Appendix C:

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HYDRODYNAMIC SUPPRESSION OF SOOT FORMATION IN LAMINAR COFLOWING JET DIFFUSION FLAMES

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Effects of flow (hydrodynamic) properties on limiting conditions for soot-free laminar non-premixed hydrocarbon/air flames (called laminar soot-point conditions) were studied, emphasizing non-buoyant laminar coflowing jet diffusion flames. Effects of air/fuel-stream velocity ratios were of particular interest; therefore, the experiments were carried out at reduced pressures to minimize effects of flow acceleration due to the intrusion of buoyancy. Test conditions included reactant temperatures of 300 K; ambient pressures of 3.7–49 8 kPa; methane-, acetylene-, ethylene-, propane-, and methane-fueled flames burning in coflowing air with fuel-port diameters of 1.7, 3.2, and 6.4 mm, fuel jet Reynolds numbers of 18–121; air coflow velocities of 0–6 m/s; and air/fuel-stream velocity ratios of 0.003–70. Measurements included laminar soot-point flame lengths, laminar soot-point fuel flow rates, and laminar liftoff conditions. The measurements show that laminar soot-point flame lengths and fuel flow rates can be increased, broadening the range of fuel flow rates where the flames remain soot free, by increasing air/fuel-stream velocity ratios. The mechanism of this effect involves the magnitude and direction of flow velocities relative to the flame sheet where increased air/fuel-stream velocity ratios cause progressive reduction of flame residence times in the fuel-rich soot-formation region. The range of soot-free conditions is limited by both liftoff, particularly at low pressures, and the intrusion of effects of buoyancy on effective air/fuel-stream velocity ratios, particularly at high pressures. Effective correlations of laminar soot- and smoke-point flame lengths were also found in terms of a corrected fuel flow rate parameter, based on simplified analysis of laminar jet diffusion flame structure. The results show that laminar smoke-point flame lengths in coflowing air environments are roughly twice as long as soot-free (blue) flames under comparable conditions due to the presence of luminous soot particles under fuel-lean conditions when smoke-point conditions are approached. This is very similar to earlier findings concerning differences between laminar smoke- and soot-point flame lengths in still environments.

Introduction

Motivated by technological and public health problems, several methods have been developed to control the soot content and emissions of hydrocarbon-fueled flames. Among these, soot-control methods based on fast mixing for non-premixed (diffusion) flames are of interest because they avoid the operational problems of additives and premixed combustion [1–3]. The objective of fast mixing is to minimize residence times of fuel and fuel-decomposition products at fuel-rich conditions so that few soot particles develop and they can be readily consumed in the soot-oxidation regions of the flame. The present investigation seeks improved understanding of fast mixing concepts based on experimental observations of laminar coflowing jet diffusion flames. Laminar diffusion flames were studied because they provide relatively tractable models of mixing and reaction within more practical but relatively intractable turbulent diffusion flames. Another advantage of the laminar coflowing jet diffusion flame configuration is that it has been widely used to study the soot-formation properties of diffusion flames (see Refs. [4–8]).

While fast mixing reduces soot formation within diffusion flames, past studies of both laminar opposed and coflowing jet diffusion flames show that the way that mixing is carried out is important as well [9–17]. In fact, existing evidence from both laminar and turbulent jet diffusion flames, and from empirical industrial practice, suggests that soot reductions can be achieved most effectively by ensuring that velocities normal to the flame sheet are directed from the fuel-rich toward the fuel-lean side. This configuration, called "soot-formation-oxidation flame conditions" by Kang et al. [13], tends to reduce the residence times of soot precursors and soot at fuel-rich soot-formation conditions by drawing these materials directly through the flame sheet toward fuel-lean oxidation conditions. In contrast, when velocities normal to the flame sheet are directed from the fuel-lean toward the fuel-rich side, called "soot-formation flame conditions" by Kang et al. [13], residence times of soot precursors and soot at fuel-rich soot-formation conditions are enhanced, making oxidation of these materials more problematic when oxidation conditions are finally reached.

