EFFECTS OF JET DILUTION AND CO-FLOW ON SOOTING AND EMISSION CHARACTERISTICS OF HYDROCARBON FUELS

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A study was conducted to understand the effects of dilution and co-flow on the sooting and pollutant emission characteristics of hydrocarbon fuels in the vicinity of their smoke point. Measurements of the critical mass flow rate of a fuel at the threshold of smoking and the mass flow rate of the dilution gas (nitrogen) required to suppress smoking at several fractions of the critical fuel mass flow rate were obtained. The curve of the variation of nitrogen flow rate that was required for smoke suppression with fuel flow rate exhibited a skewed bell shape. Two regions, a chemically controlled region and a fluid dynamics-controlled region were identified on this curve. Just prior to the condition at which the flame turned nonsmoking, at the critical (fluid dynamics-controlled region) and at the 30% of the critical fuel mass flow rate (chemically controlled region), flame characteristics including radiation emission, temperature profiles, radial concentration profiles, emission indices of NO, NOx, CO, and flame heights were also measured. Also recorded were the axial profiles of radiation power and soot volume fraction at those conditions. Propylene (C3H6) was used as the fuel in the study. A 3.2 mm ID burner was employed.

INTRODUCTION

Many researchers have studied diffusion flame characteristics due to their importance in combustion systems. Sooting is one of the major characteristics of diffusion flame combustion processes. Smoke point is a conventional means to characterize the flame’s sooting behavior.

A well-accepted definition of flame smoke point refers to a condition just before soot escapes unburned from a laminar flame. Smoke point height, taken as the visible laminar flame height at the threshold of smoking, has been used as a measure of sooting tendency of fuels, and is correlated to the flame radiation characteristics (Markstein, 1984; Markstein et al., 1984). Roper and Smith (1979) have found that at the smoke point soot oxidation becomes negligibly small. Glassman and Yaccarino (1980) defined smoke point as the condition where the ”wings” (due to soot breakthrough) reached the same height as the corresponding flame apex.

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Although there have been several methods to determine laminar flame smoke point, there is hardly any literature concerning the methods of defining or measuring the turbulent flame smoke point. Because of the proportionality of flame height and fuel flow rate in laminar flames (Burke and Schumann, 1928; Roper, 1977) the smoke point height is a good measure of sooting propensity of fuels in laminar flames. In turbulent flames, where the linearity between the fuel flow rate and flame height is not valid (Hottel and Hawthorne, 1949), the smoke point height is not a valid indication of sooting tendency. Hence, the fuel mass flow rate, which also accounts for the carbon input rate into the flame, is a better scale of sooting characteristics in turbulent flames. In a previous study (Goh et al., 1999), the authors developed a laser-based technique to determine the smoke point of turbulent diffusion flames and determine the sooting characteristics of selective hydrocarbon fuels.

At the smoke point transition, sudden and substantial changes in flame characteristics, such as flame luminosity, flame color occur. These changes suggest that significant changes in thermo-chemical mechanisms should also occur. Thus, it is logical to expect marked changes in temperature field, species concentration profiles, and pollutant emissions at the smoke point. The flame structure data, at the conditions close to smoke point are not available in the literature, which prompted this study. In this study, the flame radiation, temperature profiles, concentration profiles, flame height, and emission indices of NO, NO₅, and CO near the smoke point condition of turbulent diffusion flames of three hydrocarbon fuels have been investigated (Goh, 1999; Kusadome, 1999). This paper deals with only propylene data.

**EXPERIMENTAL TECHNIQUES**

Experiments were performed in the University of Oklahoma Combustion Test Cell in a controlled environment. The schematic of the experimental setup is shown in Figure 1.

A burner with an exit diameter of 3.2 mm ID was used. The burner was made of two concentric circular steel tubes. The inner tube was supplied with a hydrocarbon fuel or hydrocarbon fuel-nitrogen mixture and the outer tube was supplied with hydrogen to function as the flame stabilizer (to keep the flame attached to the burner; only attached flames were studied to avoid the ambiguity of the effects of complex fluid dynamics of lifted flames on smoke point). In the annular space between the tubes, fine steel wool was stuffed to provide a uniform flow of hydrogen. The average hydrogen flow rate needed to attach the flame was very small compared to the fuel flow rate (hydrogen flow rate was less than 0.7%
of fuel flow rate by volume and contributed to less than 1% of the total energy release). The fuel used in the experiments was propylene. It has been known that the addition of diluents changes smoking characteristics significantly (Gollahalli, 1977). Hence, nitrogen was used as the diluent to trigger the smoke point at fuel flow rates below the critical smoke point fuel flow rate of the pure fuel.

