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Control of the structure and sooting characteristics of a coflow laminar methane/air diffusion flame using a central air jet: An experimental and numerical study

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Abstract

Multi-port co-annular burners are widely used in practice to achieve low NO_x and soot emissions from combustion devices. However, there is lack of fundamental studies on the structure and flame regime under different flow conditions. A conventional laminar axisymmetric coflow diffusion flame burner was modified by introducing a central air jet inside the fuel tube to investigate how the central air jet velocity affects the structure and sooting characteristics of a coflow methane/air diffusion flame. The modified burner produces a double flame structure: an inner inverse diffusion flame or an inner partially premixed flame and an outer normal diffusion flame. Experiments were conducted to observe the effect of the central air jet velocity on the appearance of the flame. At a given and relatively low central air jet flow rate the inner flame can be either a partially premixed one or an inverse diffusion one, depending on how the central air jet flow rate is adjusted. The overall flame structure and sooting characteristics can be controlled effectively by varying the flow rate of the central air jet. Detailed numerical calculations were conducted using GRI-Mech 3.0 without the NO_x chemistry and a simplified soot model. Numerical results reproduce the experimental observations and provide detailed information on the flow field, temperature, and species concentration distributions. The central air jet is an effective aerodynamic means to control the flame size, structure, and sooting characteristics.

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Keywords: Laminar diffusion flame; Soot formation; Multi-port burner; Numerical simulation

1. Introduction

Co-annular multi-port burners have been used in many industrial applications to improve com-

bustion efficiency and reduce pollutant (such as NO_x and soot) emissions. These burners offer the flexibility to aerodynamically control the flame shape, combustion process, and pollutant emissions by varying the velocity and compositions of individual fluid streams to achieve the optimal performance. Several experimental studies have been carried out to investigate the effects of burner design on radiation heat output, flame

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appearance, NO_x emission, and/or CO and unburned hydrocarbon emissions [1–4]. It is worth pointing out that the key features in these novel burner designs are to take advantage of the properties of inverse diffusion flame by introducing an air stream inside the fuel jet, the properties of partially premixed flame by mixing the fuel stream with air or combustion products, and the stability of normal diffusion flames. However, the flames investigated in most of such studies are large in size and turbulent; therefore, they do not permit multidimensional numerical simulations using detailed combustion chemistry to gain fundamental understanding of the effect of flow field on the combustion processes and interactions between multiple reaction zones. Coaxial multi-port burners have also been widely employed for flame synthesis of particles, see Ref. [5] and references cited therein.

Chao and Axelbaum [5] studied the flame height and flame shape of a diffusion flame stabilized on a triaxial burner theoretically using the Burke–Schumann methodology. Their analysis, however, was limited to the case where the velocities of three streams are equal. They demonstrated the application of the theory for flame synthesis of aluminum and aluminum nitride. Ko et al. [6] extended the theoretical study of Chao and Axelbaum [5] to a more generic situation where the compositions and velocities of the three streams can be different. The theory used in Ref. [6] to predict the flame height and shape is again based on the fast chemistry (flame-sheet approximation), i.e., following the methodology of Burke and Schumann, with various assumptions such as unity Lewis number, neglect of buoyancy, and radiation heat transfer. They also conducted an experimental study to investigate the influence of stream concentrations and velocities on the shape of a laminar double diffusion flame (an inverse diffusion flame embedded inside a normal diffusion one) and conducted numerical calculations of the flame height and flame shape based on their Burke–Schumann analysis.

