EXPERIMENTAL INVESTIGATION
OF OXYGEN-ENRICHED LIFTED
AND ATTACHED TURBULENT
DIFFUSION FLAMES

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The interactions between turbulent fluid flow and chemical kinetics are examined experimentally in
attached and lifted turbulent oxygen-enriched gaseous non-premixed flames by firing a methane jet
vertically into a co-flowing oxidant shroud. Stable species, including nitrogen oxides, were measured
using physical-probing techniques. Hydroxyl radical concentrations were monitored using laser-in-
duced fluorescence (LIF). The level of oxygen enrichment in the shroud air was varied from 21% to 28% while maintaining constant average fuel-nozzle velocity and Reynolds number. Within the
operating range, attached as well as lifted flames were observed. Physical and chemical differences
in structure between attached and lifted flames are presented. With an increasing degree of oxygen,
enrichment flames become shorter and more luminous in appearance. Attached flames appear more
luminous with longer flame lengths. Significant amounts of oxygen were found on the fuel-rich side
of the stoichiometric envelope in lifted flames, underscoring partially premixed characteristics near
downstream locations.

Keywords: Nonpremixed turbulent flame; Lifted turbulent flames; Oxygen enrichment

INTRODUCTION

Recently, the enhanced temperatures achieved with oxygen-enriched combustion have found industrial use in processes ranging from glass melting to metal pro-
duction. Increased flame temperatures enhance heat transfer rates and, hence, increase thermal efficiencies. However, it is well documented that increases in oxygen levels of combustion air may lead to higher NOₓ emissions.

In the regime of modest oxygen enrichment, where potentially the most cost-
effective savings can be made in industrial processes, it is found that the increased flame temperature has a greater impact on the NOₓ formation rate than

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the corresponding reduction of the diluent nitrogen in the combustion air. In the quest for higher productivity and, hence, higher fuel throughput, non-premixed turbulent flames may be operated in a lifted mode. While oxygen enrichment enhances heat transfer, it also aids in extending the operational limits of lifted flames. Structural characteristics of oxygen-enriched attached as well as lifted flames are presented here, especially in relation to the differences in temperature and NO formation.

EXPERIMENTAL EQUIPMENT AND TECHNIQUES

The experimental apparatus consist of a turbulent diffusion burner equipped with laser-induced fluorescence (LIF), and conventional microprobe sampling and analytical train. The experiments were generally set up as shown in Figure 1. In order to avoid moving optical equipment, spatial examination of the flame was accomplished by positioning the burner with an XYZ traverse.

Turbulent Diffusion Burner Test Facilities

Atmospheric-pressure, overventilated, axisymmetrical non-premixed turbulent flames were generated with a burner in which the fuel flows from a changeable fuel tube (457 mm in length and 3.18 mm outer diameter) into coflowing oxygen-
enriched air. High-purity methane (99.9%) was used as the fuel gas. The oxygen-enriched air was delivered through a concentric outer-oxidant shroud (102 mm diameter), and the oxygen concentration of the shrouded air varied between 21–28% (flow rate = 200 liter/m STP). The flow passages contained layers of honeycomb and wire meshes, porous plates, and a contraction section to obtain a low-turbulence flow of shroud gas. The burner assembly was positioned using an XYZ traverse with 1 mm accuracy.

**Flame Gas Sampling Equipment**

A quartz gas-sampling probe mounted on an adjustable platform was used for in-flame gas sampling. The probe has a sampling orifice of 100 µm, leading to a sudden area expansion in the probe tip to quench gas-sample reactions. Filtered (25 µm filter) sample gases that were thermostatted at 375 K were either conveyed directly by means of an in-line sampling train to the gas analyzers or were collected for analysis by gas chromatography. To minimize heterogeneous catalytically-enhanced reactions on the probe surface, pacified quartz probes were used (Bowman, 1977; Heitor and Moreira, 1993).

**On-Line Gas Analyzers**

A chemiluminescence analyzer was used to measure NO and NO₂. Infrared absorption was used to measure CO and CO₂, and a paramagnetic analyzer was used to determine oxygen content. Gas chromatography was used for methane and higher hydrocarbon concentrations, and H₂, O₂, N₂, CO, and CO₂. The experimental results indicated that the repeatability was generally of the order of ±5%.