Studies of effects of velocities normal to the flame sheet on soot formation have been carried out in laminar opposed and coflowing jet diffusion flames [9–17]. During most of these studies [9–15], velocities normal to the flame sheet were varied by varying the compositions of the oxidant- and fuel-carrying streams. For example, diluting the fuel stream with an inert gas (e.g., nitrogen) while enriching the oxidant stream by removing existing diluent (e.g., removing nitrogen from air) promotes increased velocities normal to the flame sheet directed from the fuel-rich toward the fuel-lean side and yields reduced soot concentrations in the flame [9–14]. As pointed out by Sunderland et al. [12], however, these composition changes alone are sufficient to retard soot formation and enhance soot oxidation, which tends to reduce soot concentrations, obscuring the effect of hydrodynamics on soot control. In addition, the practical utility of varying reactant-stream compositions to control soot formation in diffusion flames is relatively limited.

The present investigation sought a direct evaluation of effects of velocities normal to the flame sheet on soot formation in diffusion flames by considering pure air and fuel reactant streams for laminar coflowing jet diffusion flames. In this configuration, enhanced (retarded) airstream velocities provide entrainment velocities normal to the flame sheet directed from the fuel-rich (fuel-lean) to the fuel-lean (fuel-rich) sides of the flame, which should reduce (increase) both soot concentrations within the flame and the tendency to emit soot from the flame. This behavior has been observed, with enhanced airstream velocities yielding significant increases of laminar smoke-point flame lengths—particularly for low-pressure flames, in which disturbances of the velocity field due to the intrusion of effects of buoyancy become relatively small [16]. Recent numerical simulations from Kaplan and Kailasanath [17] exhibit similar tendencies for soot concentrations within laminar coflowing jet diffusion flames to decrease for locally enhanced airstream velocities. Finally, air atomization, which is widely used for soot control in aircraft gas turbine combustors, corresponds to an enhanced airstream velocity flame configuration, which may explain this soot-control mechanism.

Prompted by these observations, the present investigation considered effects of enhanced airstream velocities on laminar soot-point properties—that is, the condition where soot is first observed in laminar diffusion flames. The main issue was to learn whether gas-phase processes (dominated by both diffusive and convective transport) could be controlled to yield soot-free flames by manipulating air/fuel velocity ratios in the same way that gas/solid processes (dominated by convective transport alone) can be controlled to eliminate soot emissions. Associated flame properties such as luminous flame lengths and flame liftoff conditions were also observed. Finally, present results define conditions where detailed numerical simulations of flame structure can be evaluated without the complications associated with soot chemistry [18–20].

Experimental Methods

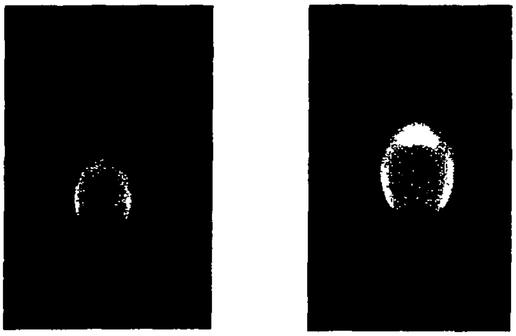
Measurements were carried out at subatmospheric pressures to control the effects of buoyancy [21]. The test burner was a vertical coaxial tube arrangement with the fuel flowing from an inner port with inside diameters of 1.7, 3.2, and 6.4 mm and the air flowing from an outer port with an inside diameter of 60 mm. The air passage used beads and screens to provide a uniform velocity distribution at the burner exit; the fuel passage provided fully developed laminar flow at its exit. The exit of the fuel port was 10 mm above the exit of the air port to provide an undisturbed region for flame attachment. The air-port diameter was sufficiently large so that the mixing layer between the air coflow and the ambient air in the vacuum chamber did not disturb the flame. The burner was operated within a windowed vacuum chamber with an inside diameter and length of 300 and 1200 mm, respectively.

Acetylene-, ethylene-, propane-, and methane-fueled laminar jet diffusion flames in coflowing air were considered with gas purities in excess of 99%, except for acetylene, which had a purity of only 98% due to contamination by the acetone that is present in commercial acetylene cylinders for safety purposes. Past work has shown, however, that effects of acetone contamination of acetylene on luminous flame shapes and laminar smoke-point flame lengths are small compared with experimental uncertainties [16]. In addition to the variations of burner-port diameters and fuels mentioned earlier, test conditions included reactant temperatures of roughly 100 K; ambient pressures of 3.7–49.8 kPa; fuel jet exit Reynolds numbers, *Re*, of 18–121; air coflow velocities of 0–6 m/s; and air/fuel-stream velocity ratios of 0.003–70. Transition to turbulent flames was never observed during the present experiments, whereas characteristic flame residence times were small so that effects of radiative heat losses from the flames were negligible [8,22].