The flame structure and overall combustion processes in co-annular multi-port burners can be very complicated. However, they can be in general viewed as interactions of three basic flame modes: normal diffusion flame, inverse diffusion flame, and partially premixed flame. These three fundamental flame modes have been extensively investigated in terms of their structure, stability, and soot formation characteristics. The structure and soot formation characteristics of coflow laminar normal diffusion flames (NDF) (fuel jet is issued into air) have been experimentally investigated by many researchers, e.g., Mitchell et al. [7], Santoro and co-workers [8], Faeth and co-workers [9], among many others, and numerically studied using relatively detailed chemistry by Kaplan and Kailas-anath [10], Smooke et al. [11,12], and Guo et al. [13], among others. Coflow inverse diffusion flames

(IDF) have also been investigated experimentally by various researchers [14–19]. It has been made clear that IDFs exhibit remarkably different soot formation features from those of NDFs due to the very different temperature, chemical environment, and residence time experienced by inception soot particles [10]. As a consequence of these differences, NDFs have a much higher soot loading than IDFs and do not emit soot when operated below smoke point. On the other hand, IDFs emit soot. Several studies have shown that the structure of IDF can be predicted by theories developed for NDF [14,18]. This is because mathematically the NDF and IDF are indistinguishable and the only difference is that fuel and air are interchanged. However, there have been very few numerical studies on coflow IDF using relatively detailed reaction mechanism [10,20].

It is interesting to observe that there have been numerous experimental and numerical studies on coflow laminar partially premixed flames (PPF), see Refs. [21–25] and the references cited therein, mainly due to its greater relevance to many practical combustion processes including non-premixed combustion. One particularly interesting feature of partially premixed flames is the occurrence of double flame structure: an inner rich premixed flame enveloped by an outer diffusion flame [22–24]. The two flames are known to be synergistically coupled [22].

The three fundamental flame modes, i.e., NDF, IDF, and PPF, encountered in two-stream co-annular burner configuration have been extensively investigated as discussed above. A laminar diffusion flame stabilized in a co-annular multi-port (more than two streams) burner is expected to possess some rich phenomena as a result of the highly non-linear interactions among reaction zones formed between adjacent streams as demonstrated by Ko et al. [6], who experimentally observed the history effect of the flame appearance, depending on increasing or decreasing the central jet velocity. Unfortunately, detailed experimental or numerical studies addressing such interactions are currently lacking. Therefore, the objective of this study is to numerically investigate the structure and sooting characteristics of a double methane–air diffusion flame established with a triaxial coflow burner (a central air jet, a co-annular fuel jet, and an outer co-annular air jet), which can be viewed as a model problem closely related to co-annular multi-port industrial gas-fired burners. Numerical calculations were conducted using GRI-Mech 3.0 without the NO_x chemistry, a semi-empirical soot formation model, and a non-gray gas radiation model to gain insights into the coupling between the inner and outer reaction zones. Additionally, experimental work was also conducted to observe the effect of varying the central air jet flow rate on the flame appearance and sooting characteristics.

Fig. 1. Top view of the double diffusion flame burner.

2. Experimental setup

The burner employed in this study is a modified version of the standard NRC co-annular laminar diffusion flame burner used in previous studies, e.g., Ref. [26]. A top view of the burner is shown in Fig. 1. It consists of a central air jet of 6.55 mm i.d. and 7.94 mm o.d., a coaxial fuel pipe of 10.95 mm i.d. and 12.75 mm o.d., and another coaxial outer air jet of 88 mm i.d. and 100 mm o.d. Before exiting their respective nozzles, both air streams and the fuel stream pass through a porous metal disk to achieve uniform flows and suppress flow disturbances. Besides the introduction of the central air jet, this burner is identical to that in the previous study [26]. It is noticed that the central air stream and the fuel stream are delivered in two separate tubes in this study. This makes the present setup different from those used in the studies of PPFs [22–25], but similar to that used by Ko et al. [6].

The flow rates of all three streams were controlled using mass flow controllers (Aalborg). For the purposes of this study, the flow rates of the fuel and the outer air stream were maintained constant (388 cm³/min for CH₄ and 284 l/min for the outer air stream) while the central air jet flow rate was varied. The effect of the central air jet flow rate on the flame appearance was recorded using a digital camera.

3. Numerical model

Descriptions of the overall flame model (governing equations, reaction mechanism, soot formation model, non-gray gas radiation model, and numerical method) have been given in previous publications [27,28]. Only a brief summary of the numerical model is given below.