**Temperature Measurements**

Small-diameter (125 µm) silica-coated Pt-13%Rh thermocouples were employed for temperature measurements. The measurements were corrected for radiative losses using

\[ T_m = \varepsilon \sigma d / n(T_m - T_a) \]

where \( T_f \), \( T_m \), and \( T_a \) are the flame, thermocouple, and ambient temperature, respectively; \( \varepsilon \) is the thermocouple emissivity; \( \sigma \) is the Stefan–Boltzmann constant; and \( h \) is the convection heat transfer coefficient between the spherical bead of the thermocouple junction and the flame gases.

**OH Detection using LIF**

The general layout of the LIF facility is shown in Figure 2. Excitation of OH was provided by an Nd:YAG (Spectra Physics GCR-11) pumped-dye laser system.
(Lumonics HD-300), using Rhodamine 590. In order to provide a tunable narrow line-width (0.075 cm\(^{-1}\)) excitation source, the fundamental beam (1064 nm) from the Nd:YAG was frequency doubled using a KDP crystal and cleaned of any fundamentals by the sequential use of two dichroic mirrors prior to the dye laser. The output from the dye laser was frequency doubled, again using a KDP crystal, and tuned to 283.55 nm, exciting the Q1 (8) line of the \(A^2\Sigma^+ \leftarrow \Sigma^2\Pi_i\) (1,0) band of OH. This line minimizes the need for a Boltzmann-population temperature correction. The fluorescence signal was collected perpendicular to the laser beam and focused onto a nitrogen-cooled intensified linear diode array, with a 10 nm FWHM band-pass filter centered around 314 nm. In order to further minimize flame luminescence and elastically scattered UV radiation, the detector was gated (50 ns) around the emission using the Q-switch advance from the Nd:YAG laser. The linear-array detector obtained instantaneous profiles of OH through the flame.

The data presented here include the concentration and distributions of the major species, the nitrogen oxides, radiation-corrected temperatures, and the normalized OH concentrations.

**RESULTS AND DISCUSSION**

Visible flame heights and lift-off heights of the lifted flame were obtained by averaging a video record of the flames over 30 frames. The luminosity of the at-
FIGURE 3  a) temperature, b) CH₄ mole fraction, and c) O₂ mole fraction versus radial distribution (r/di) at various downstream locations in the attached flame.
FIGURE 4  a) temperature, b) CH\textsubscript{4} mole fraction, and c) O\textsubscript{2} mole fraction versus radial distribution (r/di) at various downstream locations in the lifted flame.
FIGURE 5  a) NO concentration, b) NO\textsubscript{2} concentration, and c) normalized OH concentration versus radial distribution (r/di) at various downstream locations in the attached flame.
FIGURE 6  a) NO concentration, b) NO\(_2\) concentration, and c) normalized OH concentration versus radial distribution (r/di) at various downstream locations in the lifted flame.
FIGURE 7  a) mean mixture fraction, b) temperature, and c) CO mole fraction versus mean mixture fraction at various downstream locations in the attached flame.
FIGURE 8  a) mean mixture fraction, b) temperature, and c) CO mole fraction versus mean mixture fraction at various downstream locations in the lifted flame.
FIGURE 9  a) NO concentration, b) NO$_2$ concentration, and c) normalized OH concentration versus mean mixture fraction at various downstream locations in the attached flame.
FIGURE 10  a) NO concentration, b) NO$_2$ concentration, and c) normalized OH concentration versus mean mixture fraction at various downstream locations in the lifted flame.
tached flame was significantly greater than the lifted flame, suggesting the presence of higher soot loadings for the attached case. The lifted flame exhibited blue radiation from the stabilization zone to a location approximately halfway downstream, where the remaining visible flame length existed as an orange flame in a blue sheath. A series of measurements of the flame properties for the attached and for the lifted flames are given in Figures 3–6 and presented in terms of the mean mixture fraction in Figures 7–10.

The peak temperature in the attached flame [Fig. 3(a)] was 2102 K, at downstream location $x/d_i = 60$. For the lifted flame [Fig. 4(a)], the maximum temperature occurred at the downstream location $x/d_i = 30$ and was 61 K higher than the peak temperature in the attached flame; this has an important influence on heat transfer. The peak temperatures in both cases occurred at a similar radial distance from the flame centerline.

