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*Full-length article:*

## **Reaction zone weakening effects under hot and diluted oxidant stream conditions**

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### **Abstract**

This paper reports computational results, to complement experimental observations, on the turbulence–chemistry interaction of nonpremixed jet flames issuing into a heated and highly diluted oxidant stream. It is found experimentally that large-scale vortices and flame stretch can lead to spatial thinning and a decrease in OH concentration. This reduction in OH is described as a weakening of the reaction zone. Accompanying reaction zone weakening is also an increase in H<sub>2</sub>CO levels. The reduction in reaction rates is most noticeable at low oxidant stream O<sub>2</sub> levels. The heated and low oxygen oxidant conditions typify those of Moderate or Intense Low oxygen Dilution (MILD) combustion. The computational results indicate that the effects of the low oxygen levels of MILD combustion leads to both a reduction in reaction rates and an increase in transport of O<sub>2</sub> across the reaction zone. The relationship between the reaction rate and level of O<sub>2</sub> permeation suggests that a form of partial premixing can occur under MILD combustion conditions. This partial premixing leads to the formation of flame intermediates which contribute to the stabilisation of the flames. The permeation effects are most pronounced at high strain rates, which are commonly encountered in practical MILD combustors.

*Key words:* Reaction zone weakening, MILD combustion, JHC burner

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# 1 Introduction

Moderate or Intense Low oxygen Dilution (MILD) combustion is a particular combustion regime which involves the strong recirculation of exhaust gases back into the reaction zone (Cavaliere and de Joannon, 2004, Choi and Katuski, 2001, Wüning and Wüning, 1997). The subsequent reduction in localised  $O_2$  concentration leads to a distributed reaction zone, reducing pollutant emissions (notably  $NO_x$ ) and an increase in nett radiation flux (Cavaliere and de Joannon, 2004, Weber et al., 2000). Application of MILD combustion has been successfully applied in numerous applications, additionally incorporating the use of biomass fuel (HiTACG, 2008). Despite much progress in the application of MILD combustion to practical systems, there remain unresolved issues on the fundamental stabilisation, auto-ignition, and structure of the reaction zone near the jet exit under the hot and diluted conditions (de Joannon et al., 2005, Maruta et al., 2000).

To achieve the very high recirculation rates required for MILD combustion high velocity jets are often used (Cavaliere and de Joannon, 2004). The high strain rate conditions lead to a reduction in turbulence time-scales, coupled with larger chemical time-scales as a result of the low oxygen conditions, these flames are associated with low Damköhler numbers (Katsuki and Hasegawa, 1998). As an extension to this, in a furnace environment MILD combustion has been likened to a well-stirred reactor (WSR) (Plessing et al., 1998, Weber et al., 1999). Based on the concept of attempting to infer MILD combustion from a well-stirred reactor, de Joannon et al. (2000) has attempted to model a WSR with MILD combustion conditions. However as identified in that work the authors acknowledge that a well-stirred reactor is unfeasible for a practical combustor. Furthermore, the interaction between turbulence and chemistry in describing the nature of MILD combustion is expected to

26 play a significant role (Katsuki and Hasegawa, 1998). The combustion is controlled  
27 by both the kinetics and the mixing (Milani and Saponaro, 2001), and so the anal-  
28 ogy to a WSR may not always be applicable to practical systems. The importance  
29 of both chemical and turbulent timescales has been shown by Galletti et al. (2007)  
30 who reported Damköhler numbers near unity in MILD conditions.

31 To advance the fundamental understanding of practical MILD combustion furnaces  
32 beyond WSR theory, there have been efforts to study MILD combustion furnaces  
33 on an experimental level (e.g. (Plessing et al., 1998, Szegö et al., 2008)). Through a  
34 series of subsequent studies (Coelho and Peters, 2001, Dally et al., 2004, Özdemir  
35 and Peters, 2001) using the furnace of Plessing et al. (1998), it has been reported  
36 that computational studies qualitatively match the experimental findings, except  
37 in the flow-field near the jet and the NO formation. A similar observation was  
38 made between the numerical work of Mancini et al. (2002) for the furnace of  
39 Weber et al. (2000). The comparisons between the numerical and experimental  
40 results from these furnaces highlights that there are aspects of the stabilisation  
41 and turbulence–chemistry interaction under MILD combustion conditions which  
42 remain unresolved.

43 To bridge the knowledge gap between the overly simplified WSR theory, and the  
44 complex interactions within a furnace environment (even on an experimental level),  
45 Dally et al. (2002a,b) reported on the structure of turbulent nonpremixed jet flames  
46 issuing into a heated and diluted coflow. This burner configuration, referred to as  
47 a jet in hot coflow (JHC) burner, emulates MILD combustion under simplified and  
48 well-controlled conditions. These authors used single-point Raman-Rayleigh-LIF  
49 diagnostic techniques to simultaneously measure temperature, major and minor  
50 species at different locations in these flames. It was found that major changes in the  
51 flame structure occur when reducing the oxygen concentration and that, at higher

52 jet Reynolds number and low oxygen concentration, oxygen leakage from the sur-  
53 roundings is related to local extinction of the flame. Medwell et al. (2007) extended  
54 that work by simultaneous imaging of the hydroxyl radical (OH), formaldehyde  
55 ( $\text{H}_2\text{CO}$ ) and temperature in the same burner, finding evidence of partial premixing  
56 in these flames, and localised extinction in the presence of surrounding air.

57 It has been identified that the flame stabilisation mechanism appears fundamen-  
58 tally different under MILD conditions. Different stabilisation characteristics have  
59 been noted in a MILD combustion furnace (Szegö et al., 2007). Under the hot  
60 and diluted coflow conditions, in a JHC burner, pre-ignition reactions have been  
61 identified upstream of what appear to be lifted flames (Medwell et al., 2008). The  
62 presence of flame intermediates (*viz.* OH and  $\text{H}_2\text{CO}$ ) in the apparently lifted re-  
63 gion of these flames lead to the term *transitional* flames being adopted to describe  
64 this phenomenon. In both transitional and attached flames,  $\text{H}_2\text{CO}$  has been detected  
65 along the jet centreline (at  $x/D=7.6$ ), suggesting diffusion of oxygen atoms well  
66 into the reaction zone. Further differences in the stabilisation of MILD combustion  
67 is evident by noting that the apparent liftoff height decreases with an increase in jet  
68 velocity (Medwell et al., 2008). This finding is consistent with the observation in  
69 a furnace environment that increasing the jet velocity helped stabilise the jet flame  
70 through enhanced mixing (Dally et al., 2004). Furthermore, Mancini et al. (2007)  
71 concluded a similar effect of “preconditioning” of the fuel whereby significant mix-  
72 ing occurs prior to the consumption of the fuel.

73 Using a JHC burner (Dally et al., 2002a,b, Medwell et al., 2007, 2008), this study  
74 aims to examine the structure of the reaction zone of a jet in a heated and diluted  
75 coflow. Temperature, the hydroxyl radical and formaldehyde are measured instan-  
76 taneously and simultaneously using planar laser imaging techniques. The hydroxyl  
77 radical (OH) is used as a flame marker while the formaldehyde ( $\text{H}_2\text{CO}$ ) interme-

78 diate species is predominant at low temperatures typical of those found in MILD  
79 combustion. The product of  $[\text{OH}]$  and  $[\text{H}_2\text{CO}]$  has also been suggested as an indi-  
80 cator of the formyl (HCO) radical, which is closely related to the heat release rate  
81 (Najm et al., 1998).