The steady-state governing equations of mass, momentum, energy, and species in axisymmetric cylindrical coordinates and in the low Mach number limit were solved. The effect of buoyancy was accounted for by retaining the gravity term in the momentum equation in the flow direction (z , vertically upwards). The method of correction for the diffusion velocity described in Ref. [29] was employed to insure that the net diffusion flux of

all species (including the thermophoresis of soot) sums to zero in both r and z directions. The interaction between the soot chemistry and the gas-phase chemistry was accounted for through the reaction rates of the species related to soot formation and oxidation. Only the thermal diffusion velocities of H₂ and H are accounted for using the expression given in Ref. [29]. The source term in the energy equation due to radiation heat transfer was also included and calculated using the discrete-ordinates method along with the statistical narrow-band correlated- k method. Details can be found in Refs. [30,31].

A modified version of the semi-empirical two-equation formulation of soot kinetics proposed by Leung et al. [32] was used to model soot nucleation, growth, and oxidation. In this model the transport equations for the soot mass fraction and number density per unit mass are solved. These equations were given in our previous publications [27,28]. The source term in the transport equation for soot mass fraction accounts for the contributions of soot nucleation, surface growth, and oxidation. It is assumed that acetylene is the only soot nucleation and growth species and soot nucleation and surface growth proceed, respectively, via $C_2H_2 \rightarrow 2C(S) + H_2$ and $C_2H_2 + nC(S) \rightarrow (n+2)C(S) + H_2$. The rates of nucleation and surface growth are given as $R_1 = k_1(T)[C_2H_2]$ (kmol/m³/s) and $R_2 = k_2(T)A_s^{0.5}[C_2H_2]$ (kmol/m³/s), where $A_s = \pi(6/\pi)^{2/3} \rho_{C(S)}^{-2/3} \dot{V}_s^{2/3} \rho N^{1/3}$ is the soot surface area per unit volume and $[C_2H_2]$ is the mole concentration of acetylene. The nucleation and growth rate constants used in the present calculations were taken from a previous study [27]. The density of soot $\rho_{C(S)}$ is taken to be 1.9 g/cm³. It is noted that the soot surface growth rate is assumed here to be proportional to the square root of the soot surface area based on the recommendation of Leung et al. [32]. The rationale for such assumption was recently discussed by Liu et al. [28].

Gas-phase combustion chemistry was modeled using the GRI-Mech 3.0 mechanism [33], which was optimized for methane combustion, with the removal of species and reactions related to NO_x formation (except N₂). This simplified GRI-Mech 3.0 mechanism contains 36 species and 219 reactions. Thermal and transport properties of species and the mixture were obtained from CHEMKIN subroutines and the GRI-Mech 3.0 database. To identify the locations of the inner and outer flame sheets, where the mixture fraction is stoichiometric, the mixture fraction was calculated using the definition of Bilger et al. [34].

The transport equations for mass, momentum, energy, gas-phase species, soot mass fraction, soot number density, and radiation intensity are closed with the equation of state and appropriate boundary conditions on each side of the computational domain. These equations were discretized using the control volume method in axisymmetric

cylindrical coordinates. Further details on the numerical methods to solve the resultant discretized system of equations can be found in Refs. [27,28].