Figures 3(b) and 4(b) display the distribution of CH$_4$ mole fraction throughout both flames. The characteristics of the plot for the attached flame are typical for a diffusion flame, with the maximum concentration occurring at the lowest height above the nozzle on the flame centerline. At downstream locations further from the nozzle, the maximum CH$_4$ concentration was reduced by dilution of inert species.

The distribution of the O$_2$ mole fraction at various streamwise locations for both the attached and lifted flames are shown in Figures 3(c) and 4(c), respectively. In the attached flame, the maximum O$_2$ concentration at each downstream location occurred at the perimeter of the flame and depleted rapidly toward the flame centerline to zero, at a radial position that increased with progressive height above the burner nozzle. It is only at the downstream location $x/d_i = 170$, where O$_2$ is present on the flame centerline, defining the flame length. The radial position at which the O$_2$ concentration approaches zero coincides with negligible CH$_4$ concentrations, suggesting fast chemistry. The above observations are similar to typical air-fed gaseous non-premixed flames.

In the lifted flame, however, O$_2$ was present at significant levels on the flame centerline at downstream locations up to $x/d_i = 60$. At these downstream locations, the centerline O$_2$ dropped significantly to a minimum value at a radial location; this was common to all the downstream sampling locations. The O$_2$ concentrations then increased rapidly to a maximum value at the perimeter of the flame. The presence of large concentrations of O$_2$ on and adjacent to the flame centerline demonstrates premixing of the fuel by entrainment of combustion oxidant upstream of the flame stabilization zone.

Radial NO distributions in both flames are shown in Figures 5(a) and 6(a). The characteristics in both graphs in relation to the location of the maximum con-
centration of NO are similar to the NO$_x$ graphs, but the peak for the lifted flame, located at $x/d_i = 60$, has shifted further away from the flame centerline. The maximum NO concentrations for both the attached and lifted flames were 112 ppm and 148 ppm, respectively, but the reaction zones are narrower. Consequently, the downstream values for the attached flame are higher than for the lifted flame, namely, 100 compared with 90 ppm. Figures 5(b) and 6(b) show that the maximum NO$_2$ concentrations were located closer to the flame centerline than they were in the cases of NO$_x$ and NO. This trend was apparent at all the downstream locations. The peaks for both the attached and lifted flames were found at $x/d_i = 60$, but in the lifted condition this peak was at an increased radius from the flame centerline. It can be deduced that the difference in the maximum NO$\times$ concentration between the lifted and attached flames can be attributed mainly to the NO concentration, as the difference in the maximum NO$_2$ concentrations was minimal at the locations of maximum NO$_x$.

Figures 5(c) and 6(c) show the hydroxyl radical distribution. The OH radical is an important species in flame chemistry because it is an effective indicator of the location and size of the reaction zone. It together with the H atoms makes an important contribution to heat transfer. Here, normalized OH concentrations have been presented as a function of OH signal intensity. For both the attached and lifted flames, the plots of normalized OH concentration verses radial distance illustrate the reaction zone development from the locations just downstream of initial propagation, to streamwise locations further downstream. The evolution of the spatial structure of the reaction zones in terms of physical width and the progressive outward shifting of the maximums at further downstream locations was a trend in both flames. In the attached case, the peak in OH concentration occurred at $x/d_i = 60$ and at $r/d_i = 5.9$, whereas in the lifted case, the peak occurred at the same downstream location but at $r/d_i = 6.3$.

The measurements indicate wider reaction zones for the lifted flame at similar downstream locations. The differences in radial location of the maximum OH concentrations at specific downstream locations were more evident in the initial stages of flame propagation. The concentration of OH in these flames appeared to be more significant in the first quarter of the flame, with very defined peaks and maximum concentrations existing in this region.

The levels of OH at locations downstream of the first quarter, for example at $x/d_i = 115$, were very much reduced for both flames and, at downstream locations beyond $x/d_i = 170$, no defined peaks were observed.

