Burner-Stabilized Stagnation Flow Flame Approach to Probe Soot Size Distributions

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Abstract

In previous studies we used a tubular probe to sample incipient soot formed in premixed flat flames and determine the particle size distribution by scanning mobility particle sizing (SMPS). Comparison of the spatially resolved soot volume fractions by SMPS and by thermocouple particle densitometry suggests that the probe causes a positional offset of -0.35cm in comparison to TPD, and indicates that the particle size distributions measured is influenced by flame perturbation by the probe. Moreover, the flame boundary conditions are difficult to define. In the present work, we propose a burner-stabilized stagnation-flow (BSSF) flame approach, whereby the particle probe is imbedded in a water-cooled flat plate which acts both as a sample probe and flow stagnation surface. As the boundary conditions of this flow configuration can be completely defined experimentally, the flame was simulated with a quasi 1-D stagnation flow code considering radiative heat losses by CO2 and H2O. It was found that the simulated and measured gas temperature profiles compare well. The evolution of the soot size distributions obtained by the BSSF approach was found to be similar to that of the tubular probe.

Introduction

Probe sampling with scanning mobility particle sizing (SMPS) is now routinely used to follow the size evolution of soot formed in flames [1-17]. Using this technique, soot nucleation and mass/size growth may be closely investigated by resolving spatially the detailed particle size distribution (PSDF). Studies in premixed flat flames and well stirred reactors have been reported. They covered a broad range of experimental conditions and investigated the effect of temperature, fuel structure, and equivalence ratios on the detailed processes close to and beyond soot inception. Particles as small as 1.6 nm can be probed [11, 16], and the same technique has been applied to the analysis of metal oxide and other nanomaterial formation in flames [9, 18-20].

Results obtained from the probe samping/SMPS fundamental soot technique also aided model development and validation (see, e.g., [2, 17, 21-23]) and yielded useful information about the elementary processes associated with soot formation. One of the key observations is that the PSDF of incipient soot is persistently bimodal [16]. The cause for bimodality is the competition between particle inception and particleparticle coagulation [2]. It was shown that the shape of size distributions and its evolution can provide a wealth of kinetic information about elementary sooting processes [21].

Despite its increasingly widespread use, the probe sampling/SMPS technique has two fundamental drawbacks. First, care must be taken to minimize particle

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loss in the sampling probe [3]. The loss mechanisms include diffusion of the particles to the probe walls and coagulation among particles. In general, preventing particle losses requires rapid and large dilution of the flame sample by a cold, inert gas stream - a problem that can be solved with a proper probe design [3][13, 16]. The second drawback is more difficult to deal with, and indeed it has not been properly addressed. Probe sampling is inherently intrusive. An earlier study [3] demonstrated that the use of a tubular probe placed horizontally across a premixed flat flame (see, Figure 1) causes a significant drop in the local flame temperature near the sampling point, and this influence extends several millimeters from the probe. In addition, the probe also introduces local flow stagnation, leading to a longer reaction time than that in a burner-stabilized flame free of solid objects. More recently, probe perturbation and its influence on soot growth kinetics has also been discussed by Sgro et al. [15]. This problem is also expected to occur in molecular beam mass spectrometry studies of low-pressure premixed flat flames, in which a conical nozzle is inserted into the flame to sample flame species (see, e.g., [24, 25]).



Figure 1. Schematic illustrating a previous tubular probe sampling technique [2, 3].

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Modeling studies generally assume that the flow field in a premixed flat flame is unperturbed by the presence of a probe. For this reason, a comparison of the experimental and modeling results is approximate at best, since the boundary conditions of the reacting flows in the two cases are quite different. To probe the species concentration or soot PSDFs in a spatially resolved manner, a probe is usually positioned at several distances from the burner surface. Since the flow field for each position is unique, these measurements yield a series of flames, each of which are basically different from another. That is, the boundary conditions of unburned gas are expected to be identical, but those at the sampling points are different.

Historically, this problem was dealt with by shifting the computed species concentration or soot volume fraction profiles downstream by a certain distance (see, e.g., [21, 26]). Experimental evidence does support this exercise. Previous studies [13, 16] on lightly sooting flames measured temperature profiles of the free-stream gases at increasing heights above the burner surface. Using a thermophoretic particle densitometry (TPD) technique developed by McEnalley et. al. [27] the local soot volume was determined. Independently, the soot volume fraction was measured by integrating particle size distributions obtained by SMPS. The results show that the SMPS data need to be shifted upstream by 0.35 cm to yield an agreement with the TPD soot volume fraction. This shift, presumably due to probe perturbation, was found to be a constant for a set of five canonical flames over a quite wide range of maximum flame temperature (1610 K < $T_{f,max}$ < 1898 K) [13]. Of course, the agreement on soot volume fraction does not suggest that the spatial shifting is valid with regards to the various features observed for PSDFs. For probes of different size, shape and geometry, the degree to which they perturb a flame also varies-a factor that can contribute to differences in the experimental data collected on different facilities.