Results and Discussion

Flame Appearance

Photographs of typical soot-free (blue) and soot-containing ethylene/air flames at identical fuel-port



(a) (b)

FIG. 1. Photographs of ethylene/air diffusion flames for a fixed burner diameter (3.2 mm), pressure (10.2 kPa), and fuel flow rate (1.3 mg/s): left image at the laminar soot point at the largest possible air/fuel-stream velocity ratio, u_a/u_f = 0.2, at this condition; right image for a soot-containing flame at a relatively small air/fuel-stream velocity ratio, u_a/u_f = 0.004, at this condition.

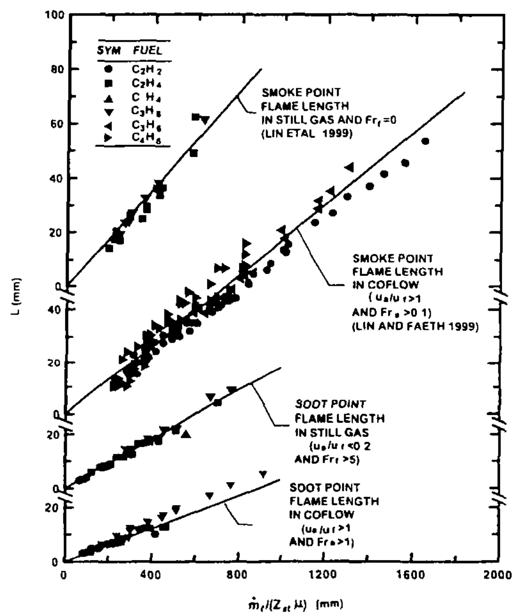


FIG. 2. Correlations between laminar soot- and smoke-point flame lengths and corrected fuel flow rates for coflowing laminar jet diffusion flames fueled with acetylene, ethylene, methane, propane, propylene, and 1-3-butadiene and burning in air based on the simplified flame shape analysis of Lin et al. [22] and Lin and Faeth [23]. Laminar smoke-point flame length correlations also are from Refs. [22] and [23].

exit conditions are illustrated in Fig. 1. Effects of buoyancy are relatively small at this low-pressure condition (10.2 kPa), so that flame properties approximate the non-buoyant behavior of greatest interest for practical applications. The flame on the left is at its laminar soot-point condition at the largest air/fuel-stream velocity ratio, u_a/u_f = 0.2, that could be used without liftoff at this jet exit condition. The flame on the right illustrates the effect of reducing the air/fuel-stream velocity ratio from the soot-point condition to a relatively small value, u_a/u_f = 0.004, while keeping all other flame properties the same. The reduced entrainment from the airstream at small u_a/u_f increases flame residence times at conditions where soot formation is favored, which causes soot to appear, as evidenced by a region of yellow flame luminosity near the flame tip.

Flame Length Correlations

Similar to the observations of luminous flame lengths at laminar smoke points by Schug et al. [5] and Lin and Faeth [14], the present luminous flame lengths at laminar soot points were closely associated with the fuel flow rate. Measurements establishing this behavior and a brief discussion of a simplified theory that helps explain the experimental findings are considered in the following.

Laminar soot- and smoke-point luminous flame lengths are plotted in Fig. 2 as a function of a corrected fuel flow rate suggested by simplified theories of flame shapes for non-buoyant laminar jet diffusion flames in still and coflowing gases [22,23] developed by extending earlier analyses [24–26]. The laminar soot-point measurement conditions from the present investigation were summarized earlier. The measured laminar smoke-point correlations are from Lin and Faeth [14] for acetylene-, propylene-, and 1-3-butadiene-fueled flames burning in air at pressures of 19–51 kPa, a burner diameter of 6 mm, and air/fuel-stream velocity ratios of 0.4–6.7. Two sets of correlations (each) are illustrated for the laminar soot- and smoke-point luminous flame lengths in Fig. 2: one for small u_a/u_f based on analysis of laminar jet diffusion flames in still air [22] and one for large u_a/u_f based on analysis of laminar jet diffusion flames in coflowing air [23]. There are good correlations between measured luminous flame lengths and the corrected fuel flow rates for both laminar soot- and smoke-point conditions (see Ref. [23] for the latter). As a result, laminar soot-point properties are represented by the laminar soot-point fuel flow rate in the following, similar to past work [14]. It is also evident that the correlation for laminar smoke-point flame lengths is roughly twice as long as that for laminar soot-point flame lengths at both large and small u_a/u_f limits.