Numerical calculations were conducted on a computational domain of $9.79\text{ cm } (z) \times 5.28\text{ cm } (r)$. Both the fuel and the central air inlet temperatures were assumed to be 350 K, which is slightly higher than room temperature to account for the preheat effect. The outer co-annular air stream temperature was assumed to be 300 K. Non-uniform grids were used in both the r and z directions to provide greater resolution in the large gradient regions without an excessive increase in the computing time. Very fine and uniform grids were placed within the fuel pipe at $r = 5.45\text{ mm}$ in the radial direction with a grid size $< 0.13\text{ mm}$. Outside the burner tip in r direction, the grid size became gradually coarser. In the flow direction (z), very fine and non-uniform grids were used in the burner exit region up to 3.6 cm (grid size $< 0.35\text{ mm}$). Further downstream, uniform but coarser grids were used. The grid size between $z = 3.6\text{ cm}$ and 6.6 cm is still fairly fine at $\Delta z = 0.55\text{ mm}$. Beyond $z = 6.6\text{ cm}$, the grid size is $\Delta z = 0.7\text{ mm}$. The computational domain was divided into $251(z) \times 115(r)$ grid lines. Uniform velocity profile was assigned to both the fuel (15.47 cm/s at 350 K) and the central air inlets. For the outer co-annular air stream a uniform velocity of 77.9 cm/s was assigned outside the boundary layer formed at the outer surface of the fuel pipe. Inside this boundary layer a boundary layer type velocity profile was assumed. Along the centerline ($r = 0\text{ cm}$) and the outer boundary, $v = 0\text{ cm/s}$ and zero-gradient for all other variables are assumed. At the top boundary ($z = 9.79\text{ cm}$), a zero-gradient condition was applied to all variables.

4. Results and discussion

4.1. Increasing the central air jet flow rate

When the methane flame was ignited with the central air jet turned off, a stable NDF with a luminous flame height of 63 mm is generated as shown in Fig. 2a, which is identical to that established with the standard NRC burner under the same fuel and outer co-annular air flow rates. The NDF is very yellow-orange in color, characteristic of soot radiation. The variations of the visible flame appearance with increasing the central air jet flow rate for $R \leq 1.92$, captured by a digital camera of identical settings, are shown in Fig. 2. As the amount of air added through the central jet (the amount of air delivered through the central air nozzle is represented by the ratio of the central air jet mean velocity to that of the fuel jet $R = U_{\text{c,air}}/U_{\text{fuel}}$, where $U_{\text{fuel}} = 15.47\text{ cm/s}$)

was gradually increased, the color of the flame changed significantly from bright yellow-orange ($R = 0$, Fig. 2a) to a pale orange structure enveloped by blue ($R = 1$, Fig. 2b), then to almost pure light blue ($R \geq 1.5$, Fig. 2c and d) for $R < 1.92$, indicating the formation of soot is almost completely suppressed. The flame height also decreases significantly. It is evident from Fig. 2 that the IDF is not formed when $R \leq 1.92$ due to the absence of an ignition mechanism for the IDF and the central air delivered into the fuel stream is simply to mix with the fuel. As such the resultant flame is basically a partially premixed one. This gesture is supported by the remarkable similarity between the present methane flames and those of partially premixed methane flames reported by Gore and Zhan [21], who found that partial premixing reduces and ultimately eliminates soot particles in methane flames. Although partially premixed methane diffusion flames were also studied by Bennett et al. [23], they did not report how partial premixing affects the sooting behavior of laminar methane diffusion flames. However, they observed a double flame structure (an inner rich premixed flame and an outer diffusion one) when the level of premixing is high enough. It is evident from Fig. 2 that dilution (premixing) of CH_4 by air is a very effective way to suppress soot formation in CH_4 diffusion flames for relatively low central air jet flow rates.

At $R = 1.92$, it is interesting to notice that there appears a light blue spot in the flame centerline region at a height of about 2.3 cm above the burner exit, Fig. 2d, which is associated with the enhanced heat release rate as more air is delivered through the central tube into the fuel stream to create a combustible mixture closer to the outer diffusion flame front. With a slightly further increase in the central air jet flow rate beyond $R = 1.92$, the blue spot in the flame centerline region shown in Fig. 2d first becomes brighter and starts to move upstream slowly for a very short distance (about 1 mm) and then moves very rapidly (estimated to be on the order of ms) upstream to ignite the inner IDF, which is then anchored at the central air nozzle rim and the soot-free blue flame suddenly becomes a yellow-orange sooty flame again, see Fig. 3a for $R = 2$ below. It is believed that the increase in the strength and upstream motion of the blue spot are associated with the further enhancement of the heat release rate and ignition of the partially premixed mixture by the outer diffusion flame when the rich mixture of fuel and air in the centerline region becomes more and more combustible and close enough to the outer flame to reach a critical condition, where ignition of the rich mixture occurs, as the central air jet flow rate increases slightly beyond $R = 1.92$. Variation of the visible flame appearance after the onset of