Profiles of nitrogen oxides, temperature, OH, and stable species as a function of mean mixture fraction ($\bar{\xi}$), have also been presented to elucidate flame structure further. The mean mixture fraction presented in this study was calculated
by a carbon-atom balance from measurements of the fuel, CO, and CO₂ (Masri and Bilger, 1986):

$$\xi = \left( \frac{12}{16.032} \bar{Y}_{\text{CH}_4} + \frac{12}{28} \bar{Y}_{\text{CO}} + \frac{12}{44} \bar{Y}_{\text{CO}_2} \right) \bar{Y}_{C,i}$$

(2)

where $\bar{Y}$ represents the mean mass fraction of species $i$ and $\bar{Y}_{C,i}$ is the mean mass fraction of carbon atoms in the main fuel ($\bar{Y}_{C,i}$ for methane).

Profiles of mean temperature and species mass fractions as a function of the conserved scalar mixture fraction (based on temporally averaged species concentrations) are presented in Figures 7–10. For the present study, the values for mass fraction of the stable species were corrected to a wet basis by calculating the water content of the flame based on the carbon/hydrogen ratio.

To illustrate levels of mixing and oxidant entrainment in these flames, radial profiles of mean mixture fraction at the various axial locations are shown in Figures 7(a) and 8(a) for the attached and lifted flames. For the attached flame, the radial gradients of the mean mixture-fraction at the two initial downstream locations are steep as shown in Figure 7(a). In addition to this, the temperature, NOₓ, OH, and various stable species profiles for this flame at these downstream locations all exhibited narrow peaks centered near the flame axis, as the flame was initially restricted to a narrow region in space at these locations. With downstream development, the mean-mixture-fraction profiles are flatter, and, consequently, the peak profiles of temperature, NOₓ, OH, and various stable species are wider. The lifted flame showed similar characteristics [Figure 8(a)]. However, at the initial downstream locations, the mean-mixture-fraction gradients were less steep consistent with partial premixing upstream of the stabilization region.

Figures 7(b) and 8(b) show the radial-temperature profiles at various streamwise locations along the length of the flame for the attached and lifted regimes, respectively. In the case of the attached flame, the peak temperature of 2102 K occurred slightly to the rich side of stoichiometric at the downstream location $x/di$ = 60. In fact, for the attached flame, generally, peak temperatures occurred in the fuel-rich zone of the flame.

For the lifted flame, the maximum temperatures at the initial individual downstream locations, from the stabilization region up to and including $x/di$ = 30, were similar in magnitude, with the peak temperature of 2163 K occurring at the downstream location $x/di$ = 30. The peak temperature in the lifted flame was spread over a significantly broader area in mixture-fraction space compared to the attached flame, which had a narrower peak. The broad peak of the lifted flame started at the stoichiometric surface and progressed well into the lean region of
the flame. Gore and Zhan (1996) suggested other characteristics that could indicate the presence of a double flame structure. These included a broad minimum O₂ mole fraction and broad peaks in the mole fraction of H₂O, both in physical space. These properties are thought to indicate the presence of two reaction zones here.

Figures 7(c) and 8(c) show the mean mole fractions of CO plotted in mean-mixture-fraction space for the attached and lifted flames, respectively. Vertical dotted lines indicate the stoichiometric mean mixture fraction (ξ). The maximum CO concentrations of both the attached and lifted flames was at the downstream location x/di = 115 on the fuel-rich side of stoichiometric [Fig. 7(c) and 8(c)]. For the lifted flame, the maximum CO concentrations up to x/di = 115, all occurred around a mean-mixture-fraction value of 0.1–0.11, with this maximum value being particularly rigid in its location in mixture-fraction space. In contrast, the peak concentrations at all the downstream locations up to x/di = 115 for the attached flame were distributed over a wider mean-mixture-fraction range (0.1–0.15). A similar observation was seen for H₂ concentrations. At similar downstream locations up to x/di = 115, the attached flame exhibited a wider range of peak concentrations in mixture-fraction space than the lifted flame. Also, with the attached flame, these peaks in both H₂ and CO mole fraction shifted to a more lean position when moving to a higher downstream location, indicating diffusion of these intermediate combustion products into the oxidant stream.

