. The HCO profile does change somewhat with the fuel type. For CH_4/H_2 the HCO is narrower but has a higher peak, whereas C_2H_4/H_2 and C_3H_8/H_2 are more spread out, but only slightly more so.

Noteworthy is that the peak temperature measured is lower than the laminar flame calculations. This is not fully understood, though the effect of radiation which is not accounted for in the calculations may be a contributing factor, along with some uncertainty in resolving the Rayleigh cross-section across the reaction zone. Significantly, the experimental results correctly capture the coflow and jet temperatures.

310 4.4.2 9% O2 oxidant

For the 9% O_2 coflow case, Figure 6 shows the same profiles as Figure 5. The temperature, OH and HCO all seem to have a similar response to the fuel type as already noted for the 3% O_2 case. Consistent with the measurements, the location of the OH peak again does not seem to be highly dependent on the fuel type, and the peak concentration is also relatively constant. At 9% O_2 the differences between the different fuels are somewhat more pronounced, but the general trends and observations are comparable.

At 9% O_2 the H₂CO distribution is again seen to be wide, extending well into the fuel rich side. Unlike at 3% O_2 , at 9% O_2 the H₂CO profiles do not

change quite as significantly with fuel type, although the differences are still 320 noticeable. The basic profile for each of the fuels is somewhat consistent, with 321 a peak just to the rich side of stoichiometry and a long tail extending further 322 to the fuel rich side. Noteworthy is that unlike the 3% O₂ oxidant, the H₂CO 323 does not increase in the fuel rich region for the 9% O₂ case. This difference is 324 a result of the reduced O_2 concentration in this region at 9% O_2 , as compared 325 with 3% O₂, inhibiting H₂CO formation via the $C_2H_3+O_2 \rightleftharpoons HCO+H_2CO$ 326 reaction. The lower O_2 on the fuel rich side of the reaction zone at higher O_2 327 oxidant stream concentration has been discussed previously [10]. 328

The stoichiometric mixture fraction, as indicated by the HCO peak, is seen to shift between the different fuels more for 9% O_2 than for 3% O_2 . This shift in stoichiometry becomes more noticeable as the stoichiometric mixture fraction becomes larger, but is still relatively small.

333 4.4.3 21% O₂ oxidant

To compare the similarity of the reaction zone structure for the different fuels 334 under the hot and diluted O₂ conditions, strained laminar flame calculation 335 results are shown for standard air $(21\% O_2, 300K)$ in Figure 7. Apparent is 336 that unlike for the MILD combustion conditions, the fuel type does lead to 337 clear differences in the species profiles. While the OH profiles remain similar, in 338 comparison to the 3% and 9% O₂, 1100K temperature oxidant stream case, the 339 OH profiles are no longer coincident. This suggests that the MILD combustion 340 conditions are fundamentally responsible for bringing about the similarity of 341 the temperature and OH profiles for the different fuels. 342

343 4.4.4 Effect of hydrogen addition

The comparison between the three different gaseous hydrocarbon fuels pre-344 sented thus far has revealed that the fuel type does not have a significant 345 effect on the reaction zone structure under MILD combustion conditions. It 346 is important to highlight that each of the primary fuels were diluted with 347 hydrogen (H_2) in equal volumetric parts. The addition of H_2 was necessary 348 to increase the flame stability to prevent blow-off. Hydrogen also shifts the 349 stoichiometric mixture fraction to the lean side, towards the edge of the shear 350 layer. The H_2 added to the fuel stream seems to influence the kinetics such 351 that the different hydrocarbon fuels show similar characteristics. 352

It was found from experimentation that when H_2 was not added to the jet the 353 flames would typically blow-off. Only the C_2H_4 flame could be sustained, and 354 even then, it appeared lifted. The importance of H_2 addition to the fuel is also 355 reflected in laminar flame calculations. For the experimental 3% O₂ oxidant 356 stream conditions, calculations show no temperature rise across the reaction 357 zone for the pure fuels without H_2 in the fuel stream, except at a very low 358 strain rate. These trends in the calculation for the undiluted fuel types are 359 in agreement with those noted during the experiments with the JHC burner. 360 These results also highlight the importance of H_2 to ignition, and thus the 361 importance of hydrogen-enrichment in the context of practical applications. 362

To examine the structural differences without H_2 addition to the fuel, Figure 8 shows selected species concentrations found from strained laminar flame calculations. The conditions for Figure 8 are identical as for the previously presented Figure 6, but without H_2 added to the fuel. As outlined in the preceding paragraph, comparisons can only be made at the 9% O_2 case, as at the $_{368}$ 3% O₂ conditions a reaction could not be sustained.

