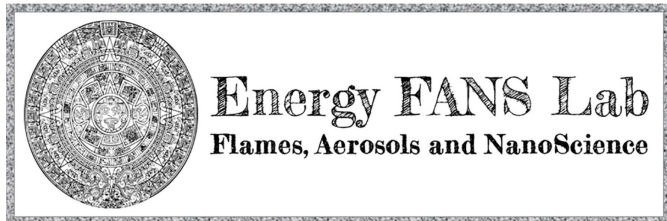


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Ultrafine particulate matter in methane-air premixed flames with oxygen enrichment

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A complementary computational and experimental study is carried out on the formation of ultrafine particulate matter in premixed laminar methane air flames. Specifically, soot formation is examined in premixed stretch-stabilized flames to observe soot inception and growth at relatively high flame temperatures common to oxygen enriched applications. Particle size distribution functions (PSDF) measured by mobility sizing show clear trends as the equivalence ratio increases from $\Phi = 2.2$ to $\Phi = 2.4$. For a given equivalence ratio, the measured distribution decreases in median mobility particle size as the maximum flame temperature increases from approximately 1950 K to 2050 K. The median mobility particle size is 20 nm or less for all flame conditions studied. The volume fraction decreases with increasing flame temperature for all equivalence ratio conditions. The $\Phi = 2.2$ condition is close to the soot inception limit and both number density and volume fraction decrease monotonically with increasing flame temperature. The higher equivalence ratio conditions show a peak in number density at 2000 K which may indicate competing soot inception processes are optimized at this temperature. Flame structure computations are carried out using detailed gas-phase combustion chemistry of the Appel, Bockhorn, Frenklach (ABF) model to examine the connection of the observed PSDF to soot precursor chemistry. Agreement between measured and computed flame standoff distances indicates that the ABF model could provide a reasonable prediction of the flame temperature and soot precursor formation for the flames currently studied. To the first order, the trends observed in the measured PSDF could be understood in terms of computed trends for the formation of

benzene, naphthalene and other soot precursors. Results of the current study inform particulate matter behavior for methane and natural gas combustion applications at elevated temperature and oxygen enriched conditions.

1. Introduction

Particulate matter formation is a crucial factor for combustion applications affecting performance [1], emissions regulations [2] and public health [3]. The most prominent particle produced in conventional combustion engines is soot originating from fuel-rich pockets. As such, a major thrust of combustion research is to develop fundamental knowledge of soot formation processes to minimize negative impacts. Natural gas is an important fuel for transportation, heating and process applications with recent boost in the United States from hydraulic fracturing resources [4]. Oxygen enriched conditions are often used with natural gas to optimize heat transfer and emissions performance [5]. The current study examines soot formation in premixed methane – enriched air flames using modeling and experimental approaches. Insights into soot inception and growth are gained for these conditions by measuring particle size distributions in the smallest particle size range. Studies on flame structure effects [6,7], parent fuel effects [8–10], and soot inception [11–13] have been reported for premixed methane flames but the current study focuses on ultrafine (< 100 nm) particle formation at elevated flame temperatures under oxygen enrichment. Maricq introduced probe sampling methods and addressed challenges for analysis of mobility size distributions in premixed flames [14–16]. Ever since several investigators have provided useful particle size experimental observations including extensions into particle sizes approaching 1 nm [17–22]. The current work uses the latest TSI mobility sizing system to examine mobility size down to 1nm.

Premixed stretch-stabilized flames are established by an aerodynamic balance rather than anchoring by heat loss [23] which enables temperatures approaching the adiabatic flame temperature. This configuration is applied in the current study to examine soot formation at elevated temperatures common under oxygen enrichment. Stretch-stabilized stagnation flames also enable systematic probe sampling under well-defined boundary conditions [24–27]. Flow perturbations at the sampling orifice are unavoidable but the stagnation surface is an explicit boundary that facilitates complementary modeling to account for probe effects on measured properties [28–30]. The time for particle growth could be reduced by up to 20% in these flames depending on the orifice and applied pressure drop [31]. With this in mind, flame structure modeling is carried out to interpret the observed soot formation behavior in terms of flame structure effects and soot precursor chemistry. conditions.