The significant amount of oxygen present on the rich side of stoichiometric in the lifted flame points directly to premixing. For the lifted flame, the minimum O₂ concentration at all the downstream locations occurred in the vicinity of the stoichiometric line, with the minimum mole fraction in all cases being approximately 0.025. The distributions of all the major species in mixture fraction space in the attached flame are significantly broader than in the lifted flame. For the attached flame, these stable species profiles, regardless of axial position in the flame, exhibit an almost unique function of mixture fraction. This is indicative of fast chemistry.

The results presented in mixture-fraction space are typically used to describe a diffusion flame. The failure of plots in mixture-fraction space to describe a lifted flame indicated the significant departure of a lifted flame from the characteristics of turbulent diffusion-flame behavior.

The relationship between NO concentration and mean mixture fraction at various streamwise locations for both the attached and lifted flames are show in Figures 9(a) and 10(a). These plots show similar trends in relation to the graphs of NO₅ verses mean mixture fraction for the respective flames. However, because of the influence of NO₂ on the NO₅ concentration, there is an obvious reduction
of product on the rich side of stoichiometric when comparing the NO to NO$_x$ concentrations.

As mentioned above, there was a significant increase in the maximum NO$_x$ and NO concentrations in the lifted flame compared to the attached flame. Also, the maximum temperature observed in the lifted flame was 61 K higher than in the attached flame, with this location in mean-mixture-fraction space coinciding with the location of the maximum NO concentration in the lifted flame, on the lean side of stoichiometric. Furthermore, the significant broadening of the high-temperature region in the lifted flame also influences the production of the temperature-sensitive NO. It has been well established (Miller and Bowman, 1989; Missaghi et al., 1990) that the thermal mechanism of NO formation becomes prominent at elevated temperatures in excess of 2000 K. Hence, the increased magnitude and width of the maximum temperature profile in the lifted flame suggest increased NO concentrations attributed to thermal NO formation.

The presence of the OH radical in both flames in the high-temperature, high-NO concentration regions suggests that within the reaction zones of the respective flames the OH radical may contribute to the formation of thermal NO. The contribution of OH within the "extended Zeldovich mechanism" may be more influential in the slightly fuel-rich areas of the flame where the presence of O radicals is deficient, as suggested by Miller and Bowman (1989). In fact, detailed calculations in opposed diffusion flames suggest that N$_2$ + OH = NO + H is part of the most prolific channel for NO formation in diffusion flames (Yap et al., 1998).

The role of the prompt mechanism in the formation of NO is thought to be significant in methane-jet flames. The OH radical plays an important role in the oxidation process during the prompt NO-reaction scheme (Miller and Bowman, 1989). The prompt mechanism is associated with lower temperatures than the extended Zeldovich mechanism, short residence times (increased strain rate), and fuel-rich environments.

Figures 9(b) and 10(b) show the distribution of NO$_2$ in mean-mixture-fraction space for both the attached and lifted flames, respectively. For both flames, the NO$_2$ concentrations peaked on the rich side of stoichiometric. Both flames exhibited maximum concentrations of NO$_2$ at the downstream location $x/di = 60$. Also, at the furthest downstream location, the maximum NO$_2$ concentrations were significantly reduced.

Formation and destruction of NO$_2$ is believed to occur via the reactions of NO with O$_2$, O, OH, and HO$_2$ within the flame zone of a combustion process (Hanson and Saliman, 1984). Hori (1988) suggested that NO converted to NO$_2$ in the mixing region of jets, mainly via oxidation with the HO$_2$ radical. Further ex-
periments (Hori et al., 1992) established that the conversion of NO to NO\textsubscript{2} is dominated by reactions of NO with HO\textsubscript{2} and are strongly promoted by the presence of small concentrations of fuel. Higher conversions are obtained from fuels that easily decompose to produce active species and, consequently, produce high HO\textsubscript{2} concentrations. Also, the conversion of NO to NO\textsubscript{2} appeared to proceed strongly within a certain temperature range.

The present observation that NO\textsubscript{2} in both the attached and lifted flames was more prominent on the rich side of stoichiometric suggests that the formation route in these flames was via the reaction of NO with HO\textsubscript{2}. This is because the regeneration of HO\textsubscript{2} is generally coupled with the presence of H\textsubscript{2}, CO, and hydrocarbon fragments. These zones coincide with areas of significant NO\textsubscript{2} contributions. Bonturi et al. (1996) also concluded in their work that a peak in NO\textsubscript{2} was found on the rich side of stoichiometric, with its presence in a similar region in mixture-fraction space to a peak in HO\textsubscript{2} radicals.