Comparison of Figure 8 (no H_2 addition) to Figure 6 (with H_2 addition) reveals 369 major differences in the selected species concentrations. Unlike with H₂ diluted 370 fuels, Figure 8 shows that the OH profiles are dependent on the fuel type. 371 For C_2H_4 , the peak OH concentration is 30% higher, and the FWHM (full-372 width half-maximum, in the normalised mixture fraction space) is 40% wider, 373 than for CH_4 fuel. Significant differences for the various fuel types in the 374 H_2CO and HCO profiles are also far more pronounced than seen with H_2 375 addition. Comparison of the peak concentration in the reaction zone of H₂CO 376 and HCO reveals that both increase by approximately a factor of 3.5 without 377 H_2 addition. 378

The role of H_2 leading to a similar reaction zone structure is important for the use of low calorific value fuels. These results suggest that H_2 not only improves reactivity but also changes the structure of the reaction zone.

382 5 Discussion

From the instantaneous images and the radial profiles that have been presented there does not seem to be any significant effect of the fuel type on the structure of the reaction zone. Similar trends are also observed at other measurement locations. The trends obtained from methane and propane fuels have previously been noted as being similar in a heated and diluted oxidant stream in a spectral emission study [59], and are consistent with the similar global behaviour reported previously [25, 26, 28, 29].

 $_{390}$ Of the measured scalars, the H₂CO number density changes the most between

the different fuel types considered. Nonetheless, the basic behaviour of the H_2CO is essentially constant between the different fuel cases. Changes in the flow structure are also apparent for the different fuel types. These differences arise because of the different jet velocity for each fuel type (required to maintain constant Reynolds number). Despite these minor differences, the fuel type does not lead to any major changes in the overall flame characteristics at the measurement locations under MILD conditions.

The photographs of the flames (Figure 2) provide supplemental evidence supporting the similarity of the different fuel types when in the confines of the hot and diluted coflow. Further downstream, after the effects of the coflow have diminished, each of the flames visually appear significantly different. This is suggestive of the importance of the coflow in establishment of the unique conditions which lead to the similarity between the different fuels.

Table 1 shows the average of the peak OH values in each of the images for a 404 particular flame. Also included is the standard deviation (as a percentage) of 405 the values. In determining the peak value in each of the images, only the central 406 3mm portion of the image is included to avoid over-corrected values towards 407 the edges of the images where the low laser power makes sheet corrections 408 less reliable. Table 1 reiterates the similarity of the OH levels for the different 409 fuel compositions. At either O_2 level the mean peak OH number density is 410 very similar for each fuel. Consistent with the radial profiles, the similarity is 411 especially noted for the 3% O₂ case. 412

At the 35mm location, where the oxidant composition is well defined and not yet affected by surrounding air entrainment, comparisons of the trends can be made to the strained laminar flame calculations. Table 2 presents the peak temperature, and the maximum number density of OH and H_2CO for each fuel type and at both O_2 levels obtained from the OPPDIF calculations.

Comparing Tables 1 & 2 it is seen that calculations support the measurements, 418 in that the OH concentration does not change significantly with the fuel com-419 position. The experimental peak OH value for the $3\% O_2$ flames is higher than 420 from the calculations, whereas at 9% O₂ the calculated value is higher than 421 that found from the experiment. The typical differences between the measured 422 value and calculations is approximately 25% for 3% O_2 and ~40% for 9% O_2 . 423 Nevertheless, there is good similarity of the trends, and also a similar order of 424 magnitude between the experiments and calculations. 425

Table 2 shows that the H_2CO in the C_2H_4/H_2 flame behaves the opposite 426 to the other two fuels. For C_2H_4/H_2 the H_2CO significantly increases at the 427 lower O_2 case. In contrast, the effect of O_2 on H_2CO is comparatively minor 428 for the other fuels. For C_3H_8/H_2 there is little difference between the two O_2 429 levels. In the CH_4/H_2 flame the trend is reversed and H_2CO slightly increases 430 with O_2 level. The different behaviour of the H_2CO in the C_2H_4/H_2 flames 431 to the other fuels was also apparent in the experimental data presented. Of 432 particular note in the C_2H_4/H_2 flames was that the H_2CO levels in the 3% 433 O_2 flames was much higher than the other cases, which is consistent with the 434 laminar flame calculations (Table 2). At 9% O_2 , Table 2 indicates that the 435 C_2H_4/H_2 flame should have lower H_2CO than the other flames, which is not 436 seen in the experimental data. Nevertheless, in general, the trends of H_2CO 437 largely follow those predicted by the flame calculations shown in Table 2. 438

It is important to highlight that each of the primary fuels are diluted with hydrogen (H_2) in equal volumetric parts. The addition of H_2 is necessary to increase the flame stability to prevent blow-off. The H_2 added to the fuel stream seems to influence the kinetics such that the different hydrocarbon fuels show similar characteristics.