To provide a small velocity co-flow air stream around the flame, an arrangement consisting of a steel box and a blower was fabricated. A 101.6 cm × 101.6 cm square cross-section opening and 50.8 cm high steel box was connected to a blower with a 20.3 cm diameter duct. Air entered through the sidewall of the box and exited through the open area on the top. To ensure a uniform flow, two screens were placed at the opening of the box separated by a distance of 7.6 cm. The velocity of air exiting the box was uniform on the middle two thirds of the opening where the flames existed. The air velocity was measured using a hot-wire air anemometer. The average co-flow velocity was 1.07 m/s. The air velocity varied only by 0.15 m/s from burner plane to 1.2 m above the burner exit.
The fuel gas supply train consisted of propylene and hydrogen cylinders, pressure regulators, rotameters, and manifolds. Each cylinder was mounted with a two-stage pressure regulator. There were three different gas supply trains, one each for fuel, hydrogen, and nitrogen.

A set of ball-rotameters was used to measure fuel, nitrogen, and hydrogen flow rates. These flow meters were calibrated with a wet test meter for all fuels, nitrogen and hydrogen, with different floats and tubes to cover the wide range of flow rates desired. Before entering the burner, both the fuel and nitrogen lines were combined using a Y connector. The hydrogen line was similar to the fuel and nitrogen lines. Downstream of this connection, the line was 2 m long to ensure a homogeneous mixture of fuel and diluent at the burner exit.

Several smoke point determination methods were tried (Goh, 1999), but the one that was selected due to its good repeatability is described here. This method involved observing the visibility of a laser beam just above the flame. When a flame started to smoke, the laser beam became visible due to scattered radiation, and the intensity of the illumination of the laser beam was a function of the concentration of smoke. A viewing angle of about fifteen degrees from the laser beam in the forward scatter mode was found to yield repeatable and sensitive results. The top view of the position of the observer relative to the laser beam is shown in Figure 2. The condition at which the laser beam located approximately 7.5 cm above the flame tip just became visible as a continuous beam was taken as the smoke point. The nominal experimental conditions and the measures of repeatability are given in Tables I and II.

A wide-angle (150-degree viewing angle) highly sensitive pyrheliometer of absorptivity 0.96 was used for flame radiation measurement. The radiometer was mounted on a tripod at approximately the mid-height of the flame and far enough to satisfy the inverse-square law. The radiative fraction of heat release was calculated assuming complete combustion, which was reasonable because the flame was at the verge of becoming smokeless. To record the radiation profile data, the

![FIGURE 2 Laser beam visible location.](image)
view-angle of the radiometer was modified with masks to capture only the radiation from a horizontal slice of the flame and the radiometer was traversed parallel to the flame axis.

The temperature was measured with an in-house-made thermocouple of type R (Platinum-Platinum 13% Rhodium) with bead diameter 280 µm. The output of

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<th>TABLE I  Experimental conditions</th>
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<td>Fuel jet diameter</td>
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<td>Characteristic diameter of hydrogen jet</td>
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<td>Burner length</td>
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<td>Fuel property: Propylene</td>
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<td>(Vol. %)</td>
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<td>Propane</td>
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<td>Ethane</td>
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<td>Fuel or mixture velocity range</td>
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<td>Fuel or mixture Reynolds number range</td>
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<td>Diluent (Vol. %) Nitrogen</td>
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<td>Flame stabilizer agent: Hydrogen</td>
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<td>Temperature profiles</td>
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*95% confident level
the thermocouple was acquired with a PC computer equipped with an A/D converter at a sampling rate of 1 kHz. The instantaneous data were time-averaged for two minutes. The temperature profiles were measured at three locations, near-burner (25% of the visible flame length), mid-burner (50% of the visible flame length), and far-burner (75% of the visible flame length). The measured temperatures were corrected to account for radiation, convection, and conduction losses.