The profiles of normalized OH concentration as a function of mean mixture fraction for the attached and lifted flames are presented in Figures 9(c) and 10(c). For both flames, the maximum OH concentrations occurred on the lean side of stoichiometric, at the downstream location \(x/d_i = 60\). The concentration of OH remained high in relation to the maximum concentration at the downstream locations up to \(x/d_i = 60\), with significant reductions observed at the axial locations beyond the maximum. The reduction in the OH concentrations between the downstream locations of \(x/d_i = 60\) and \(115\) was 47.5\% for the attached flame and 23.4\% for the lifted flame.

The significant reduction in the OH concentration beyond \(x/d_i = 60\) could be explained via the following routes. First, it has been well documented (Fristom, 1995; Puri and Santoro, 1994) that the principle route of CO oxidation is by reactions with the OH radical. The presence of CO\textsubscript{2} — this being a product of the CO oxidation process — is prominent at downstream locations further than \(x/d_i = 60\) in both of these flames. Second, the OH radical is thought to be a significant player in the formation of NO via both the thermal and prompt routes (Barlow and Carter, 1996). Third, evidence presented by Puri and Santaro (1994) indicated not only the oxidation of CO by OH, but also the oxidation of soot by OH in the sooty regions of a flame, which may justify the differences in OH concentrations seen between the attached and lifted flames at the downstream location \(x/d_i = 115\).

In the attached flame, this downstream location provided the highest soot concentrations in relation to other sampling locations, with the lifted flame showing the presence of no significant quantities of soot in any regions. From the differences in the OH distributions for the two flames up to the downstream location
x/d_l = 60, it is apparent that the peaks in the attached case shifted to a more lean position compared to the lifted case at similar streamwise locations. The lifted flame also had broader peaks in mixture fraction space, but the overall range of the attached flame in mixture-fraction space was wider.

It has been concluded (Bonturi et al., 1996) that the significance of the prompt mechanism in the formation of NOx becomes more apparent as the strain rate within the flame increases. They also concluded that as the strain rate was reduced, the thermal NO mechanism was enhanced. Hence, it is plausible that both attached and lifted flames exhibit thin NO-production zones near the flame base, but broaden further downstream [Fig. 5(a) and 6(a)]. This observation is consistent with OH and temperature profiles [Fig. 5(c) and 6(c)]. The influence of local strain on the narrow reaction zones near the flame base is thought to reduce temperatures and suppress the formation of radicals, leading to the sensitivity of NO formation at these initial downstream locations.

Further downstream, scalar dissipation decreases, and as a result, temperature and radical concentrations approach equilibrium, allowing for broader temperature zones to develop and culminating in an increase in NO-production rates (Barlow and Carter, 1996). The presence of significant concentrations of both O2 and N2 at the flame centerline indicate partial premixing. This influences the formation of NOx in the lifted flame. As both O2 and N2 are the major reactants in the formation of NO, the presence of these reactants along the flame centerline could also account for the increased NOx levels observed in the lifted flame, especially at the initial downstream locations.

**CONCLUSIONS**

The physical and chemical appearances of the attached and lifted flames were significantly different. In the lifted flame, partial premixing was observed and the lifted flame data suggest the presence of a “double-flame structure.” Consequently, the peak in-flame NOx and temperature measurements were observed to be higher in the lifted case, with a lean shift of these peaks in mixture-fraction space apparent in the lifted flame compared to the attached flame.

These changes will have a significant influence on the application of these flames in practice. The flame temperatures of the lifted flame is higher than the attached flame, 2163 K compared with 2102 K, and this will have a marked influence on the heat transfer rate, increasing it by typically 8%. This is further enhanced by the increase in the width of the reaction zone and the increase in the OH, so that, overall, the increase in heat transfer is higher. The formation of NO shows slightly different behavior. While the reaction zone values show a marked
increase in NO concentrations, the downstream values are slightly less for the lifted flames. Consequently, in practice the lifted flames show considerable advantages.

References