82 In this paper we report on the effect of turbulence–chemistry interaction on the reac-  
83 tion zone structure under hot and diluted oxidant stream conditions. Using the same  
84 experimental conditions presented in a companion paper (Medwell et al., 2007), a  
85 JHC burner is used to emulate MILD combustion. The fuel used is a mixture of  
86 natural gas & hydrogen, and issues into a coflow with a 3%  $\text{O}_2$  level. Laminar  
87 flame calculations are used to complement, and provide additional insight into, the  
88 reaction zone structure effects which were observed experimentally.

## 89 **2 Experimental Details**

90 The MILD combustion burner used in this study is the jet in hot coflow (JHC)  
91 burner used previously (Medwell et al., 2007, 2008), and shown in Figure 1. It con-  
92 sists of a central insulated fuel jet ( $\varnothing 4.6\text{mm}$ ) within an annular coflow ( $\varnothing 82\text{mm}$ )  
93 of hot exhaust products from a premixed secondary burner mounted upstream of  
94 the jet exit plane. The coflow  $\text{O}_2$  level is 3% (volumetric), with an exit temperature  
95 of 1100K. The fuel used in the jet is natural gas (92%  $\text{CH}_4$ ) diluted with hydrogen  
96 ( $\text{H}_2$ ) in an equal volumetric ratio. The addition of hydrogen reduces the levels of  
97 soot (advantageous for laser techniques) and improves flame stability. Addition of  
98  $\text{H}_2$  also has implications for the potential use of hydrogen as a supplemental fuel  
99 additive. The jet Reynolds number for the experimental data presented in this paper  
100 is 15,000.

101 Laser induced fluorescence (LIF) is used to image OH and H<sub>2</sub>CO, and tempera-  
102 ture is inferred from Rayleigh scattering measurements. The laser pulses are fired  
103 sequentially to reduce interferences on the other systems, with the entire sequence  
104 occurring in 300ns to ensure the flow field is effectively frozen with respect to  
105 the fluid time scales. The in-plane resolution of all three ICCD cameras is 160μm,  
106 after spatial matching. The laser sheet heights were all ~12mm, of which the cen-  
107 tral 8mm portion is presented herein. All images are corrected for laser power and  
108 profile variations shot-to-shot based on the signal from a laminar slot burner. De-  
109 scription of the experimental details is described in-depth in a previous publication  
110 by the authors (Medwell et al., 2007).

### 111 **3 Laminar Flame Calculations**

112 Laminar flame calculations have been performed to extend the study beyond the  
113 available measurements, and shed more light on the structure of the reaction zone  
114 and molecular transport. The OPPDIF routine of the Chemkin package is used to  
115 compute temperature and species concentration for opposed-flow diffusion flames.  
116 For all calculations the GRI-Mech 3.0 mechanism is used. Previous studies have  
117 shown the GRI mechanism to provide agreement with experimental measurements  
118 of the JHC burner used in this study under similar conditions (Christo and Dally,  
119 2005, Dally et al., 2002a, Kim et al., 2005).

120 The opposed-flow laminar diffusion flame configuration represents a one-  
121 dimensional flame, analogous to the traverse across the well-defined reaction zone  
122 from fuel to oxidant. By increasing the velocity of the flow from two facing noz-  
123 zles, the strain rate imposed on the flame front can be varied. The strain rate quoted  
124 throughout most of this paper is the average normal strain rate reported in the OP-

125 PDIF post-processor output. The use of a laminar, one-dimensional, configuration  
126 provided by the OPPDIF models is a well established methodology to enable the  
127 role of strain alone to be de-coupled from the more complex turbulent interactions  
128 that are observed experimentally.

129 Three different oxidant stream temperatures and compositions are used for the cal-  
130 culations, as listed in Table 1. The major species concentrations for the experimen-  
131 tal conditions used in the previous publication (Medwell et al., 2007) are found  
132 from equilibrium calculations based on the coflow inlet flowrates. The oxidant  
133 stream temperature is 1100K (to match the measurement from the JHC burner).  
134 For comparison, standard air conditions are included as well. The jet composition  
135 is an equal molar ratio of CH<sub>4</sub> & H<sub>2</sub> (i.e. the natural gas is assumed pure methane).

136 Since the coflow oxidant stream consists of combustion products (H<sub>2</sub>O and CO<sub>2</sub>),  
137 the standard definition of mixture fraction is not appropriately defined for calcu-  
138 lations based on the mass fraction of H & C (hydrogen & carbon) atoms. A nor-  
139 malised mixture fraction,  $\xi^* = (\xi - \xi_{oxi}) / (\xi_{fuel} - \xi_{oxi})$  is used instead, where  
140  $\xi_{fuel}$  &  $\xi_{oxi}$  refer to the standard definition of mixture fraction at the fuel and oxi-  
141 dant stream boundaries, respectively.

## 142 **4 Results and Discussion**

### 143 *4.1 Motivation*

144 Figure 2 shows a selection of instantaneous image triplets of OH, H<sub>2</sub>CO, and  
145 temperature from the JHC burner (repeated from Medwell et al. (2007)). The jet  
146 Reynolds number is 15,000 and the coflow O<sub>2</sub> concentration 3% (volumetric). The



147 images are centred at 35mm downstream of the jet exit plane. The corresponding  
148 size of each image is 8mm in height and 30mm wide. The jet centreline is marked  
149 by the vertical dashed line.

150 Typically, near the jet exit the images do not show evidence of large-scale vor-  
151 tices. The images suggest that the reaction zone appears unconvoluted, as shown  
152 in Figure 2a. Nonetheless, a proportion of the images do show signs of large-scale  
153 vortices, as indicated by convolution of the temperature and/or flame species. As  
154 the flow becomes more convoluted and stretched by vortices, the OH images can  
155 show a localised decrease in concentration and a spatial thinning. The phenomenon  
156 relating to reductions in the OH layer is described as a “weakening” of the flame  
157 front, and forms the basis of this paper. An example of a weakened reaction zone is  
158 seen in Figure 2b.

159 The interaction of the vortices with the reaction zone intuitively suggests that flame  
160 stretch leads to the observed reduction in OH concentration. Over a range of strain  
161 rates up to extinction, the laminar flame calculations presented in Figure 3 sug-  
162 gest that the peak OH number density does have a dependence on the strain rate.  
163 Similarly, the peak temperature is also dependent on the strain rate. The strain rate  
164 presented in Figures 3 & 4 is defined as the maximum absolute normal strain rate  
165 across the reaction zone. Apparent from Figures 3 & 4 is that the flames in the  
166 heated and diluted oxidant conditions can sustain a much higher strain rate be-  
167 fore extinction. It is also noted that the OH number density measured is of a similar  
168 magnitude to that predicted by the laminar flame calculations presented in Figure 3.

169 For comparison of the strain rate values to turbulent conditions, the scalar dissipa-  
170 tion ( $\chi$ ) is determined from the gradient of the mixture fraction ( $\xi$ );

$$\chi = 2\mathcal{D} (\nabla\xi \cdot \nabla\xi) \quad (1)$$

171 The diffusion coefficient ( $\mathcal{D}$ ) is determined from the mole-fraction weighted mean  
 172 of the mixture averaged diffusion of the fuel ( $k = \text{CH}_4$  and  $\text{H}_2$ ) components, defined  
 173 as;

$$\mathcal{D}_{km} = \frac{1 - Y_k}{\sum_{j \neq k}^K X_j / \mathcal{D}_{jk}} \quad (2)$$

174 The binary diffusion coefficients ( $\mathcal{D}_{jk}$ ) are determined based on polynomial fit with  
 175 temperature provided by the TRANSPORT library of CHEMKIN.