A helium-neon laser beam of wavelength 635 nm provided the light source for the attenuation measurement. The laser beam passed horizontally through the flame field to the receiver on the opposite side (Figure 2). The power incident on the receiver was recorded with and without flame for each measurement. The signal fluctuation due to the turbulent flame structure was averaged in the analog meter. The laser attenuation measurements were carried out on the flame centerline at selected heights to obtain axial profiles. Soot volume fraction was calculated from attenuation data using Mie theory (Yagi and Iino, 1962).

An NDIR (nondispersive infrared) analyzer was used for the CO and CO$_2$ concentration measurements. Oxygen concentration was obtained by using galvanic analyzer. The NO and NO$_x$ concentrations were measured with a chemilu-
minescent analyzer. The exhaust gas was collected through a conical duct and mixed well before measuring its composition. The sampling gas flow rate was kept constant and measured by a rotameter.

The radial concentration profiles of CO, CO₂, NO, and O₂ were measured by sampling the emission gas in flame with an uncooled quartz probe. The probe was mounted on a two-dimensional traverse system that allowed movement in the radial and vertical directions. The instrumentation was the same as that for the emission index measurement except the CO analyzer. A higher range NDIR CO analyzer was employed because in-flame CO concentration was higher than the global-flame CO emission. Figure 3 shows the instrumentation setup.

A Canon SLR camera with Kodak Gold 100 ASA film and a Sony digital camera were used to obtain the flame images with three different shutter speeds (one second, 1/125 second, and 1/250 second). The flame height was taken as the distance from the burner tip to the end of contiguous image of the visible flame on the one-second exposure photographs.

RESULTS AND DISCUSSION

Smoke Point

The results are presented in Figure 4 in the form of the variation of the critical fuel mass flow rate versus the mass flow rate of nitrogen to be added to the fuel to achieve the smoke point. The critical mass flow rate is defined as the mass flow rate of the pure hydrocarbon fuel at the smoke point of its attached flame. Nitrogen flow rate rose from zero at the 100% critical mass flow rate, attained a

![Figure 4](image-url)
maximum value at a certain fraction of the critical flow rate, and then decreased to zero at the zero fuel mass flow rate.

The co-flow (approximately 1.07 m/s) provided in this experiment did not have a significant influence on the smoke point. Hence, the rest of experiments were conducted in a quiescent environment.

The maximum amount of nitrogen needed to achieve the smoke point occurred at approximately 0.006 kg/min (around 25% of the critical mass flow rate for propylene). The maximum nitrogen mass flow rate at that point was 0.007 kg/min.

Smoke emission from diffusion flames basically depends on the soot formation and burnout processes occurring in the flame. Both these processes are functions of the soot-precursor species concentrations, temperature distribution, soot trajectories, and residence time of the soot particles in the flame field. In a diffusion flame, if soot concentration increases with residence time, then chemical control is more important; on the other hand, if the residence time does not change, diffusion control is more important (Kent and Wagner, 1984). Both these distinct processes were exhibited in the results. Two distinct regions separated by the maximum nitrogen mass flow rate can be observed in Figure 4. Let us define Region I as the region on the right of the maximum value and that on the left as Region II.

In Region I, the amount of nitrogen needed to achieve smoke point increased with decreasing fuel mass flow rate. As the fuel mass flow rate was decreased, the fuel jet velocity and consequently the air entrainment into the flame also decreased, the latter at a higher rate than the former. Thus, in order to reach the smoke point condition, (completely suppress smoke liberation) more nitrogen was needed to intensify the flame turbulence and air-fuel mixing rate. Hence, it can be concluded that this region was fluid-dynamics-controlled or it may be described as a momentum-dominated region. This phenomenon had also been observed by Markstein (1984) in an experiment on an ethylene-nitrogen mixture. The addition of nitrogen to the ethylene flame resulted in an increase of smoke point height, which was attributed to the transition from natural to the forced convection. The evidence of the transition was the change of the flame structure, which was the disappearance of the large eddies and the appearance of small-scale turbulent structures.