An explanation of the flame length behavior observed in Fig. 2 can be obtained from the flame

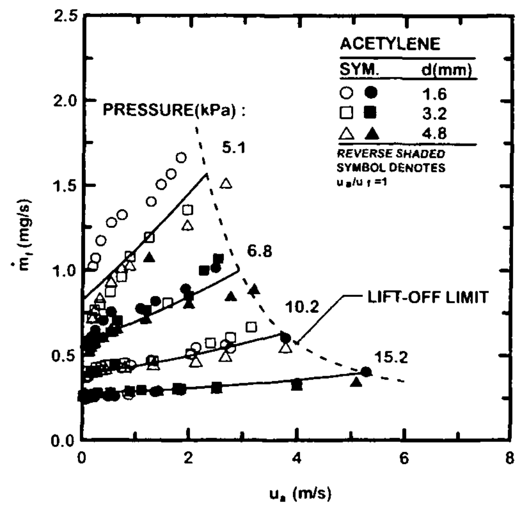


FIG. 3 Fuel flow rates at laminar soot-point and liftoff conditions as a function of air coflow velocities, fuel-port diameter, and pressure for acetylene/air flames.

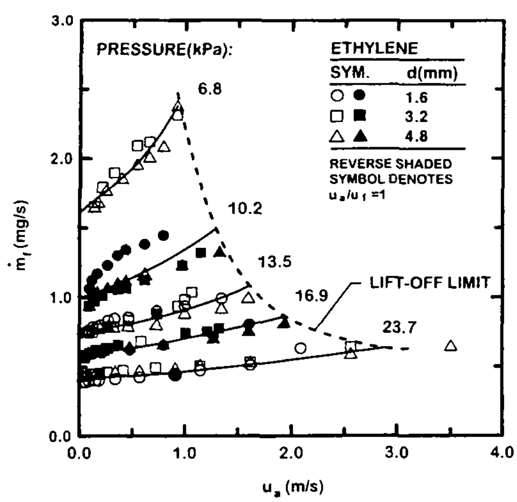


FIG 4. Fuel flow rates at laminar soot-point and liftoff conditions as a function of air coflow velocities, fuel-port diameter, and pressure for ethylene/air flames.

shape correlations based on the simplified analyses of Refs. [22] and [23]. Ignoring small effects of virtual origins, both these correlations can be written to yield the luminous flame length as a function of the corrected flow rate parameter used in Fig. 2, as follows:

$$L = (C_n C_f Sc/(8\pi))\ \dot{m}_f/(Z_{st}\mu) \qquad (1)$$

Following Refs. [22] and [23], a simple correlation of equation 1 was fitted to measurements of flames in air environments by using values of the Schmidt number and viscosity for air at the average of the adiabatic flame temperature and the ambient temperature. Similarly, $C_n = 3$ for non-buoyant flames in still gases, whereas $C_n = 2$ for non-buoyant flames in coflowing gases [23]. The measurements of Refs. [27] and [28] yield $C_f \approx 0.5$ for soot-free blue flames and $C_f \approx 1.0$ for flames at the laminar smoke point for flames in still air [22]. These assignments provide the good correlations of the present results in coflowing air seen in Fig. 2, as well as an explanation of the increased luminous flame lengths caused by reduced air coflow velocities and the presence of soot near the flame tip for these conditions seen in Fig. 1.

Laminar Soot-Point Properties

Both laminar soot-point and liftoff properties were measured during the present experiments. The tests were conducted by varying the pressure range for each fuel based on its propensity to soot, so that effects of reasonable variations of air/fuel-stream velocity ratios could be measured for flames fueled with each fuel in spite of limitations due to effects of liftoff and the intrusion of buoyancy.