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2. Materials and Methods

2.1 Experimental - A series of methane-air flames with oxygen enrichment is designed to systematically observe the effects of flame temperature and equivalence ratio on soot formation in premixed laminar flames. Flame conditions spanning $2.2 < \Phi < 2.4$ and $1965 \text{ K} < T_{f,\max} < 2095 \text{ K}$ are summarized in Table 1. The nozzle-to-stagnation surface separation distance is $L = 1.5 \text{ cm}$ for all flames studied and the computed particle time ($t_p \sim 15 \text{ ms}$) is also comparable for all flames. The particle time can be considered to be the residence time for a particle nucleated at the flame zone and sampled at the stagnation surface. For a given equivalence ratio, the flame temperature is adjusted by independently adjusting the flow rate of diluent nitrogen gas fed to the flame. The experimental setup, summarized in Fig. 1, centers upon an aerodynamic nozzle ($D_{\text{nozzle}} = 1.43 \text{ cm}$) which issues the premixed fuel/air flow. The nozzle takes contours defined by Bergthorson [32] to induce a plug flow at the nozzle boundary. A concentric flow of nitrogen surrounds the flame to reduce perturbation from the surrounding environment. The temperature at the nozzle and stagnation surface boundary are monitored by Type K thermocouples with temperatures maintained at $T_{\text{nozzle}} = 330 \pm 20 \text{ K}$ and $T_{\text{stagnation}} = 473 \pm 20 \text{ K}$. Calibrated critical orifices are used to control all gas flow rates with oxygen enrichment established by using independent oxygen and nitrogen gas supply.

Table 1: Details of flame conditions studied

flame	Φ	T_{max} (K)	$X_{\text{CH}_4}^a$	$X_{\text{O}_2}^a$	$X_{\text{N}_2}^a$	v_o^b (cm/s)	t_p^b (ms)
2.2a	2.23	1980	0.323	0.289	0.388	32.5	16
2.2b	2.23	2010	0.349	0.313	0.364	31.2	15
2.2c	2.23	2050	0.365	0.327	0.338	30.0	14
2.2d	2.23	2095	0.368	0.329	0.308	28.7	14
2.3a	2.33	1980	0.354	0.304	0.343	29.6	17
2.3b	2.33	2025	0.370	0.318	0.312	28.3	16
2.3c	2.33	2040	0.378	.0325	0.297	27.7	16
2.4a	2.43	1965	0.383	0.316	0.301	27.4	18
2.4b	2.43	1980	0.399	.0329	0.272	26.3	17
2.4c	2.43	2050	0.420	0.347	0.233	24.9	16

A sample probe with orifice diameter of 130 micron is embedded into the stagnation surface with sampling procedures established to minimize particle diffusion losses and

artificial coagulation [33–39]. Particle size is measured by mobility sizing using a TSI 1 nm Scanning Mobility Particle Sizer (TSI 3838E77, SMPS) in the “compact” configuration to minimize diffusion loss of the smallest particles. This SMPS system contains a dual voltage classifier (TSI 3082), a Kr-85 bipolar diffusion charger (Neutralizer TSI 3077A), 1 nm differential mobility analyzer (DMA) (TSI 3086), a diethylene glycol-based (DEG) condensation particle counter (CPC) (so-called Nanoenhancer, TSI 3777) and a butanol-based CPC (TSI 3772). TSI Aerosol Instrument Manager Software (version 10.2) is used to collect and export the measured PSDFs. An insert supplied by the vendor is now mounted onto the inlet of the neutralizer to minimize flow recirculation for more predictable attainment of the equilibrium charge distribution. TSI also measured the penetration of ultra-fine particles through the system flow path recently and a new diffusion loss correction [40] is applied to the measured particle size distribution function (PSDF). The mobility size is corrected to properly account for the transition in gas-particle collision regimes for ultra-fine soot particles [41]. The dilution ratio is calibrated using independent flow-meter and CO₂ detector measurements. As discussed previously [30,42–45], the dilution ratio is correlated to indicated pressure drop at the sample probe dilution flow inlet and outlet. The dilution ratio applied to all flames is 2100 for the current study. The flame position is determined experimentally by analysis of flame projection images obtained from a Nikon D5300 DSLR camera.

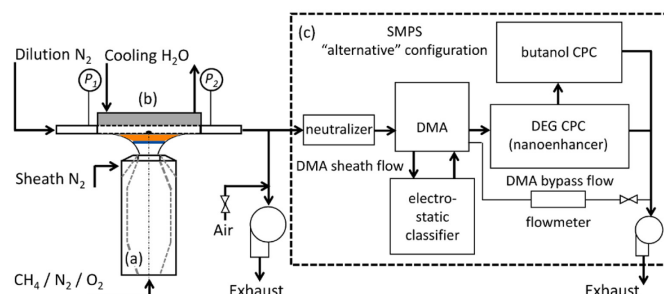


Figure 1 Experimental Setup including an aerodynamic nozzle for premixed flames (a), a stagnation surface / sampling probe assembly (b), and a TSI SMPS system in the compact configuration (c).

2.2 Computational - Premixed stretch-stabilized flames are a relatively simple axisymmetric flow field that can be solved using a similarity solution [46–49]. Previous studies have indicated that the similarity solution is reasonably accurate for stretch-stabilized flames with a wide range of nozzle-to-stagnation surface separations [50–53]. The OPPDIF flame solver [54] is used in the current study to compute the flame structure with a similarity solution in the Chemkin framework [55]. Detailed gas-phase combustion chemistry and transport is modeled with the Appel, Bockhorn, Frenklach (ABF) model [56] to examine flame structure and the effect of flame structure on production of PAH soot precursors. The current computations do not consider soot formation processes because extensive development of chemical reversibility, graphitization and other processes at elevated flame temperatures will be left to future work.