In Region II, the amount of nitrogen supplied decreased with a decrease in fuel mass flow rate. In the study by Glassman and Yaccarino (1981), nitrogen was added to the fuel to alter the flame temperature. They found that increasing nitrogen mass flow rate could reduce flame temperature. The lower the flame temperature, the lower was the flame tendency to smoke. This was because the
fuel pyrolysis process was dependent strongly upon the flame temperature; the lower the temperature, the lower the rate of pyrolysis reactions. The temperature data of the flames are available in the thesis by Kusadome (1999). The Region II results agreed with the studies of Glassman and Yaccarino (1981), Kent and Wagner (1984), Markstein (1984), and Orloff et al. (1992). Both Markstein (1984) and Orloff et al. (1992) showed that the soot temperature dropped below 1300 K to 1600 K, which was the critical temperature for soot to cease its burnout regardless of location. Hence, it could be concluded that the Region II was a chemical reaction dominant region.

The curves in Figure 4 resemble the curve of critical smoke point ethylene flow rate versus oxygen index (mole fraction of oxygen in the co-flow stream) given by Glassman and Yaccarino (1980). They also distinguished two regions, the first in which the fuel flow rate increased with the increase of oxygen index (similar to the reduction of nitrogen in our study), and the second in which fuel flow rate decreased with the increase of oxygen index. They attributed these regimes to the dominance of particle burn-up and fuel pyrolysis processes with significantly different activation energies. It is interesting to note that in the experiments of Glassman and Yaccarino (1980), the local oxygen concentration was changed by changing the diluent content of the air stream, whereas in our experiments we added the diluent to the fuel stream; nonetheless, similar regimes are noted.

**Flame Appearance**

The color photographs of propylene flames at various fuel flow rates at the smoke point conditions are available in Goh (1999). At the critical smoke point flow rate of propylene, the entire flame appeared luminous, except for a small region near the burner rim, which was bluish in color. The bluish region was due to the gas-phase oxidation reactions caused by the premixing of the fuel and air in the flame-anchoring region. The overall yellow flame starting close to the burner indicates the formation of soot close to the burner and soot combustion over most of the flame length. As nitrogen was added, the flame became less luminous, while still maintaining yellow color. Even at the smoke point threshold, the flame was still yellow.

The visible flame length was measured as the distance from the burner exit to the contiguous part of the flame on one-second exposure photographs. The flame length was 220$d$, 216$d$, 190$d$, and 128$d$, at 100%, 75%, 50%, and 15% of the critical smoke point flow rate, respectively, where $d$ is the fuel jet exit diameter.
Flame Temperature

Figure 5 shows the radial temperature profiles at these axial locations: near-burner (0.25 $H$), mid-flame (0.5 $H$), and far-burner (0.75 $H$), where $H$ is the visible flame length. It is interesting to note that even at the critical smoke condition the temperature profiles exhibit the same shapes as in diffusion flames at other conditions (Gollahalli, 1977). The double hump structure with peak temperature of 1800 K is noticed in the near-burner region, which is comparable to the values obtained in non-smoking propane flame (Gollahalli, 1977). The double-hump structure occurs because of the gas phase-dominated interfacial reactions. The fact that these peaks occur even at the smoke-point threshold illustrates that the gas-phase reactions are still dominant in the near-burner region, although the pyrolysis reactions leading to soot formation have already begun, as seen in flame photographs.

At the mid-flame, the double hump structure vanishes and a single axial peak appears in radial temperature profiles. The absence of even flattened peaks, which are often noted in the mid-flame regions of non-sooty flames, is a definite indication of the dominance of soot combustion. The peak temperature of 1700 K is less than that in the near-burner region, which further confirms the slower and more spatially distributed soot oxidation reactions and the concomitant heat losses.

In the far-burner region, a single peak still continues with a peak temperature of 1475 K. The drop in the peak temperature is due to reduction of soot oxidation and radiative cooling. This value is in the range of 1300 to 1600 K, where soot ceases to burn. Markstein and De Ris (1984), quote this value as 1600 K for ethylene and propylene, whereas Kent and Wagner (1984) state that this value is in the range of 1300 to 1400 K for several fuels including ethylene, ethane, pro-

![FIGURE 5 Flame radial temperature profiles at 100% critical fuel mass flow rate.](image-url)
pane, butylenes, and acetylene. The discrepancy in their values was due to the difference in the criterion used to specify the smoke liberation location. Kent and Wagner take it as the axial location where the soot volume fraction curves flatten out, whereas Markstein and De Ris (1984) give the value at the laminar smoke-point height.