Fig. 2. The visible flame appearance of the double CH_4 flame at low central air jet flow rates ($R \leq 1.92$) with increasing the central air jet flow rate from the normal diffusion flame mode. The corresponding equivalence ratios of the fuel and central air (as if they were fully mixed) are ∞ , 12.63, 8.42, and 6.59.

the inner IDF for $R > 1.92$ with the central air flow rate is displayed in Fig. 3. Once it is formed, the inner IDF remains anchored over a relatively narrow range of the central air jet flow rate of $R < 3$, Fig. 3a and b. The essentially soot-free

Fig. 3. The visible flame appearance of the double CH_4 flame at central air jet flow rates between $R = 2$ and 6 with the onset of the inner IDF. The corresponding equivalence ratios of fuel/central air (as if they were fully mixed) are 6.32, 5.05, 4.21, 3.61, 3.16, and 2.11.

structure of the inner blue-colored IDF is clearly visible. The sooting characteristics of the flame displays a remarkable difference before and after the onset of the inner IDF, compared Fig. 2d for $R = 1.92$ and Fig. 3a for $R = 2$. With increasing the central air jet flow rate the inner IDF becomes taller, while the visible flame height of the outer diffusion flame decreases significantly, Fig. 3a and b. When the central air jet flow rate reaches $R = 3$ and higher, the inner IDF starts to lift off and the height of the outer diffusion flame continues to decrease. As more air is added through the central air jet nozzle, soot formation is also greatly suppressed, as the luminous zone becomes shorter and fainter, and the flame becomes essentially soot-free at $R = 4$. This can be attributed to the dual actions of the central air: suppression of C_2H_2 formation, which reduces the soot surface growth rate, and direct oxidation of soot. It is also interesting to observe that the luminous sooting region always appears at about 8 mm above the burner exit, regardless the central air flow rate. Unlike the anchored IDFs at low central air flow rates, where the tip of the IDF is closed, the lifted IDFs at higher central air flow rates are open-tipped, Fig. 3d–f, and display a strong interaction with the outer NDF as indicated by the branching of the inner blue ring (the base of the IDF) and the outer blue shell, Fig. 3e and f. The lifted inner flame should be more adequately classified as a triple flame than an IDF.

Numerical results for the temperature distribution at $R = 0$, 1, and 2 for increasing the central air jet flow rate (from the case of $R = 0$) are shown in Fig. 4. The results for $R = 1$ and 2 were calculated with the converged model solution for $R = 0$ as the initial field. The white contours in Fig. 4 indicate the stoichiometric mixture fraction ($f_{\text{st}} = 0.05496$). It is evident that the flame height decreases, the peak flame temperature decreases, and the centerline temperature gradients increase with increasing the central air flow rate. The decreased flame height with increasing R is in agreement with the experimental observation. Although it was observed experimentally that the inner IDF is formed when R is slightly higher than 1.92, the numerical model predicts that the onset of the inner IDF does not occur until $R = 2.4$. The possible causes for such discrepancy can be attributed to the difficulty in specifying adequate inlet conditions (velocity profile and temperature) and certain deficiency in the reaction mechanism used in the modeling.

The predicted soot fields for $R = 0$, 1, and 2 are shown in Fig. 5 with the peak soot volume fraction indicated in each case. For the case without the central air flow, $R = 0$, the predicted peak soot volume fraction of 0.528 ppm is in good agreement with available data in the literature [30]. The predicted visible flame height of about

Fig. 4. Predicted temperature distributions for $R = 0, 1$, and 2 in the case of increasing the central air jet flow rate. The stoichiometric mixture fractions are marked by the white contours.