176 For the two heated oxidant streams the approximate maximum scalar dissipation  
 177 rate prior to extinction is  $\sim 1200 \text{ s}^{-1}$  for the 3%  $\text{O}_2$  and  $\sim 6000 \text{ s}^{-1}$  for the 9%  $\text{O}_2$   
 178 oxidant. At standard oxidant conditions the peak scalar dissipation of  $\sim 150 \text{ s}^{-1}$  was  
 179 estimated. These results confirm that MILD combustion can sustain much higher  
 180 strain/scalar dissipation than under conventional oxidant stream conditions.

181 Experimental measurements of  $\text{H}_2\text{CO}$  show an increase with Reynolds number  
 182 (Medwell et al., 2007). An increase in  $\text{H}_2\text{CO}$  with strain is also noted in the lam-  
 183 inar flame calculations presented in Figure 5.  $\text{H}_2\text{CO}$  has also been shown to in-  
 184 crease with the extent of partial premixing (Mc Enally and Pfefferle, 2000, Med-  
 185 well et al., 2007). To investigate the possibility of a connection between the increase  
 186 in  $\text{H}_2\text{CO}$  with strain and partial premixing, the  $\text{O}_2$  concentration at the location of  
 187 peak  $\text{H}_2\text{CO}$  is plotted against strain in Figure 6.

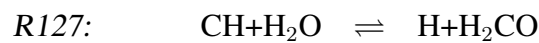
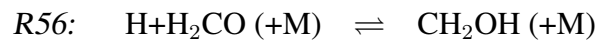
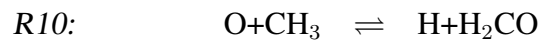
188 Figure 6 suggests a relationship between strain and partial premixing is plausible.  
 189 Since  $\text{H}_2\text{CO}$  is formed on the fuel-rich side of the reaction zone, the  $\text{O}_2$  concentra-

190 tion at the location of peak  $\text{H}_2\text{CO}$  is indicative of the amount of  $\text{O}_2$  which has been  
191 transported across the flame front. The molecular transport of  $\text{O}_2$  to the fuel-rich  
192 side is suggestive of a form of partial premixing, and is seen to increase with the  
193 strain rate. The presence of oxygen on the fuel-rich side of the reaction zone has  
194 been noted previously in MILD combustion conditions (Kim et al., 2005).

#### 195 4.2 Formaldehyde Production

196 The  $\text{H}_2\text{CO}$  production rates have been analysed from laminar flame calculations.  
197 For each oxidant stream composition, two strain rate conditions have been chosen  
198 to differentiate between low and high strain cases, *viz.*  $30 \text{ s}^{-1}$  and  $200 \text{ s}^{-1}$  (aver-  
199 aged normal strain rate, corresponding to maximum absolute normal strain rates of  
200  $\sim 50 \text{ s}^{-1}$  and  $\sim 500 \text{ s}^{-1}$ ). Previous studies have shown the validity of laminar flame  
201 calculations to model these flames when using moderate strain rates, of the order  
202 of  $30 \text{ s}^{-1}$  to  $200 \text{ s}^{-1}$  (Dally et al., 2002a).

203 Five main reactions have been found to be responsible for the majority of  $\text{H}_2\text{CO}$   
204 formation and consumption. The key reactions identified are (numbered in accor-  
205 dance with GRI);



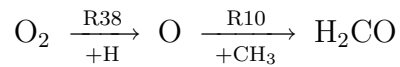
207 For the 3% O<sub>2</sub> oxidant stream case, Figures 7a & 7b show the production rate of  
208 H<sub>2</sub>CO plotted against mixture fraction for the five selected reactions at two differ-  
209 ent strain rates. Reactions R10 and R127 are seen to be the main contributors to  
210 H<sub>2</sub>CO production. H<sub>2</sub>CO production via R10 increases by a factor of ten between  
211 the two strain rates considered. In comparison, R127 increases by a factor of three.  
212 The increase of H<sub>2</sub>CO production as the strain rate is increased is consistent with  
213 Figure 5. Accompanying the increase in H<sub>2</sub>CO production is also an increase in  
214 the consumption rate. Reactions rates for R58 and R101 increase by a factor of  
215 five and three, respectively, between the two strain rate cases. Nevertheless, the  
216 large increase in the reaction rate of R10 with strain over-compensates for the dif-  
217 ferences in consumption, leading to the higher H<sub>2</sub>CO concentration at the higher  
218 strain condition.

219 The rate of H<sub>2</sub>CO production via reaction R10 for the two strain cases under con-  
220 sideration is shown in Figures 8a & 8b. Also shown in these figures are the species  
221 relevant to H<sub>2</sub>CO production via reaction R10 (*viz.* O and CH<sub>3</sub>). A vertical line  
222 is included as a reference point at the location of the peak nett H<sub>2</sub>CO production  
223 rate. At this mixture fraction, it is noted that the O concentration is very low for the  
224 low strain case, but is significantly (five fold) higher at the high strain case. This  
225 difference in O concentration between the two strain cases, coupled with a two fold  
226 increase in CH<sub>3</sub>, seems to explain the relative reaction rate of R10.

227 An increase in the concentration of O at the location of peak H<sub>2</sub>CO production  
228 seems to be one of the main contributors to the increase of H<sub>2</sub>CO with strain. In-  
229 vestigation of reaction rates reveals that the major production of O is from O<sub>2</sub>, via  
230 reaction R38:  $\text{H} + \text{O}_2 \rightleftharpoons \text{O} + \text{OH}$ . The production rate of O via reaction R38,  
231 and H & O<sub>2</sub> mole fractions, are also shown in Figures 8a & 8b. It is apparent that  
232 the production rate of O at the location of peak nett H<sub>2</sub>CO is ten fold greater at

233 the higher strain rate. This is predominately attributed to a five fold increase in O<sub>2</sub>  
234 concentration at the location of peak nett H<sub>2</sub>CO between the two strain cases.

235 It is acknowledged that there are many concurrent processes that occur as the strain  
236 rate is increased. Nevertheless, the most significant increase in H<sub>2</sub>CO with strain  
237 seems to occur via the following path;



238 The identified pathway relates the increase in H<sub>2</sub>CO with strain to an increase in  
239 O<sub>2</sub> concentration at the location of peak nett H<sub>2</sub>CO production. For these laminar  
240 nonpremixed calculations, any O<sub>2</sub> at the location of peak H<sub>2</sub>CO must have been  
241 transported from the oxidant stream. This transport of O<sub>2</sub> across the reaction zone  
242 is likened to partial premixing. The possibility of O<sub>2</sub> existing on the fuel rich side  
243 of the reaction zone is not unheard of; in conventional nonpremixed flames O<sub>2</sub> has  
244 been measured along the centreline towards the base of an attached flame (McE-  
245 nally and Pfefferle, 1999).

246 It is worth noting that both the experimental measurements (Figure 2) and the lam-  
247 inar flame calculations (Figure 8) show a broad H<sub>2</sub>CO distribution on the fuel-rich  
248 side of the reaction zone. Figure 7 indicates a cessation of significant H<sub>2</sub>CO produc-  
249 tion/consumption above  $\xi^* \gtrsim 0.015$ , yet high H<sub>2</sub>CO concentrations are seen much  
250 further into the fuel-rich side (Figure 8). This is strongly indicative of diffusion  
251 of H<sub>2</sub>CO towards the fuel-rich boundary, and is consistent with previous studies  
252 that have shown broad H<sub>2</sub>CO distribution is a result of diffusive and convective  
253 transport effects (Fotache et al., 1997a,b).