In the nitrogen-diluted flame at the 30% critical smoke point condition, the shapes of temperature profiles (Figure 6) remain the same as that at the critical point flow rate of pure fuel. Nevertheless, we notice that the flame is narrower and the peak temperature in the near-burner region is higher (approximately 1900 K). It can be attributed to the lowering of endothermic pyrolysis reactions in that region due to nitrogen that leads to lower soot formation. The peak temperature in both the mid-flame and far-burner regions are also higher by about 100 K due to lower soot concentrations in the diluted fuel flame. Nonetheless, the far-burner peak temperature is still in the range of temperature at which soot oxidation ceases.

**Radiation**

Figure 7 shows the radiation data. Radiation and the flame height of a diffusion flame are strongly coupled with the soot concentration in the flame. A luminous flame indicates the oxidation of soot, which leads to a high radiation emission; also, if the residence time of soot is long, the flame length increases. The radiation power and the radiative fraction of heat release rate \( (F) \) varied approximately linearly, with the critical fuel mass flow rate. The maximum radiation power was emitted from the propylene flame at the critical fuel mass flow rate. The radiant
power of propylene flame was 7.0 kW and 2 kW at the 100% and 30% critical fuel mass flow rate.

Orloff et al. (1992) related the radiant fraction in a diffusion flame to three parameters: the fuel smoke point, the oxidant to fuel stoichiometric mass ratio, and the adiabatic stoichiometric flame temperature. They developed a correlation $F = 0.374 - 0.334 Q_{sp} \text{ (kW)}$, for turbulent flames, where $F$ is the radiative fraction of heat release, and $Q_{sp}$ is the heat release rate at laminar smoke point. The value of $F$ obtained by these authors at standard conditions ($T_{ad} = 2200$ K, and stoichiometric oxidant to fuel ratio = 15) for propylene is 0.38 and for the flame in pure air is 0.44. The maximum value of $F$ in Figure 7 is 0.39 at the critical smoke-point fuel flow rate, which confirms the range of values obtained by Orloff et al. (1992).

The axial radiation power profile was measured at the 100% critical fuel mass flow rates. Figure 8 shows the results in the form of flame radiation emission versus normalized flame height. The shapes of the curves are similar for all flames. In the near-burner and far-burner regions the flame radiation is low, but at the mid-flame region the flame radiation is the highest because most of the soot oxidation took place in the mid-flame region. The radiation power curve is similar in shape to the axial soot concentration profile (Figure 9) for the pure fuel and the peaks in both curves occur at about $h/H = 0.4$. Nevertheless, the peak radiation occurs in the diluent-fuel flame at about $h/H = 0.4$, whereas the peak in soot concentration occurs at $h/H = 0.7$. This clearly demonstrates the strong influence of soot concentration through emissivity on radiation in pure fuel flame at its smoke point. Nevertheless, in the diluent-fuel flame, the lower soot concentra-
tion makes its influence on radiation smaller and the temperature effect becomes significant.

**Pollution Emission**

Figure 9 shows the axial concentration profiles of the soot concentration in at the critical and 30% critical smoke point conditions. The profiles are somewhat similar to those given by Kent and Wagner (1984) for ethylene flames. We notice that the soot concentration levels off around the visible flame height, which is in agreement with their results. Further, the axial location of the peak soot concentration also agrees with their value given for an acetylene turbulent diffusion
flame at the smoke point. Nevertheless, at the 30% critical smoke point, the peak occurs later, at about 0.75 $H$, which can be attributed to slower formation of soot in nitrogen-diluted-fuel flame.