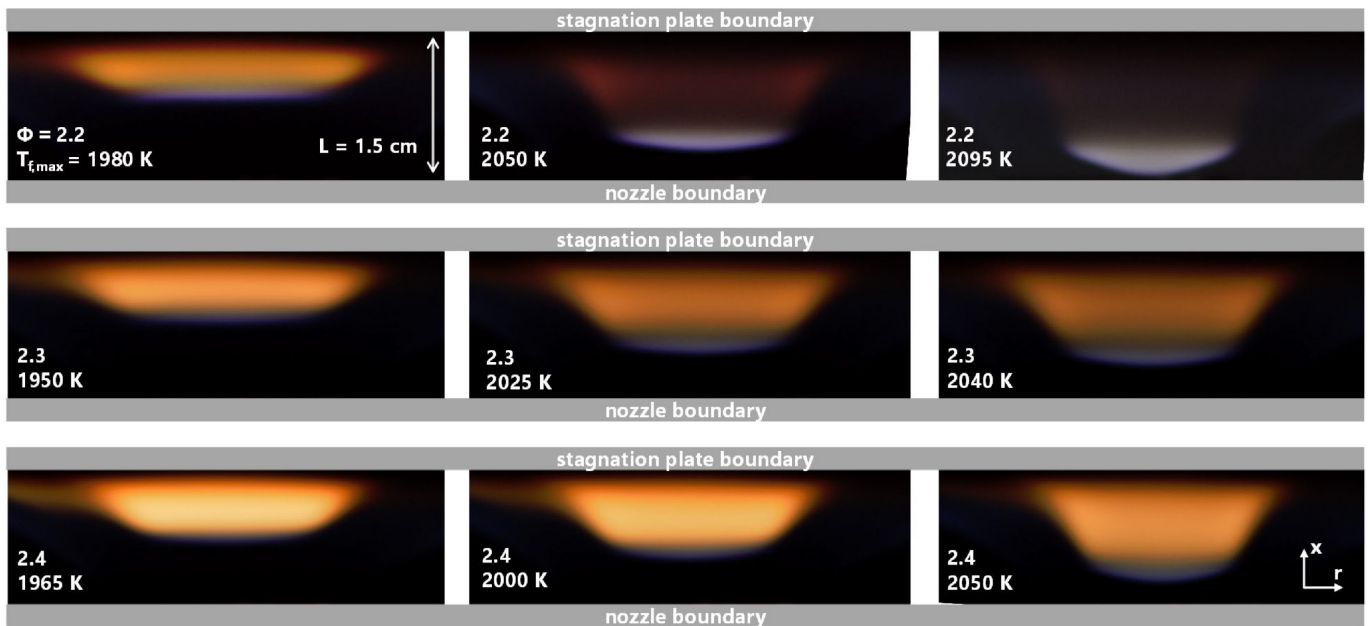


Figure 3 Images for the series of flames currently studied.