254 *4.3 Transport Effects*

255 As the strain rate increases, it has been demonstrated in this paper that the O<sub>2</sub>  
256 concentration on the fuel-rich side of the reaction zone increases. The O<sub>2</sub> level on  
257 the fuel-rich side of the reaction zone is controlled by the competition between  
258 transport from the oxidant stream and consumption. The net O<sub>2</sub> consumption rate  
259 (i.e. the opposite of production rate) for the two strain cases is shown in Figure 9.  
260 For reference purposes, key species mole fractions are also included on this plot. To  
261 aid in visualisation, since transport is a physical process, Figure 9 is plotted against  
262 distance, instead of mixture fraction. The distance origin is defined as the location  
263 of peak HCO; with the fuel-rich side denoted as negative, and the fuel-lean side  
264 positive. Although constant between the two plots, the range extends only over the  
265 region of interest, and not to the fuel and oxidiser boundaries. As expected, as the  
266 strain rate is increased it is apparent there is a spatial compression of the reaction  
267 zone. The width of the reaction zone (defined as the distance between the peak  
268 OH and H<sub>2</sub>CO, as markers of the fuel-lean and -rich peaks, respectively) reduces  
269 ~2-fold with the increase in strain.

270 From Figure 9, the peak O<sub>2</sub> consumption rate is seen to be ~5 times higher at  
271 200s<sup>-1</sup> compared to 30s<sup>-1</sup>. The increase in peak consumption rate is due in part to  
272 the spatial compression of the physical distances at the higher strain and therefore  
273 higher concentrations of the radicals involved. Nevertheless, the integrated O<sub>2</sub> con-  
274 sumption is ~3 times higher at the higher strain rate case.

275 Despite an increase in the consumption of O<sub>2</sub> with increased strain rate, it has  
276 been seen that more O<sub>2</sub> permeates the reaction zone. To compare the differences in  
277 transport, the velocity of O<sub>2</sub> for the two strain cases is also included in Figure 9.

278 The  $O_2$  velocity ( $v_{O_2}$ ) is defined as the combination of the bulk velocity ( $u$ ) and the  
279 diffusion velocity ( $V_{O_2}$ ) (Yamada et al., 2003). Using the  $O_2$  velocity, the mass flux  
280 transport of  $O_2$  ( $j_k = \rho_k Y_k v_k$ , with  $k = O_2$  (Reaction Design, 1999)) has also been  
281 added to Figure 9. It is apparent that the transport flux is significantly higher as the  
282 strain rate is increased. Coupled with the spatial compression of the reaction zone  
283 with increased strain rate, there is a significant increase in the transport of  $O_2$ .

284 It is therefore apparent that the increased  $O_2$  transport, and reduced spatial scales,  
285 associated with the increase in strain rate over-compensate for the increased reac-  
286 tion consumption rates. The result is that more  $O_2$  is capable of penetrating the re-  
287 action zone. This observation is consistent with the instantaneous images showing  
288 increases in  $H_2CO$  near the location of local weakening of the flame front (Fig-  
289 ure 2b).

#### 290 4.4 Comparison between oxidant stream conditions

291 In the conditions analogous to MILD combustion presented so far, an increase  
292 in transport of  $O_2$  across the reaction zone has been observed. To compare these  
293 heated and diluted conditions to more conventional flame conditions, Figure 10  
294 presents the same details as shown in Figure 9, but now for a 21%  $O_2$  & 300K  
295 oxidant stream. The permeation effects under consideration are primarily prevalent  
296 at high strain rates, and so data is presented only for the  $a \approx 200s^{-1}$  condition. The  
297 scaling for each of the species is the same between the plots, but the vertical axis  
298 range is different to accommodate the differences in the relative intensities. De-  
299 spite the seven-fold increase in oxidant  $O_2$  concentration, it is apparent that under  
300 the conventional oxidant stream conditions there is significantly lower  $O_2$  levels on  
301 the fuel-rich side of the reaction zone compared to MILD conditions. In order for

302 the higher O<sub>2</sub> level in the oxidant stream to be reflected on the fuel-rich side, the  
303 transport must also increase compared to the increase in consumption. The peak  
304 O<sub>2</sub> consumption rate is 7 times higher at the 21% O<sub>2</sub> level, whereas the O<sub>2</sub> mass  
305 flux only increases approximately 4-fold in comparison to the 3% O<sub>2</sub> & 1100K  
306 condition. In combination, the increase in consumption relative to transport coun-  
307 teracts the higher oxidant stream O<sub>2</sub> concentration in conventional conditions. In  
308 effect, the reaction zone becomes more permeable to O<sub>2</sub> under MILD combustion  
309 conditions. The increase in O<sub>2</sub> concentration on the fuel-rich side with a reduction  
310 in oxidant O<sub>2</sub> level is consistent with Figure 6.

311 To bridge the gap between the 3% O<sub>2</sub> & 1100K and 21% O<sub>2</sub> & 300K oxidant,  
312 Figure 11 shows the same details again, but for a 9% O<sub>2</sub> & 1100K oxidant stream.  
313 As expected (from Figure 6), it is seen in Figure 11 that the O<sub>2</sub> level on the fuel-rich  
314 side is significantly reduced at 9% O<sub>2</sub> as compared to the 3% O<sub>2</sub> oxidant stream.  
315 The three-fold difference in the O<sub>2</sub> level is accompanied by a three-fold increase in  
316 O<sub>2</sub> consumption rate. The increased O<sub>2</sub> level at 9% as compared to 3% leads only  
317 to a two-fold increase in O<sub>2</sub> flux. Again, the increase in consumption is greater than  
318 the increase in flux, such that the O<sub>2</sub> permeation to the fuel-rich side is less.

319 It is apparent that the lower the O<sub>2</sub> concentration in the oxidant stream the higher  
320 the degree of permeation of O<sub>2</sub> across the reaction zone. The lower reaction rates  
321 as a result of the low O<sub>2</sub> conditions associated with MILD combustion therefore  
322 lead to a form of partial premixing. These findings suggest that molecular transport  
323 and finite rate chemistry effects are essential in order to capture the stability and  
324 structure of these flames. The challenge remains, however, to capture the transition  
325 from a MILD regime to conventional flames (Christo and Dally, 2005).



#### 326 4.5 Stabilisation

327 In the low oxygen & high temperature oxidant stream conditions presented, it has  
328 been seen that transport of O<sub>2</sub> across the reaction zone is responsible for an increase  
329 in the formation of key flame intermediates (notably H<sub>2</sub>CO). As the strain rate is  
330 increased, these intermediates continue to be generated. The production of such  
331 flame intermediates is responsible for the stabilisation of the reaction.

332 For flames similar to those presented in the current paper, Medwell *et al.* Medwell  
333 *et al.* (2008) identified flame intermediates and pre-ignition reactions upstream of  
334 an apparent liftoff height. Due to the presence of intermediates in what is generally  
335 considered the liftoff region, the term “transitional” flames was adopted to describe  
336 these unique conditions.