In the following, effects of air coflow on laminar soot-point and liftoff properties are presented as plots of laminar soot-point fuel flow rates as a function of air coflow velocities because this approach provides a compact presentation of the measurements. Effects of air coflow velocities on laminar soot-point fuel flow rates were qualitatively similar for the four fuels that were considered. This can be seen from the plots of fuel mass flow rate at soot-point conditions as a function of air coflow velocities for the various pressures and fuel-port diameters that are illustrated in Figs. 3–6. To indicate the transition between soot-formation and soot-formation-oxidation configurations at the base of the test flames, the condition of $u_a/u_f = 1$ is denoted by reverse-shaded symbols on the plots (note that the soot-formation and soot-formation-oxidation configurations occur for test conditions in the left and right of the reverse-shaded symbols, respectively). Liftoff conditions are denoted by the symbol at the highest air flow rate for each pressure and fuel-port diameter, with the extreme liftoff limit denoted by a dashed line.

The measurements illustrated in Figs. 3–6 show that increased air coflow velocities increase laminar soot-point fuel rates. Notably, this behavior is observed for air/fuel-stream velocity ratios both smaller and larger than unity. Increasing pressures generally reduce allowable fuel mass flow rates and flame lengths for soot-free flames due to increased soot-formation rates and flame residence times for a given flame length. The relative enhancement of laminar soot-point fuel flow rates between small and

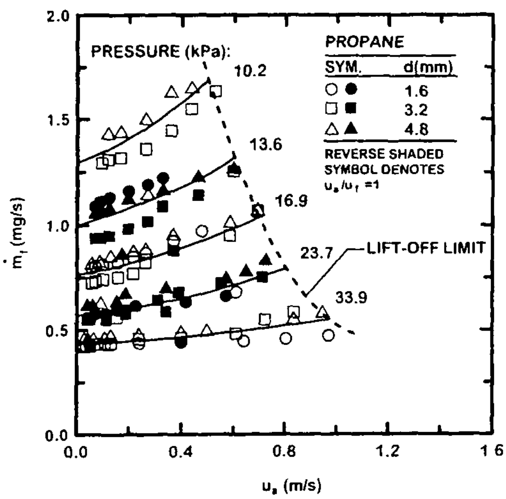


FIG. 5. Fuel flow rates at laminar soot-point and liftoff conditions as a function of air coflow velocities, fuel-port diameter, and pressure for propane/air flames.

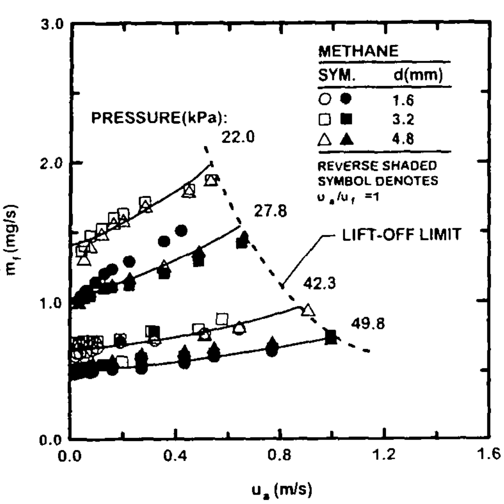


FIG 6. Fuel flow rates at laminar soot-point and liftoff conditions as a function of air coflow velocities, fuel-port diameter, and pressure for methane/air flames.

maximum allowable values of air coflow velocities before liftoff, however, tends to be relatively independent of the pressure for a particular fuel. This behavior comes about because generally more intense reaction rates at elevated pressures accommodate large air coflow velocities before liftoff, which tends to compensate for faster soot reaction rates at elevated pressures. Taken together, it is clear that sufficiently large air coflow velocities are capable of completely suppressing the formation of particulate soot for these conditions, supporting the soot-suppression argument discussed in the introduction. The resulting soot-free flames also provide potentially useful conditions for evaluating detailed models of diffusion flame chemistry and transport at the computationally tractable limit of soot-free laminar diffusion flames for light hydrocarbons.

For the present tests, the propensity of a fuel to soot can be associated with the pressure range for observing soot-free flames. On this basis, the present tests indicate that the propensity to form and emit soot progressively decreases in the order acetylene, ethylene, propane, and methane. This finding agrees with conventional determinations of laminar smoke-point properties based on observations of buoyant laminar jet diffusion flames [4–8]. In addition, the general behavior of the laminar soot-point properties in Figs. 3–6 is qualitatively similar to earlier observations of laminar smoke-point properties as a function of air coflow velocities in Ref. [16].