Radial profiles of CO and NO concentrations at the critical smoke point flow rates at three axial locations of propylene flames are shown in Figures 10a and 10b. The corresponding profiles at the 30% critical smoke-point flow rate are given in Figures 11a and 11b. As expected, NO concentration profiles follow temperature profiles for mid-flame and far-burner regions because of the dominance of the Zeldovich mechanism. The highest peak concentrations occur in the near-burner region for NO, which decrease significantly in the mid-flame and far-burner regions. Zeldovich mechanism dominated most part of the flame, except at the near-burner region. At the near-burner region, fuel pyrolysis was still taking place in the flame. As a result, this region contained a high concentration of hydrocarbon radicals. Hence, the prompt NO mechanism may be important in this region. Because most of the hydrocarbon radicals concentrate at the fuel rich core, due to the fast formation of NO through prompt mechanism, a high NO concentration was observed at the center region; as seen in the NO concentration profiles in the near-burner region for both 30% and 100% CFMFR flames. The mid-flame NO concentration for 100% CFMFR flame possesses dual peaks, though the temperature profile does not show significant dual peaks. Besides temperature, the $O_2$ concentration is also an important factor in the Zeldovich mechanism. The $O_2$ concentration at this location (Kusadome, 1999) attained a minimum at the center of the flame. Hence, the low availability of $O_2$ could cause a dip in the NO concentration at the mid-flame of the 100% CFMFR flame. The NO concentration profile in the 30% CFMFR flame, which was short and narrow, did not resolve the two-peak structure, which is due the limited spatial resolution of measurement. It is interesting to note that at the critical smoke-point flow rate, the peak CO concentration in the near-burner and the mid-flame is substantially higher than in propane flames in air (Gollahalli, 1977). The higher concentrations are due to the smoke-point critical flow rate and due to the differences in molecular structure between the fuels. Nevertheless, the peak CO concentration drops to significantly low values (about 1%) in the far-burner region, presumably due to the fact that soot oxidation is about to cease in that location, at the critical smoke point.

At the 30% critical smoke-point flow rate, the peak concentrations of both NO and CO are smaller (note the change in the scale of the ordinate in Figures 10 and 11). This again confirms the effects of the diluent. Even at the smoke point the concentrations of both CO and NO reduce, which again is in conformity with the results of nonsmoking propane flames (Gollahalli, 1977).
Figure 12 shows the emission indices of NO, NO\textsubscript{x}, and CO expressed in the units of g/kg of fuel in propylene flames at various fractions of the critical smoke-point flow rate. The emission indices are calculated following Turns (1996) using the concentrations of CO\textsubscript{2} measured in the exhaust. Although the assumption of complete combustion is somewhat questionable in the smoking flame, it is still reasonable here, at least for comparative discussions, in view of the fact that the flames were at the smoke point, almost at the verge of becoming nonsmoking.

It is interesting to note that the NO and NO\textsubscript{x} emissions decrease slightly with the flow rate increase towards the critical value. This low rate of decrease is

![Figure 10: Radial NO concentration for 100% CFMFR at three different axial locations.](image_url)
due to the mutually compensating effects of reduction in nitrogen and the increase of heat release level as the fuel flow is increased. Nevertheless, all flames were at the threshold of smoke point, and hence it is logical to expect some similarity in thermo-chemical processes in all these flames, which further makes the emission indices less sensitive to fuel flow rate. One interesting factor appears to be in the CO emission index, which clearly shows a jump at a flow rate of 0.018 kg/min. This jump could perhaps be explained by the critical change in flame turbulence levels, which entrains more air into the flame, and thus results in a higher oxidation rate of soot.

FIGURE 11 Radial NO concentration for 30% CFMFR at three different axial locations.
CONCLUSIONS

A small co-flow velocity does not change the smoke point results obtained at quiescent conditions. The smoke point curves (critical fuel mass flow rate versus nitrogen mass flow rate required to suppress smoking) exhibited a skewed bell shape. The two regions on either side of the maximum nitrogen flow rate are controlled by different mechanisms related to chemical processes and fluid dynamics.

The radiation power varied rapidly with the critical fuel mass flow rate. The variation of radiative fraction of heat release rate as a function of the critical fuel mass flow rate showed an approximately linear relation. The value of this fraction for propylene was 0.39 and was in the range of values obtained from the correlation based on laminar smoke point of the fuel. The axial radiation power profiles of the flames showed a low radiation in both the near-burner and far-burner regions, but a high peak value in the mid-flame region. Flame temperature profiles and soot concentration profiles corroborate the radiation data. The far-burner temperature at the critical smoke point was approximately 1475 K and agreed with the data in the literature. The peak concentrations of NO, NOx, and CO decreased when the fuel flow rate was decreased from its critical smoke-point value to 30% of it. The emission indices of NO and NOx decreased gradually with the increase in the critical smoke point flow rate; the CO emission index, however, increased.

FIGURE 12  Propylene flame emission indices for NO, NOx, and CO at different critical fuel mass flow rates.
Acknowledgments
The authors acknowledge the financial support of the Oklahoma Center for Advancement of Science and Technology, and John Zink Company with Mr. Lawrence Berg as the contract monitor.

References