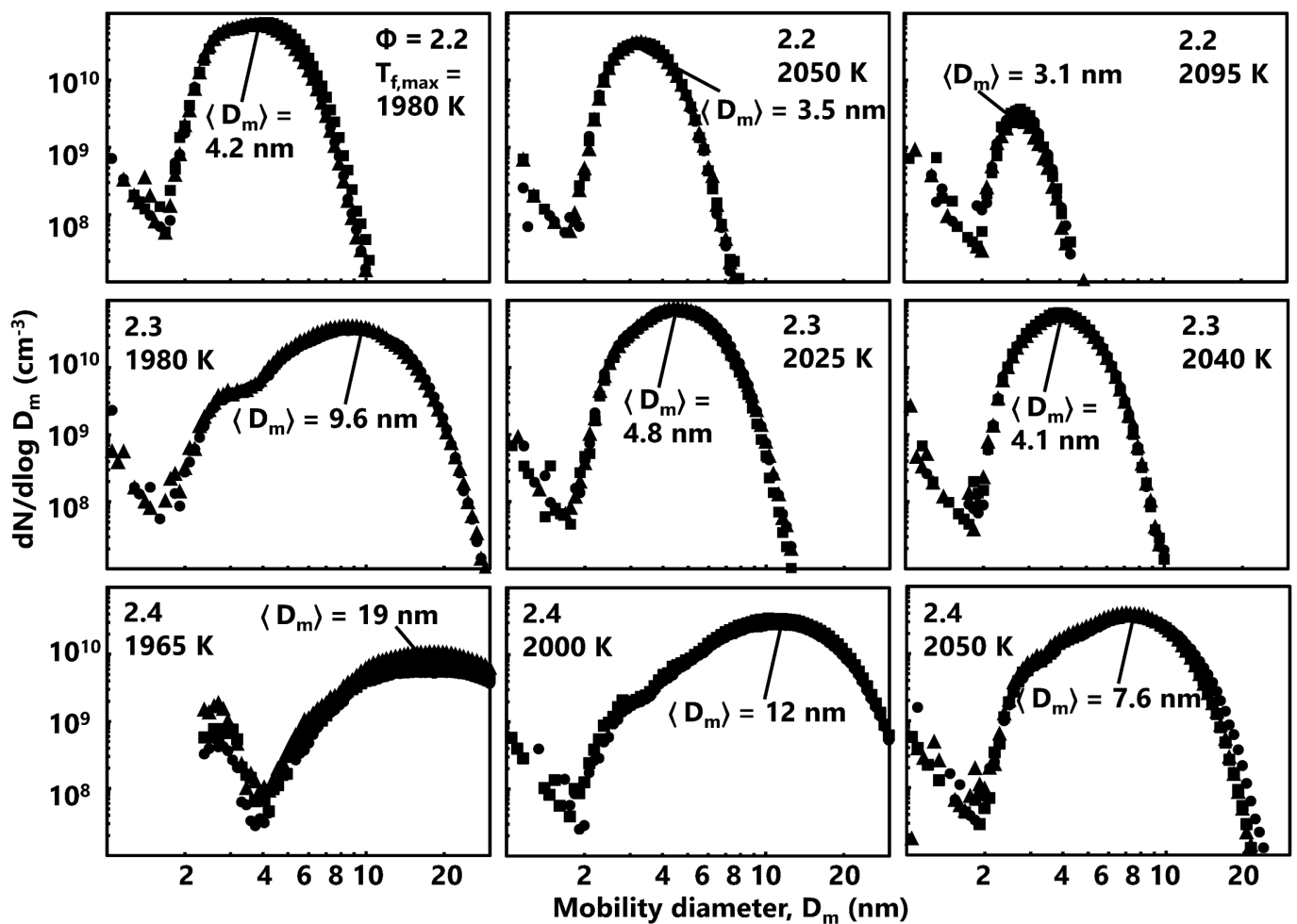


Figure 2 Measured PSDF for the series of flames currently studied. The PSDF measurement is repeated three times and different symbols are used to highlight repeated measurements. The median diameter, $\langle D_m \rangle$, is labelled for each flame condition as well.

3. Results and discussion

Images of the series of flames currently studied are shown in Fig. 2. The image is a projection of the axisymmetric stagnation flow with a steady flame position indicated by the thin blue disc. For flames sitting close to the nozzle boundary, non-ideal deviations from the flat disc shape are observed due to the tendency of the flame to anchor. The intensity of the orange luminosity in the post-flame region is an indication of the amount of soot produced in the flame. As expected, the most intense luminosity is observed for the most fuel rich flames.

Measured PSDF for the series of flames currently studied is shown in Fig. 3. For a given equivalence ratio, the median mobility particle size decreases as the flame temperature increases. This indicates that soot formation processes are hindered at elevated temperatures due to reversibility in precursor formation and soot growth [6,57], increase in OH oxidation [58] and a reduction in coagulation efficiency [59–62]. For comparable flame temperature, the median mobility diameter increases over a factor of two from $\langle D_m \rangle = 4.2$ nm to 9.6 nm to 19 nm as the equivalence ratio increases incrementally from $\Phi = 2.2$ to 2.3 to 2.4. With exception of the $\Phi = 2.4$, $T_{f,max} = 1965$ K condition, the median mobility particle diameter is on the order of 10 nm and below. Global properties derived from the measured PSDF are shown in Fig. 4 for the series of flames currently studied. The number density is the area under curve for the measured number weighted PSDF but extraction of the volume fraction requires an assumption of the particle morphology. The measured mobility diameter is interpreted to correspond to spherical particles as evidence suggests for particles in the current size range [30,56,63]. For each mobility diameter bin, a volume weighting corresponding the sphere volume is applied and the volume fraction is the area under the volume weighted PSDF. For the lowest equivalence ratio condition, a clear downward trend in number density and volume fraction is observed as the flame temperature increases. The higher equivalence ratio conditions also show decreasing volume fraction with increasing flame temperature, but the number density shows a broad peak in this temperature range. This broad peak in the number density but down trend in volume fraction may indicate that soot inception is optimal for 2000 K flames but growth processes are favored at lower flame temperatures. A peak in global sooting properties is a known temperature dependent behavior reported ever since early flame [64] and shock-tube studies [65]. As Fig. 2 shows, the lowest equivalence ratio series is close the limit of visible soot luminosity. The downward trend for both number density and volume fraction in this case may indicate that soot inception is favored at lower temperature for sooting limit flame conditions. Experimental observation of the detailed PSDF for the earliest inception stages provides a valuable guide for developing soot formation models including recent hypotheses for soot inception [66–70].