An important issue concerning the results illustrated in Figs. 3–6 is the mechanism for increased resistance to soot formation as the air coflow velocity increases for a particular fuel, fuel-port diameter, and pressure. Consider the simplest case, when the flame is in the soot-formation-oxidation condition for air/fuel-stream velocity ratios greater than unity, which generally involves conditions in which buoyancy does not significantly affect flame velocities. The results discussed in connection with Fig. 2 then indicate that the flame shape (length) is largely controlled by the fuel flow rate and is relatively independent of fuel velocity at the burner exit (or the burner-port diameter). In contrast, the characteristic flame residence time, t_a, is proportional to the flame length divided by the air coflow velocity [23]. Thus, given a critical residence time for the appearance of soot for a particular fuel and pressure, the fuel flow rate at the laminar soot-point limit progressively increases with increasing air coflow velocity, relatively independent of fuel-port diameter, which is typical of the behavior seen in Figs. 3–6 for reasonably large air/fuel-stream velocity ratios.

The mechanism of increased resistance to soot formation as the air coflow velocity increases for a particular fuel, fuel-port diameter, and pressure is more complex when the flame is in the soot-formation configuration (at least near the flame base). This generally involves conditions in which buoyancy affects flame velocities and air/fuel-stream velocity ratios are less than unity. For such conditions, increasing the air coflow velocity causes the flame to shift from the soot-formation toward the soot-formation-oxidation configuration, which reduces the proportion of the flame residence time spent at soot-formation conditions compared with soot-oxidation conditions and thus tendencies for soot formation. Behavior of this nature can be observed from the soot-concentration measurements near laminar smoke-point conditions in Ref. [16], where variations of soot concentrations as a function of residence time become path independent as the soot-formation-oxidation condition is approached. Similarly, this effect is not uniform for all soot precursor paths through the present flames, whereas all paths are affected to some extent by reduced flame residence times as air coflow velocities are increased. These effects, and the intrusion of buoyancy, introduce greater effects of fuel-port diameter on laminar soot-point conditions for these flames for the simple soot-formation-oxidation flame configuration discussed earlier, as seen in Figs. 3–6. Nevertheless, in spite of variations of flame behavior depending on the range of air/fuel-stream velocity ratios and effects of the intrusion of buoyancy, the general capability of increased air coflow velocities to reduce the content and emissions of soot for the present flames is evident.

Flame Stability Properties

Limiting conditions for flame liftoff are plotted in Figs. 3–6 as a function of pressure for each fuel. At high pressures, fuel-port velocities are small at liftoff conditions, and this limit correlates quite nicely as a function of coflow velocity and pressure, relatively independent of fuel-port diameter. At low pressures, however, fuel-port velocities become relatively large and also begin to affect liftoff conditions, with small fuel-port diameters (which yield the largest fuel-port velocities) generally contributing to reduced flame stability.

Conclusions

The present experimental investigation considered the effect of air/fuel-stream velocity ratios on soot processes within laminar coflowing jet diffusion flames for the experimental conditions summarized earlier. Major conclusions of the study are as follows:

1. Laminar soot-point flame lengths and fuel flow rates were increased with increasing air/fuel-stream velocity ratios; these effects were most pronounced at low pressures, where effects of buoyancy were minimized, and initial air/fuel-stream velocity ratios are reasonably representative of the entire visible portion of the flame for the present test conditions. These results are qualitatively similar to earlier measurements of laminar smoke-point properties, as well as recent predictions of soot-concentration properties [17], for similar flame conditions.
2. Laminar soot-point flame lengths were conveniently correlated in terms of a corrected fuel flow rate parameter based on an earlier simplified analysis of the structure of non-buoyant laminar coflowing jet diffusion flames [23]. It was found that laminar smoke-point flame lengths in both coflowing and still air environments are roughly twice as long as soot-free (blue) flames under comparable conditions due to the presence of luminous soot particles under fuel-lean conditions as laminar smoke-point conditions are approached.
3. The mechanism of increased resistance to soot formation with increasing air/fuel-stream velocity ratios at low pressures (where buoyancy does not significantly affect flame velocities) and large air/fuel-stream velocity ratios (where the flame is in the soot-formation-oxidation configuration) involves progressive reduction of flame residence times for soot production, eventually reaching the soot-free (blue) flame limit. Given a critical residence time for the appearance of soot for a particular fuel and pressure, this behavior is consistent with present measurements and the simplified analysis of the shape of non-buoyant laminar jet diffusion flames in coflowing air [23]. Notably, the shape (length) of these flames is largely controlled by the fuel flow rate, while the characteristic residence time is proportional to the flame length divided by the air coflow velocity. Then, laminar soot-point fuel flow rates should increase with increasing air coflow velocities for a given fuel and pressure, relatively independent of fuel-port diameter, as observed at low pressures and large air coflow velocities in Figs. 3–6.
4. The mechanism of increased resistance to soot formation with increasing air/fuel-stream velocity ratios is more complex at high pressures (where buoyancy significantly affects flame velocities) and at small air/fuel-stream velocity ratios (where the flame is in the soot-formation configuration). Then, increasing air/fuel-stream velocity ratios causes the flame to shift from the soot-formation toward the soot-formation-oxidation configuration, which reduces the proportion of the flame residence time spent at soot-formation conditions compared with soot-oxidation conditions, reducing tendencies for soot formation accordingly. However, this effect is not uniform for all soot precursor paths through the flame, whereas all paths are affected to some degree by reduced flame residence times with increasing air/fuel-stream velocity ratios, as discussed in conclusion 3 above.