337 As the velocity (hence strain rate) approaches lift-off, rather than the transitional  
338 flames becoming completely lifted, the formation of intermediates continues. It is  
339 proposed that in the “lift-off” region a pool of intermediates is produced. These  
340 precursors initiate the combustion reaction, and are believe to greatly enhance the  
341 stabilisation of the flame. The build-up of a pool of precursors (notably H<sub>2</sub>CO) in  
342 jet flames in a similar high temperature coflow has been suggested by Gordon *et al.*  
343 (2007).

## 344 5 Conclusion

345 MILD combustion is a promising combustion regime offering simultaneous reduc-  
346 tions in emissions and an increase in thermal efficiency. Laminar flame calculations  
347 in the low oxygen and high temperature conditions of MILD combustion have been

348 shown to lead to simultaneous reduction in reaction rates and increased transport  
349 across the reaction zone. Enhancing these effects, experimentally it is shown that  
350 interaction with large-scale vortices can lead to a spatial thinning of the reaction  
351 zone. The turbulence–chemistry interaction under the heated and highly diluted  
352 oxidant stream is referred to as reaction zone weakening. The effects of strain is  
353 particularly important for MILD combustion since practical combustors typically  
354 rely on high velocity jets to achieve the highly diluted conditions.

355 As the flame-front is stretched at the low oxygen (3% O<sub>2</sub>) and high temperature  
356 ( $T_{oxi}=1100\text{K}$ ) conditions, reaction zone weakening manifests itself in two ways;  
357 the strain effect leads to an increase in H<sub>2</sub>CO, and large-scale vortices lead to a  
358 reduction in OH concentration. The low O<sub>2</sub> conditions lead to a reduction in reac-  
359 tion rate, that is, a weaker reaction zone (as compared to higher O<sub>2</sub> oxidant stream  
360 conditions). When the strain rate is increased, the effects of weakening, combined  
361 with a spatial compression of the reaction zone, contribute to a greater degree of O<sub>2</sub>  
362 transport across the reaction zone. Therefore, in MILD combustion a form of partial  
363 premixing occurs. Large-scale turbulent mixing due to large-scale vortices can lead  
364 to a weakening of the flame front. This observation demonstrates the importance of  
365 turbulent mixing in the establishment of the MILD combustion regime.

366 Reaction zone weakening, which leads to a form of partial premixing, may con-  
367 tribute to the stabilisation of the MILD combustion reaction zone. Previous studies  
368 have shown that an increase in jet velocity helps stabilise flames under MILD con-  
369 ditions (Dally et al., 2004, Medwell et al., 2008). The partial premixing effects that  
370 have been identified provide an increase in the formation of flame intermediates,  
371 and thus an improvement in the stabilisation with jet velocity.

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376 **References**

- 377 A. Cavaliere and M. de Joannon. Mild combustion. *Progress in Energy and Com-*  
378 *bustion Science*, 30:329–366, 2004.
- 379 G.-M. Choi and M. Katuski. Advanced low NO<sub>x</sub> combustion using highly pre-  
380 heated air. *Energy Conversion and Management*, 42:639–652, 2001.
- 381 F. C. Christo and B. B. Dally. Modeling turbulent reacting jets issuing into a hot  
382 and diluted coflow. *Combustion and Flame*, 142:117–129, 2005.
- 383 P. J. Coelho and N. Peters. Numerical simulation of a mild combustion burner.  
384 *Combustion and Flame*, 124:503–518, 2001.
- 385 B. B. Dally, A. N. Karpetis, and R. S. Barlow. Structure of turbulent non-premixed  
386 jet flames in a diluted hot coflow. *Proceedings of the Combustion Institute*, vol-  
387 ume 29, pages 1147–1154, 2002a.
- 388 B. B. Dally, A. N. Karpetis, and R. S. Barlow. Structure of Jet Laminar Non-  
389 premixed Flames under Diluted Hot Coflow Conditions. *2002 Australian Sym-*  
390 *posium on Combustion and The Seventh Australian Flame Days*, Adelaide, Aus-  
391 tralia, 2002b.
- 392 B. B. Dally, E. Riesmeier, and N. Peters. Effect of fuel mixture on moderate and  
393 intense low oxygen dilution combustion. *Combustion and Flame*, 137:418–431,  
394 2004.
- 395 M. de Joannon, A. Cavaliere, T. Faravelli, E. Ranzi, P. Sabia, and A. Tregossi.  
396 Analysis of process parameters for steady operations in methane mild combus-  
397 tion technology. *Proceedings of the Combustion Institute*, volume 30, pages  
398 2605–2612, 2005.
- 399 M. de Joannon, A. Saponaro, and A. Cavaliere. Zero-dimensional analysis of di-  
400 luted oxidation of methane in rich conditions. *Proceedings of the Combustion*  
401 *Institute*, volume 28, pages 1639–1646, 2000.

- 402 C. G. Fotache, T. G. Kreutz, and C. K. Law. Ignition of Hydrogen-Enriched  
403 Methane by Heated Air. *Combustion and Flame*, 110:429–440, 1997a.
- 404 C. G. Fotache, T. G. Kreutz, and C. K. Law. Ignition of Counterflowing Methane  
405 versus Heated Air under Reduced and Elevated Pressures. *Combustion and  
406 Flame*, 108:442–470, 1997b.
- 407 C. Galletti, A. Parente, and L. Tognotti. Numerical and experimental investigation  
408 of a mild combustion burner. *Combustion and Flame*, 151:649–664, 2007.
- 409 R. L. Gordon, A. R. Masri, S. B. Pope, and G. M. Goldin. Transport budgets in  
410 turbulent lifted flames of methane autoigniting in a vitiated co-flow. *Combustion  
411 and Flame*, 151:495–511, 2007.
- 412 HiTACG. In *7<sup>th</sup> International Symposium of High Temperature Air Combustion  
413 and Gasification*, Phuket, Thailand, 13-16 January 2008.
- 414 M. Katsuki and T. Hasegawa. The science and technology of combustion in highly  
415 preheated air combustion. *Proceedings of the Combustion Institute*, volume 27,  
416 pages 3135–3146, 1998.
- 417 S. H. Kim, K. Y. Huh, and B. Dally. Conditional moment closure modeling of  
418 turbulent nonpremixed combustion in diluted hot coflow. *Proceedings of the  
419 Combustion Institute*, volume 30, pages 751–757, 2005.
- 420 M. Mancini, R. Weber, and U. Bollettini. Predicting NO<sub>x</sub> emissions of a burner  
421 operated in flameless oxidation mode. *Proceedings of the Combustion Institute*,  
422 volume 29, pages 1155–1163, 2002.
- 423 M. Mancini, P. Schwöppe, R. Weber, and S. Orsino. On mathematical modelling  
424 of flameless combustion. *Combustion and Flame*, 150:54–59, 2007.
- 425 K. Maruta, K. Muso, K. Takeda, and T. Niioka. Reaction zone structure in flameless  
426 combustion. *Proceedings of the Combustion Institute*, volume 28, pages 2117–  
427 2123, 2000.
- 428 C. S. Mc Enally and L. D. Pfefferle. Experimental study of nonfuel hydrocarbons

429 and soot in coflowing partially premixed ethylene/air flames. *Combustion and*  
430 *Flame*, 121:575–592, 2000.

431 C. S. McEnally and L. D. Pfefferle. Experimental Study of Nonfuel Hydrocarbon  
432 Concentrations in Coflowing Partially Premixed Methane/Air Flames. *Combustion and Flame*, 118:619–632, 1999.