Complementary flame structure calculations are carried out for the series of flames currently studied. The flame standoff distance is considered to be the distance from the blue flame disc to the stagnation surface. This is measured in the current study from analysis of pixel intensities in the flame projection images. The computed flame standoff distance is considered to be the location of the peak CH^* concentration in the centerline

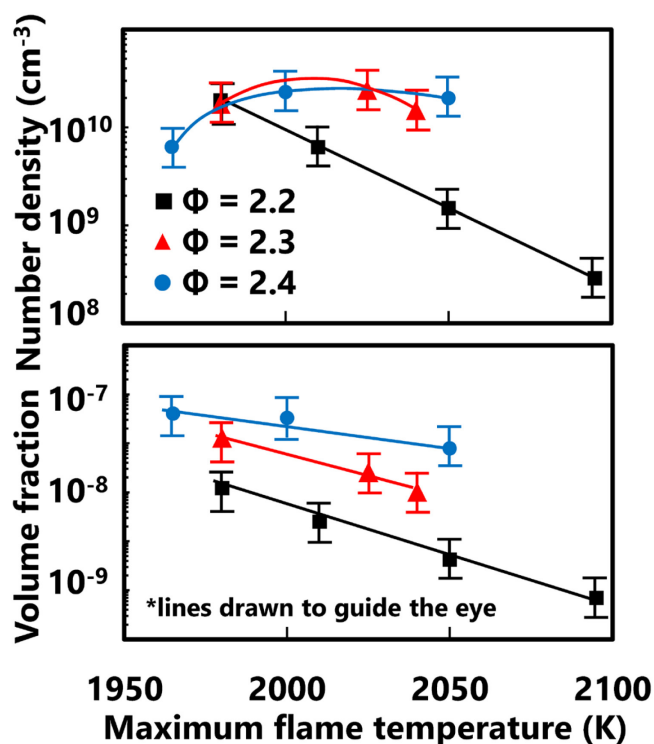


Figure 4 Global particle distribution properties derived from the measured PSDF. Lines are drawn to guide the eye.

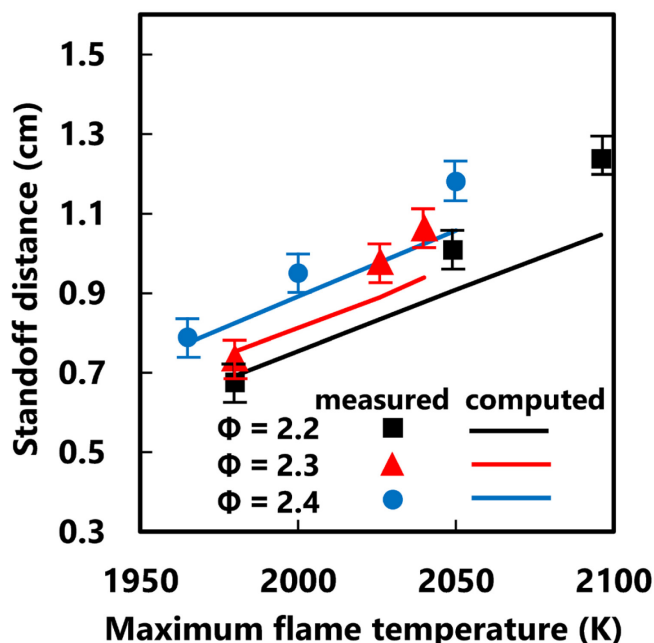


Figure 5 Measured and computed flame standoff distance for the series of flames currently studied.

axial profile. A comparison between measured and computed values is shown in Fig. 5. Reasonable agreement is observed as long as the flame does not approach the vicinity of the nozzle (high standoff distance). As discussed above, the flame disc distorts significantly for flame position close to the nozzle due to the tendency to anchor onto any solid surface. This effect causes a significant disagreement between the measured flame standoff distance for the $\Phi = 2.2$, $T_{f,max} = 2095$ K condition.

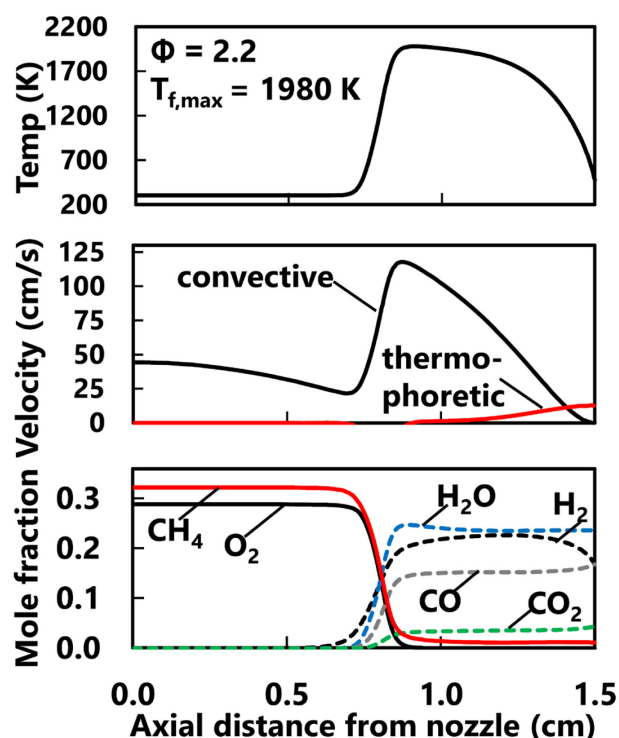


Figure 7 Computed axial temperature profile (top), axial velocity profiles (middle) and major products (bottom) at the centerline for the $\Phi = 2.2$, $T_{f,max} = 1980$ K flame.