Other effects observed during the present investigation generally are consistent with earlier findings concerning the propensity of diffusion flames to form and emit soot [7–8]: laminar soot-point fuel flow rates and flame lengths tend to progressively increase with decreasing pressure, and the propensity to form and emit soot with variations of fuel type progressively decreases in the order acetylene, ethylene, propane, and methane. Finally, in spite of limitations due to the intrusion of buoyancy, the results of the present investigation support the earlier findings of Ref. [16] that effects of enhanced air/fuel-stream velocity ratios contribute to the mechanism of reduced sooting tendencies for non-premixed flames using air atomization techniques. Nevertheless, more work is needed to resolve the specific contributions of enhanced air/fuel-stream velocity ratios and improved atomization to reducing the sooting tendencies of practical spray flames.

Nomenclature

C_f	flame length empirical parameter
C_n	flame length configuration parameter
d	fuel-port diameter
D	mass diffusivity
Fr_a, Fr_f	air- and fuel-stream Froude numbers, $(u_a^2$ or $u_s^2)/(2gL)$
g	acceleration of gravity
L	laminar smoke- and soot-point flame lengths
$\dot{m}_f$	fuel mass flow rate
p	pressure
Re	Reynolds number, $4\,\dot{m}/(\pi d\mu)$
Sc	Schmidt number, ν/D
t_t	characteristic residence time, L/u_a
u	streamwise velocity
Z_{st}	stoichiometric mixture fraction
μ	dynamic viscosity
ν	kinematic viscosity

Subscripts

a	initial property of airstream
f	initial property of fuel stream

Acknowledgments

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COMMENTS

C. H. Priddin, Rolls Royce, UK. In the 80s-style fuel atomizers you showed, the overall AFRs are of the order 4–6, that is, still overall rich. Do you think your analysis still applies in this situation, or is the flame somewhere else?

Author's Reply The general success of air atomization to reduce soot emissions from aircraft gas turbine engines for a variety of fuel atomizer AFRs [Ref. 1 in paper] suggests that effects of increasing air/fuel velocity ratios persist even when AFRs are small. We believe that this is reasonable based on present findings because small fuel stream velocities should generally provide conditions where air stream velocities are larger than fuel stream velocities throughout the combustion process, leading to generally desirable soot emissions properties, e.g., soot-formation-oxidation conditions as defined by Kang [Ref. 13 in paper]. Direct demonstration of this conjecture, however, would be desirable.

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Cary Presser, NIST, USA. Please describe your thoughts regarding the use of different gases in place of air. Is the propensity to soot purely an aerodynamic effect (and thus other gases may be used) or is the pressure of oxygen required to assist in the oxidation of soot? It is assumed that ambient (or secondary) air is present to sustain a stable flame.

Author's Reply For the same reasons discussed in the reply to C. H. Pridden, we believe that the nature of the atomizing gas used in the fuel atomizer is not the most critical aspect of soot control using air atomization. It seems to us that the crucial elements are relatively good atomization with relatively small fuel momentum (velocities). This should generally yield desirable air/fuel stream velocity ratio properties when the region of the flame sheet is approached, e.g., soot-formation-oxidation conditions as defined by Kang (Ref. [13] in paper). Direct assessment of the conjecture, however, would also be desirable.