444 6 Conclusions

The comparison between the three different gaseous hydrocarbon fuels in this 445 paper reveals that the fuel type does not have a significant effect on the re-446 action zone structure under MILD combustion conditions, when mixed with 447 hydrogen. The primary fuels considered were natural gas, ethylene, and LPG 448 (each diluted with H_2 1:1 by volume). Both experimental measurements and 449 strained laminar flame calculations indicate that the reaction zone structure 450 is very similar for the different fuels considered when hydrogen is added to 451 the fuel stream. Hydrogen was found necessary for the experimental flames to 452 stabilise. The OH concentration results were seen to be quite constant. Only 453 minor changes of the OH spatial distribution were noted, and attributable to 454 differences in fuel velocity required to maintain constant Reynolds number. 455 The only significant changes with the fuel type were noted in the H_2CO lev-456 els, most notably with the C_2H_4/H_2 flame. The trends in the measured H_2CO 457 levels were seen to be consistent with flame calculations. The similarity of the 458 combustion characteristics for the various gaseous hydrocarbon fuels consid-459 ered suggests that MILD combustion should be readily adapted for different 460 fuel types. The insensitivity to fuel type is potentially a significant advan-461 tage for the implementation and application of MILD combustion to practical 462 systems. 463

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645

Fuel	Peak n _{OH} $[\sigma]$					
compos-	$(\times 10^{16} {\rm cm}^{-3})$					
ition	$3\% O_2$		$9\% O_2$			
$\rm NG/H_2$	0.56	[11%]	1.28	[11%]		
C_2H_4/H_2	0.56	[10%]	1.53	[30%]		
LPG $/H_2$	0.54	[9%]	1.33	[8%]		

Table 1

Mean and standard deviation (σ , in brackets) of peak OH number density measurements. Central 3mm strip of images used. Axial location 35mm above jet exit.

Fuel	Peak temp.		Peak n _{OH}		Peak n_{H_2CO}	
compos-	(K)		$(10^{16} \mathrm{cm}^{-3})$		$(10^{14} {\rm cm}^{-3})$	
ition	3%	9%	3%	9%	3%	9%
CH_4/H_2	1400	1876	0.40	2.04	1.15	1.93
C_2H_4/H_2	1403	1856	0.46	2.11	2.59	0.87
C_3H_8/H_2	1384	1807	0.43	1.97	1.60	1.45

Table 2 $\,$

Peak temperature and OH & H₂CO number density (molecules/cm³) from strained laminar flame calculations for 3% and 9% O₂ oxidant stream conditions ($a\approx 200 \text{ s}^{-1}$).

List of Figure Captions

Figure 1: Cross-sectional diagram of jet in hot coflow (JHC) burner.

Figure 2: Photographs of natural gas (NG), ethylene (C₂H₄) & LPG flames, each diluted with hydrogen (1:1 vol/vol) at two coflow O₂ levels. Jet Reynolds number of 10,000. Note the different exposure times (all other camera parameters held constant). Horizontal lines indicate measurement locations (35mm & 125mm downstream of jet exit plane). Photograph height: 300mm.

- Figure 3: Selection of instantaneous OH, H₂CO and temperature image triplets of natural gas/H₂, C₂H₄/H₂ and LPG/H₂ flames showing typical features. OH shown in number density (molecules/cm³), H₂CO in arbitrary units, temperature in Kelvin. Re_{jet} =10,000. Each image 8×35mm. Jet centreline marked with dashed line. Axial location 35mm above jet exit.
- Figure 4: Mean and RMS radial profiles of OH, H₂CO and temperature for natural gas/H₂, C₂H₄/H₂ and LPG/H₂ flames. OH shown in number density (molecules/cm³), H₂CO in arbitrary units, temperature in Kelvin. *Re_{jet}=10,000*. Central 3mm strip of images used. Axial location 35mm above jet exit.
- Figure 5: Temperature and species mole fractions from strained laminar flame calculations in (normalised) mixture fraction space for 3% O₂ coflow composition ($a \approx 100 \text{s}^{-1}$).
- Figure 6: Temperature and species mole fractions from strained laminar flame calculations in (normalised) mixture fraction space for 9% O₂ coflow composition ($a \approx 100 \text{s}^{-1}$).
- Figure 7: Temperature and species mole fractions from strained laminar flame calculations in (normalised) mixture fraction space for $(21\% O_2, 300K)$ air

 $(a \approx 100 \mathrm{s}^{-1}).$

Figure 8: Temperature and species mole fractions from strained laminar flame calculations in (normalised) mixture fraction space for 9% O₂ coflow composition ($a \approx 100 \text{s}^{-1}$).



Fig. 1. Cross-sectional diagram of jet in hot coflow (JHC) burner.



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Fig. 6. Temperature and species mole fractions from strained laminar flame calculations in (normalised) mixture fraction space for 9% O_2 coflow composition $(a \approx 100 \text{s}^{-1})$.



Fig. 7. Temperature and species mole fractions from strained laminar flame calculations in (normalised) mixture fraction space for (21% O₂, 300K) air ($a \approx 100 \text{s}^{-1}$).



Fig. 8. Temperature and species mole fractions from strained laminar flame calculations in (normalised) mixture fraction space for 9% O_2 coflow composition $(a \approx 100 \text{ s}^{-1})$.