Reasonable agreement for the other conditions indicates that the ABF combustion chemistry model reasonably predicts the

fuel burn rate and flame temperature. The delicate kinematic balance between opposing burning and flow velocity determines the flame position and the ABF model is able to capture the experimental behavior.

With the performance of the ABF model established for the current flames, the flame structure calculations could provide insight into temperature and flame chemistry effects. The computed flame structure for the $\Phi = 2.2$, $T_{f,max} = 1980$ K case is shown in Fig. 6 to demonstrate the profiles for a typical premixed stretch-stabilized flame. The axial temperature profile shows the sharp temperature increase at the flame reaction zone significantly downstream of the nozzle exit. The axial convective velocity also increases at the flame and falls down to zero at the stagnation surface. The thermophoretic velocity of the soot particles is calculated based on the assumption that the particles follow the gas streamlines and the hard sphere approximation applies. Assuming inception of the first particles occurs at the flame reaction zone, the time for the particle to traverse the flame zone to the stagnation surface is comparable across all flames (see Table 1). The computed major species for this fuel rich flame show CO production is much higher than CO₂ but H₂ and H₂O are produced at comparable levels. Computed profiles for light and heavy soot precursors are shown in Fig. 7 for the series of flames currently studied. As Table 1 shows, the computed particle residence times are all within 13% which effectively minimizes growth time effects on the measured PSDF and computed species profiles. Interestingly, the predicted concentrations of acetylene and propargyl radical are not substantially different across the current range of equivalence ratio and flame temperature. In contrast, the