434 P. R. Medwell, P. A. M. Kalt, and B. B. Dally. Simultaneous imaging of OH,  
435 formaldehyde, and temperature of turbulent nonpremixed jet flames in a heated  
436 and diluted coflow. *Combustion and Flame*, 148:48–61, 2007.

437 P. R. Medwell, P. A. M. Kalt, and B. B. Dally. Imaging of diluted turbulent ethylene  
438 flames stabilized on a Jet in Hot Coflow (JHC) burner. *Combustion and Flame*,  
439 152:100–113, 2008.

440 A. Milani and A Saponaro. Diluted combustion technologies. Technical Report  
441 Article Number 200101, IFRF Combustion Journal, February 2001.

442 H. N. Najm, P. H. Paul, C. J. Mueller, and P. S. Wyckoff. On the adequacy of certain  
443 experimental observables as measurements of flame burning rate. *Combustion*  
444 *and Flame*, 113:312–332, 1998.

445 I. B. Özdemir and N Peters. Characteristics of the reaction zone in a combustor  
446 operating at MILD combustion. *Experiments In Fluids*, 30:683–695, 2001.

447 T. Plessing, N. Peters, and J. G. Wüning. Laseroptical investigation of highly  
448 preheated combustion with strong exhaust gas recirculation. *Proceedings of the*  
449 *Combustion Institute*, volume 27, pages 3197–3204, 1998.

450 Reaction Design. TRANSPORT: Chemkin Collection Release 3.5. User Manual  
451 TRA-035-1, 1999.

452 G. G. Szegö, B. B. Dally, and G. J. Nathan. Stability limits of a parallel jet mild  
453 combustion burner system. *Proceedings of the Australian Combustion Symposium*, pages 62–65, 2007.

454  
455 G. G. Szegö, B. B. Dally, and G. J. Nathan. Scaling of NO<sub>x</sub> emissions from a

- 456 laboratory-scale mild combustion furnace. *Combustion and Flame*, 154:281–  
457 295, 2008.
- 458 R. Weber, A. L. Verlaan, S. Orsino, and N. Lallemand. On emerging furnace design  
459 methodology that provides substantial energy savings and drastic reductions in  
460 CO<sub>2</sub>, CO and NO<sub>x</sub> emissions. *Journal of the Institute of Energy*, 72:77–83, 1999.
- 461 R. Weber, S. Orsino, N. Lallemand, and A. Verlaan. Combustion of natural gas  
462 with high-temperature air and large quantities of flue gas. *Proceedings of the*  
463 *Combustion Institute*, volume 28, pages 1315–1321, 2000.
- 464 J. A. Wüning and J. G. Wüning. Flameless oxidation to reduce thermal NO-  
465 formation. *Progress in Energy Combustion and Science*, 23:81–94, 1997.
- 466 E. Yamada, M. Shinoda, H. Yamashita, and K. Kitagawa. Experimental and numer-  
467 ical analyses of magnetic effect on OH radical distribution in a hydrogen-oxygen  
468 diffusion flame. *Combustion and Flame*, 135:365–379, 2003.

Oxidant	3% O <sub>2</sub>	9% O <sub>2</sub>	21% O <sub>2</sub>
$T_{oxi}$	1100K	1100K	300K
O <sub>2</sub>	0.03	0.09	0.21
N <sub>2</sub>	0.84	0.78	0.79
H <sub>2</sub> O	0.10	0.10	0.00
CO <sub>2</sub>	0.03	0.03	0.00

Table 1  
Oxidant stream temperature and composition (molar basis) used for laminar flame calculations.



## List of Figure Captions

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

Figure 2: Selection of instantaneous OH, H<sub>2</sub>CO, and temperature image triplets (Medwell et al., 2007). Each image 8×30mm. Jet centreline marked with dashed line. Axial location 35mm above jet exit.

Figure 3: Calculated peak OH number density plotted versus maximum absolute strain rate found from laminar flame calculations at various oxidant stream O<sub>2</sub> levels.

Figure 4: Calculated peak temperature plotted versus maximum absolute strain rate found from laminar flame calculations at various oxidant stream O<sub>2</sub> levels.

Figure 5: Calculated peak H<sub>2</sub>CO number density plotted versus maximum absolute strain rate found from laminar flame calculations at various oxidant stream O<sub>2</sub> levels.

Figure 6: Calculated O<sub>2</sub> mole fraction (%) at location of peak H<sub>2</sub>CO concentration plotted versus maximum absolute strain rate found from laminar flame calculations at various oxidant stream O<sub>2</sub> levels.

Figure 7: H<sub>2</sub>CO production rate via major reactions, found from laminar flame calculations. 3% O<sub>2</sub>, 1100K oxidant stream for two different strain rates. (Note the different vertical axis scaling.)

Figure 8: Selected species and production rates found from laminar flame calculations. Production rate of H<sub>2</sub>CO is shown for reaction R10. Production rate of O is shown for reaction R38. 3% O<sub>2</sub>, 1100K oxidant stream for two different strain

rates. Vertical dashed line indicates location of peak  $\text{H}_2\text{CO}$  production rate. (Note the different vertical axis scaling.)

Figure 9: Selected species mole fractions ( $\text{OH}$ ,  $\text{H}_2\text{CO}$ ,  $\text{HCO}$ ,  $\text{O}_2$ ), nett  $\text{O}_2$  consumption rate ( $p_{\text{O}_2}$ ),  $\text{O}_2$  velocity ( $v_{\text{O}_2}$ ), and  $\text{O}_2$  mass flux ( $j_{\text{O}_2}$ ), found from laminar flame calculations. 3%  $\text{O}_2$ , 1100K oxidant stream for two different strain rates.

Figure 10: Selected species mole fractions ( $\text{OH}$ ,  $\text{H}_2\text{CO}$ ,  $\text{HCO}$ ,  $\text{O}_2$ ), nett  $\text{O}_2$  consumption rate ( $p_{\text{O}_2}$ ),  $\text{O}_2$  velocity ( $v_{\text{O}_2}$ ), and  $\text{O}_2$  mass flux ( $j_{\text{O}_2}$ ), found from laminar flame calculations. 21%  $\text{O}_2$ , 300K oxidant stream for strain rate,  $a \approx 200\text{s}^{-1}$ .

Figure 11: Selected species mole fractions ( $\text{OH}$ ,  $\text{H}_2\text{CO}$ ,  $\text{HCO}$ ,  $\text{O}_2$ ), nett  $\text{O}_2$  consumption rate ( $p_{\text{O}_2}$ ),  $\text{O}_2$  velocity ( $v_{\text{O}_2}$ ), and  $\text{O}_2$  mass flux ( $j_{\text{O}_2}$ ), found from laminar flame calculations. 9%  $\text{O}_2$ , 1100K oxidant stream for strain rate,  $a \approx 200\text{s}^{-1}$ .

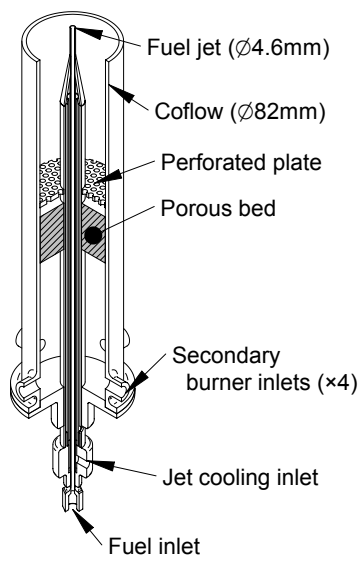


Fig. 1. Cross-sectional diagram of jet in hot coflow (JHC) burner.