62 mm is also in very good agreement with the experimental observation of 63 mm shown in Fig. 2a. However, the model predicts a much taller visible flame height for $R = 1$ (52 mm vs. 40 mm), compare Figs. 5b and 2b. Nevertheless, the model predicts the rapid decrease in the soot concentration as the central air flow rate increases, which is again in good qualitative agreement with the experimental observation.

Fig. 5. Predicted soot volume fraction distribution for $R = 0, 1$, and 2 in the case of increasing the central air jet flow rate. The stoichiometric mixture fractions are marked by the white contours.

Fig. 6. The visible flame appearance for $R = 1$ and 1.5 by decreasing the central air flow rate from a state where the inner IDF is established.

Examination of the heat release rate distribution reveals that two heat release regions exist for $R = 1$ and 2 : one is the expected outer one along the stoichiometric contour and the other is an inner region exhibiting its maximum value at the centerline and not along the inner stoichiometric contour. At $R = 2$, the inner heat release rate is quite high and peaks at about 2.8 cm above the burner exit, which corresponds to the light blue spot shown in Fig. 2d.

The predicted flame structure and sooting characteristics after the onset of the inner IDF at $R \geq 2.4$ also reproduce those observed experimentally very well in a qualitative sense. The model shows soot drops to ppb level at $R = 4$, which corresponds well to the blue flame shown in Fig. 3e. Numerical results also show that for $R \geq 4$ the two stoichiometric contours merge to form a single contour off the centerline. This is consistent with the tip-opening of the flame for $R \geq 3.5$ shown in Fig. 3. At $R \geq 3$, the predicted inner IDF becomes lifted and the luminous zone is further reduced to a very small region. The lift-off height of the IDF first increases with the air flow rate and then remains almost constant for $R > 4$.

Fig. 7. Predicted temperature distributions for $R = 1$ and 2 .

4.2. Decreasing the central air jet flow rate

Once the inner IDF has been established for $R > 1.92$ as discussed above, it remains even when $R < 1.92$. The observed visible appearance of the flame for $R = 1$ and 1.5 is shown in Fig. 6. The sooting characteristics of the double diffusion flame (DDF), Fig. 6a and b, are strikingly different from those with an inner partially premixed flame, Fig. 2b and c, under conditions of identical air and fuel flow rates. Clearly, the DDF with the establishment of the inner IDF strongly favors soot formation compared to its counterpart without the IDF for $R \leq 1.92$ shown in Fig. 2. The enhancement of soot formation with the onset of the inner IDF can be explained primarily by the flame temperature effect on soot formation in diffusion flames. The occurrence of the inner IDF significantly increases the temperatures along the flame centerline region, which in turn greatly accelerates the pyrolysis of the fuel, leading to enhanced soot formation. Compared to the case without central air, $R = 0$, the luminous flame height of the DDF decreases significantly with the increase in the central air flow rate. This is easy to understand since the fuel requires less oxygen from the outer air stream to fully react when more air is added from the central air nozzle.

Numerical calculations for $R = 1$ and 2 with a converged solution for $R \geq 3$ also indicate the existence of the inner IDF and relatively high soot volume fractions with a peak value around 0.3 ppm. The calculated temperature distributions for $R = 1$ and 2 are shown in Fig. 7. The model predicts the liftoff of the outer NDF, though it is not observed experimentally. Overall the numerical results again agree qualitatively with the experimental observations.

5. Conclusions

Experimental observation and detailed numerical simulation of a double methane/air diffusion flame established in a three-port co-annular burner were carried out. The central air jet has a significant effect on the flame structure and sooting characteristics of the flame. At relatively low central air jet flow rates, two flame states exist, depending on increasing or decreasing the central air flow rate. The two flame states exhibit a remarkable difference in the flame appearance and soot loading. The numerical model reproduces very well the experimental observations, though differences in the critical central air jet flow rate for the onset of the inner inverse diffusion flame and in the visible flame height when the inverse diffusion flame is absent exist between the model and the experiment. The double diffusion flame shows different features

from the corresponding partially premixed flame and is an interesting flame configuration for control of soot formation in methane diffusion flames.

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