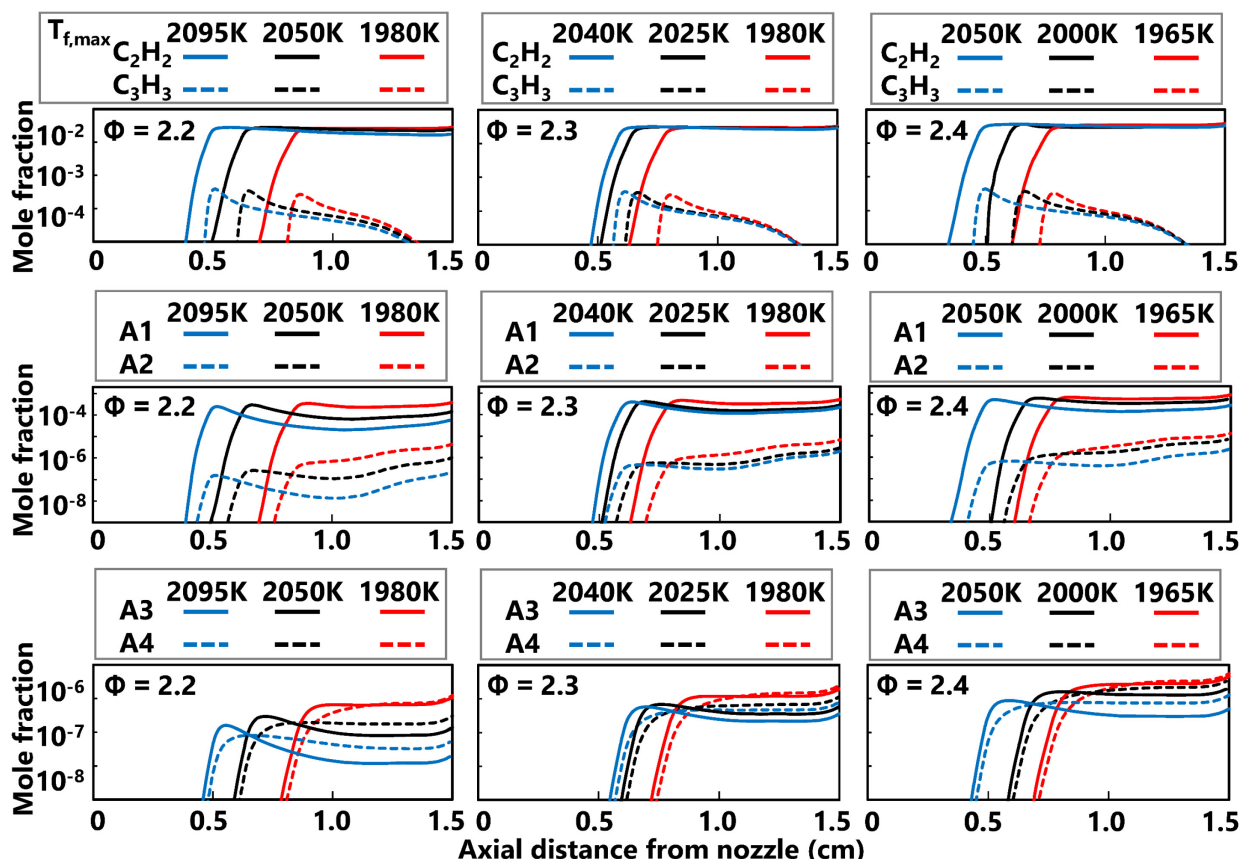


Figure 6 Computed centerline axial profiles of soot precursors for the series of flames currently studied. The top row shows the production of acetylene (C₂H₂) and propargyl radical (C₃H₃), the middle row shows the production of benzene (A1) and naphthalene (A2), and the bottom row shows the production of phenanthrene (A3) and pyrene (A4).

computed profiles for benzene and PAH predict concentrations show some sensitivity to the flame temperature. The measured volume fraction decreases with increasing flame temperature for all equivalence ratio series but the trends for computed PAH are not strong in all cases. For the $\Phi = 2.2$ case, the computed A1-A4 profiles show a clear decreasing trend with increasing flame temperature. This downward trend is generally true for the $\Phi = 2.3$ case but narrower range in flame temperature results in a narrower spread in predicted concentrations. The highest equivalence ratio conditions show a relatively weak sensitivity of the computed A1-A4 mole fractions even though the predicted range of temperatures is wider.

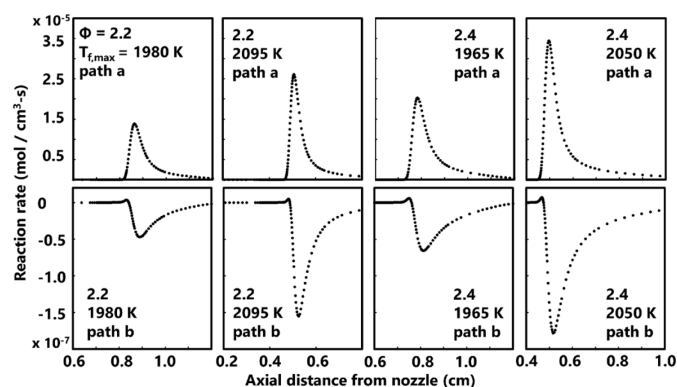


Figure 9 Computed rates of two common benzene (A1) production reactions for four flames spanning the range of equivalence ratios and flame temperatures currently studied. The top row is the axial profile for the net rate of $2C_3H_3^\bullet = A_1$ (path a) and the bottom row is the axial profile for the net rate of $n-C_4H_5^\bullet + C_2H_2 = A_1 + H^\bullet$ (path b).

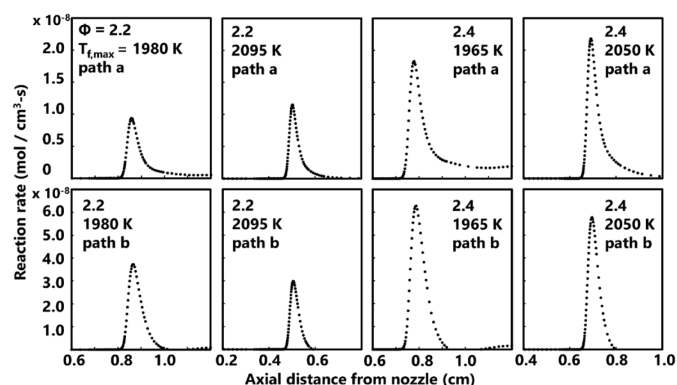


Figure 10 Computed rates of two common naphthalene (A2) production reactions for four flames spanning the range of equivalence ratios and flame temperatures currently studied. The top row is the axial profile for the net rate of $A_1^\bullet + C_4H_4 = A_2 + H^\bullet$ (path a) and the bottom row is the axial profile for the net rate of $A_2^\bullet + H^\bullet = A_2$ (path b).

Reaction rates predicted by the ABF model are also shown here to provide insight into soot precursor formation pathways. Computed profiles for rates of propargyl recombination ($2C_3H_3^\bullet = A_1$) and acetylene + butadienyl ($n-C_4H_5^\bullet + C_2H_2 = A_1 + H^\bullet$) is shown in Fig. 8 for four flames spanning the range of equivalence ratios and flame temperatures currently studied. The most significant reaction pathway for benzene production is propargyl recombination for the current series of methane flames. As shown in Fig. 8, the C2 pathway is not favored as the reverse reaction consumes a small fraction of benzene in the flame. Predicted production rates for naphthalene based on the $A_1^\bullet + C_4H_4 = A_2 + H^\bullet$ pathway and the $A_2^\bullet + H^\bullet = A_2$ pathway

are shown in Fig. 9. These two pathways are the most significant for the series currently studied based on the rates predicted by the ABF model. The phenyl radical pathway is slightly slower than the A2 radical pathway but this channel is a culmination of multiple pathways to form the A2 radical. The selected reaction rate profiles for benzene and naphthalene show a modest sensitivity to flame temperature and equivalence ratio.