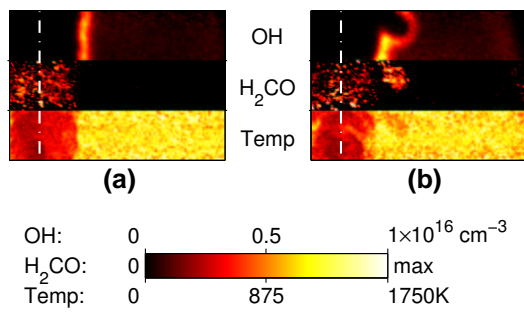


Fig. 2. Selection of instantaneous OH, H<sub>2</sub>CO, and temperature image triplets (Medwell et al., 2007). Each image 8×30mm. Jet centreline marked with dashed line. Axial location 35mm above jet exit.

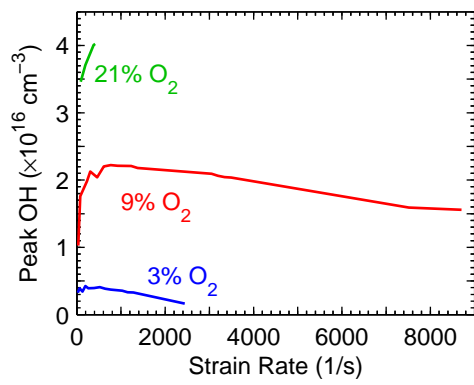


Fig. 3. Calculated peak OH number density plotted versus maximum absolute strain rate found from laminar flame calculations at various oxidant stream  $\text{O}_2$  levels.

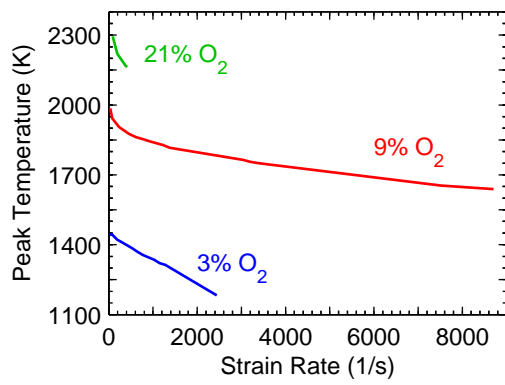


Fig. 4. Calculated peak temperature plotted versus maximum absolute strain rate found from laminar flame calculations at various oxidant stream O<sub>2</sub> levels.

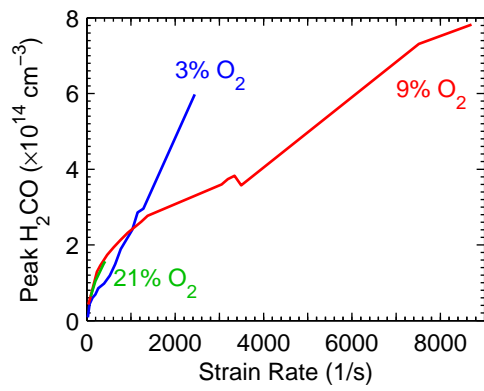


Fig. 5. Calculated peak H<sub>2</sub>CO number density plotted versus maximum absolute strain rate found from laminar flame calculations at various oxidant stream O<sub>2</sub> levels.

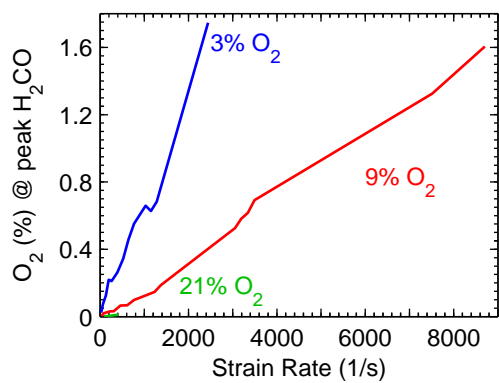
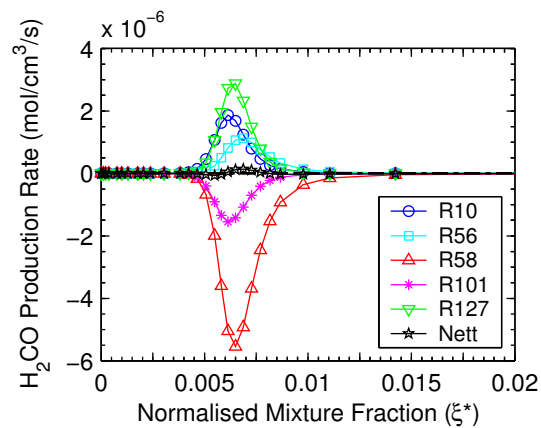
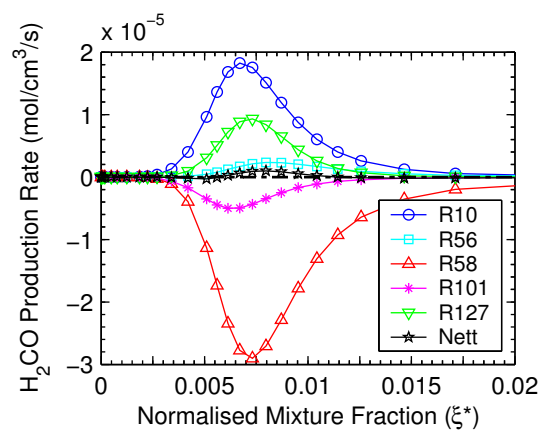


Fig. 6. Calculated O<sub>2</sub> mole fraction (%) at location of peak H<sub>2</sub>CO concentration plotted versus maximum absolute strain rate found from laminar flame calculations at various oxidant stream O<sub>2</sub> levels.



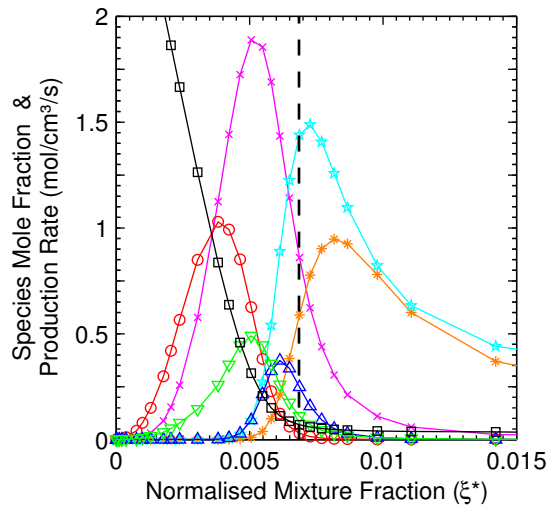


(a) Average strain rate,  $a \approx 30\text{s}^{-1}$ .

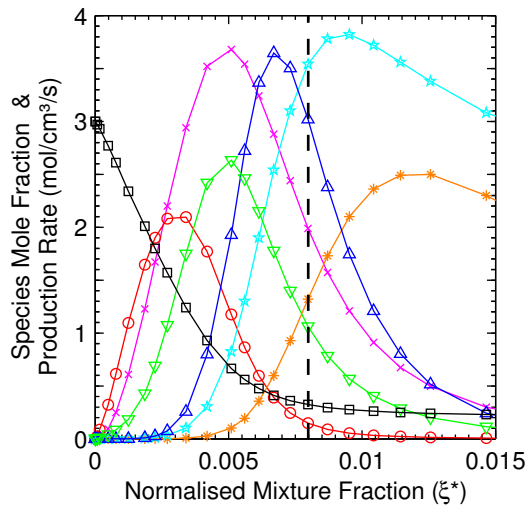


(b) Average strain rate,  $a \approx 200\text{s}^{-1}$ .