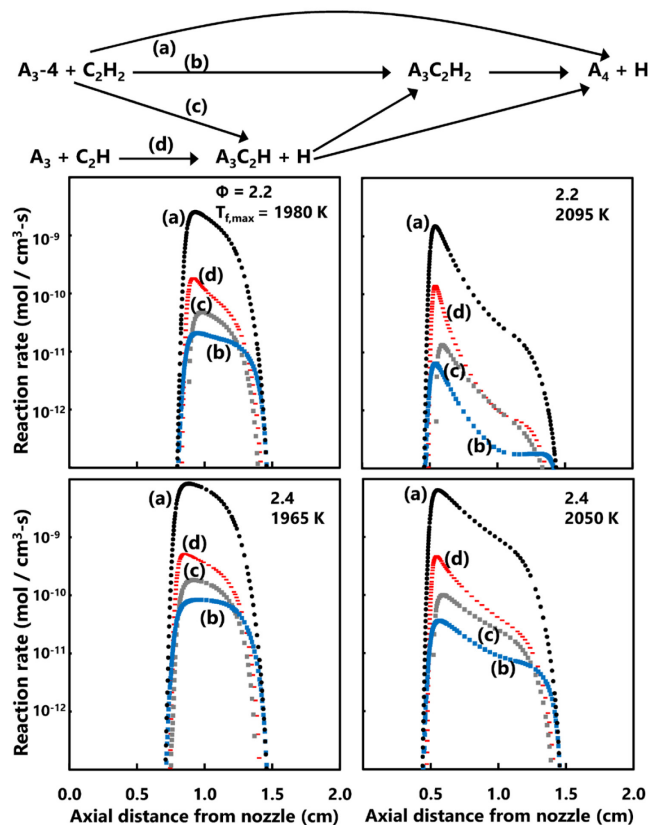


Figure 8 Reaction pathways for production of pyrene (A4) considered by the ABF model (top) and axial profiles of computed reaction rates for four main pyrene production pathways (bottom).

A common soot inception model is to consider pyrene dimerization as the effective onset of soot particles [71–74]. The ABF model considers four major pathways, summarized in Fig. 10, to form pyrene all stemming from phenanthrene species. Phenanthrene radical (A_3-4) is postulated to combine with acetylene to either form pyrene directly or to form radicals with unclosed rings. Phenanthrene can also react with C_2H radical to form an unclosed ring. For the current series of flames, the ABF model predicts that the direct formation of pyrene from the phenanthrene radical is an order of magnitude faster than the other pathways. Increasing flame temperature also results in somewhat lower rates of pyrene production. The species and reaction rate profiles provide insight into the underlying soot precursor flame chemistry. The experimental PSDF provide a guide for future soot formation modeling in these premixed, enriched air methane flames.

4. Conclusions

Measured PSDF of ultrafine particulate matter formed in premixed methane – air flames are examined with complementary flame structure computations. Relatively high flame temperatures are investigated by using oxygen enrichment and premixed stretch-stabilized flames. For a given equivalence ratio, the measured distribution decreases in median mobility particle size as the maximum flame temperature increases from approximately 1950 K to 2050 K. The lowest equivalence ratio, highest temperature flame condition corresponds to a PSDF having a median mobility diameter of 3 nm while the highest equivalence ratio, lowest flame temperature condition corresponds to 20 nm. The soot number density shows a broad peak on the order of 10^{10} cm^{-3} for the higher equivalence ratio conditions and decreases to 108 for the lowest equivalence ratio. The measured soot volume fraction shows a monotonic decrease with increasing flame temperature from 10^{-7} .

Flame structure computations are carried out using detailed gas-phase combustion chemistry of the Appel, Bockhorn, Frenklach (ABF) model to examine the connection of the observed PSDF to soot precursor chemistry. Agreement between measured and computed flame standoff distances indicates that the ABF model could provide a reasonable prediction of the flame temperature and soot precursor formation for the flames currently studied. To the first order, the trends observed in the measured PSDF could be understood in terms of the computed trends for the formation of benzene, naphthalene and other soot precursors. Propargyl recombination is the dominant pathway to benzene formation for the current methane flames. According to the ABF model, naphthalene formation occurs by combination of phenyl radical + C₄H₄ and other pathways leading to naphthalene radical formation. Pyrene formation is predicted to be fastest through direct formation after combination of phenanthrene radical and acetylene. Results of the current study inform particulate matter behavior for methane and natural gas combustion applications at elevated temperature and oxygen enriched conditions.

Conflicts of interest

There are no conflicts to declare

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