Fig. 7.  $\text{H}_2\text{CO}$  production rate via major reactions, found from laminar flame calculations. 3%  $\text{O}_2$ , 1100K oxidant stream for two different strain rates. (Note the different vertical axis scaling.)



(a) Average strain rate,  $a \approx 30\text{s}^{-1}$



(b) Average strain rate,  $a \approx 200\text{s}^{-1}$

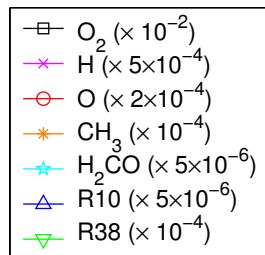
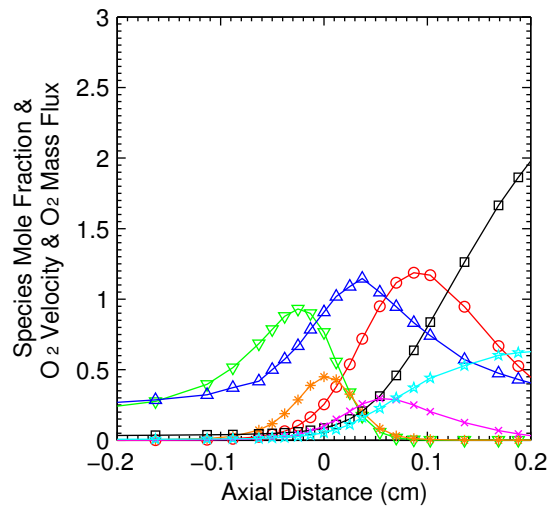
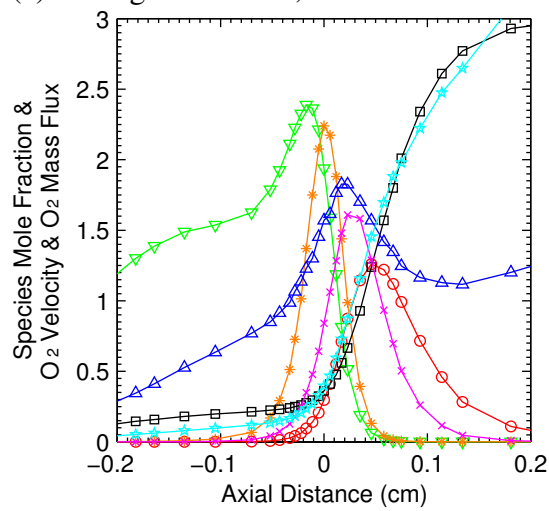


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(a) Average strain rate,  $a \approx 30 \text{s}^{-1}$ .



(b) Average strain rate,  $a \approx 200 \text{s}^{-1}$ .

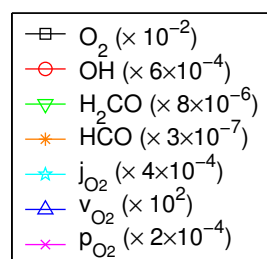


Fig. 9. Selected species mole fractions (OH, H<sub>2</sub>CO, HCO, O<sub>2</sub>), nett O<sub>2</sub> consumption rate ( $p_{\text{O}_2}$ ), O<sub>2</sub> velocity ( $v_{\text{O}_2}$ ), and O<sub>2</sub> mass flux ( $j_{\text{O}_2}$ ), found from laminar flame calculations. 3% O<sub>2</sub>, 1100K oxidant stream for two different strain rates.

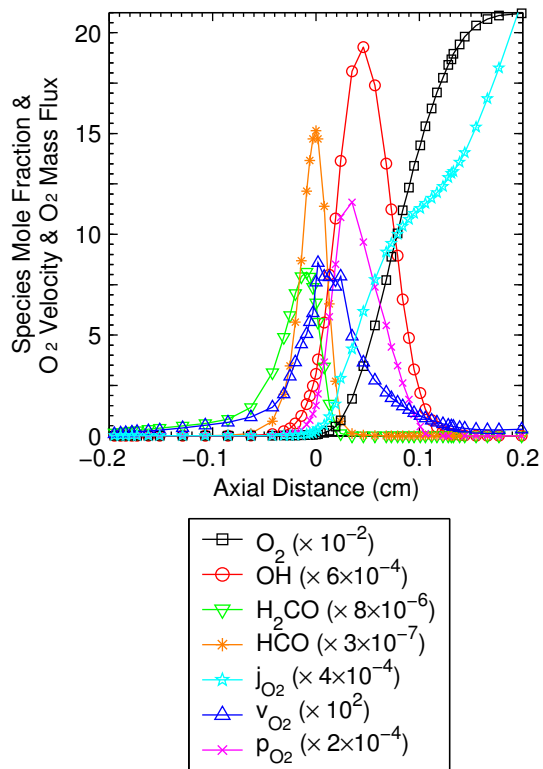


Fig. 10. Selected species mole fractions (OH, H<sub>2</sub>CO, HCO, O<sub>2</sub>), nett O<sub>2</sub> consumption rate ( $p_{O_2}$ ), O<sub>2</sub> velocity ( $v_{O_2}$ ), and O<sub>2</sub> mass flux ( $j_{O_2}$ ), found from laminar flame calculations. 21% O<sub>2</sub>, 300K oxidant stream for average strain rate,  $a \approx 200s^{-1}$ .

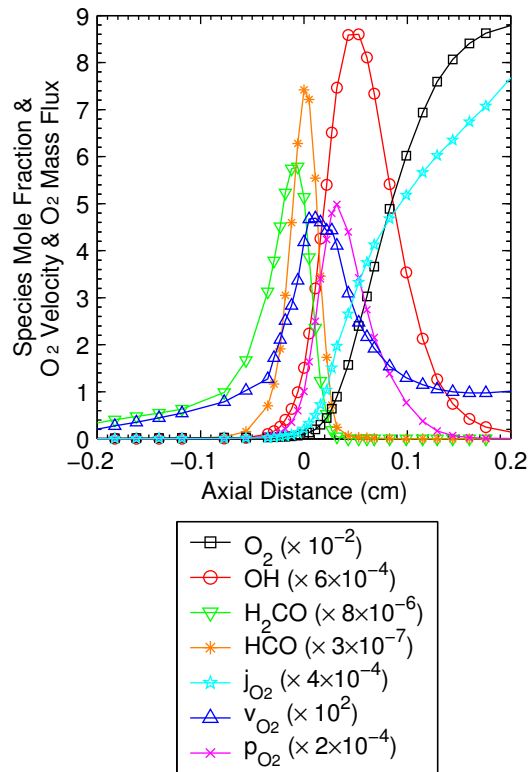


Fig. 11. Selected species mole fractions (OH, H<sub>2</sub>CO, HCO, O<sub>2</sub>), nett O<sub>2</sub> consumption rate ( $p_{O_2}$ ), O<sub>2</sub> velocity ( $v_{O_2}$ ), and O<sub>2</sub> mass flux ( $j_{O_2}$ ), found from laminar flame calculations. 9% O<sub>2</sub>, 1100K oxidant stream for average strain rate,  $a \approx 200s^{-1}$ .