The purpose of the present paper is to report and discuss a new approach that can resolve the problems just discussed. Specifically, we introduce the burnerstabilized stagnation (BSS) flame approach, in which a sampling probe is embedded in a water cooled plate, which also acts as a flow stagnation surface. Unlike the tubular-probe approach, the BSS flame approach allows for the flame boundary conditions to be fully specified and the divergent reacting flow can be simulated by the 1-dimensional OPPDIF standard code [28, 29]. Compared to the use of one-dimensional free-flow code (e.g., PREMIX [30]), the computational cost in the BSS flame approach is only moderately more expensive, but a rigorous comparison of the experimental and modeling results is now possible.

In fact, a stagnation surface has been traditionally used for burner stabilized flame studies to further stabilize the flame downstream from the main reaction zone, but the condition on the surface is often poorly characterized. Its influence on the flame is, again, almost never considered in modeling studies. It was shown recently [31] that depending on the details of experimental setup, including whether or not a stagnation surface was used, the flame soot profiles could be drastically different. With the use of BSS flame technique, we utilize the flame stabilizer as the probe itself in such a way that the various uncontrolled parameters, as discussed in [31], are either fully controlled or rendered unimportant.

To demonstrate the above ideas, we characterized the temperature profiles in a well-studied, fuel-rich ethyleneoxygen-argon flame, comparing several burner setups, including the BSS flame approach, the use of a tubular probe, and a flame free of probes. Numerical calculations were carried out to aid in data interpretation and to examine how appropriate the employed flame boundary conditions are in the BSS flame approach. We also provide a comparison of the soot PSDF, volume fraction, and number density data obtained by the BSS technique and by the previous tubular-probe approach.

Experimental Methods

The experimental setup is similar to what was described in an earlier work [2]. In the BSS flame approach, modifications were made to the sampling probe, as will be discussed later. Soot was generated by a water-cooled flat flame burner on which a 16.3% ethylene-23.7% oxygen-argon flame (equivalence ratio $\phi = 2.07$) was stabilized at atmospheric pressure. The burner is made of a sintered porous plug with an outer diameter of 7.6 cm. The cold gas velocity is 8 cm/s (STP), which is identical to Flame C3 reported in a previous study [13]. The flame was isolated from the ambient air by a shroud of nitrogen flowing at 43.6 cm/s (STP) through a concentric porous ring. The reactant gases were metered by critical orifices.

The burner temperature was measured by a type-K thermocouple embedded at the bottom of the porous plug. In each experiment, the burner was allowed to reach a steady-state temperature of 353 ± 10 K. The gas temperature was determined using a Type-S thermocouple coated with a Y/BeO mixture to prevent surface catalysis. The diameter of the coated thermocouple wire was 125 µm. Radiation corrections were made using the method of Shaddix [32], with gas properties estimated by solving flame structure and species profiles using detailed reaction kinetics and transport [13], as will be discussed later. Literature emissivity values for Y/BaO ceramic coating ranges from 0.3 to 0.6 [33]. Here we used the range of emissivity values to yield the upper and lower



Figure 2. Schematic of burner-stabilized stagnation (BSS) flame approach. Left panel: plate sampler as seen from the bottom up of the burner exit, right panel: actual flame image and illustration of the coordinate system and flow field.

limits for the radiation correction. The radiationcorrected temperature is estimated to be the average of the two limiting values.

The BSS flame approach is shown schematically in Figure 2. A tubular probe was embedded in a circular aluminum plate positioned in parallel to the burner surface at the separation distance of H_p , which can be resolved to within a positional accuracy of ±0.015 cm. The top of the plate is water cooled. The flow streamlines are that of an impinging jet on a stagnation surface. The plate temperature is measured by a type-K thermocouple embedded at the bottom of the plate towards the incoming flame gas. Care was taken to position the thermocouple flush with the bottom of the plate so as to maintain a flat stagnation surface above the burner. A thin slit is cut into the bottom of the plate such that the orifice in the probe tube is flush with the bottom of the plate, facing the incoming flame gas. The probe is made of a stainless steel tube 0.635 cm in outer diameter and has a wall thickness of 125 µm. The flame sample was drawn into the probe through an orifice 127 µm in diameter. The orifice was placed on the central axis of the burner. The pressure across the orifice was precisely controlled and measured by manometers placed upstream and downstream of the orifice. To compare the BSS flame approach with the previous tubular-probe method, a second setup used identical flame and sampling conditions, with the exception that the stagnation plate was removed.

The flame sample entered the probes through the orifice and was immediately diluted with a cold nitrogen flow at 30 L/min (STP). To ensure that particle losses are negligible in the sampling line, we used a dilution procedure identical to previous studies [2, 3]. The dilution ratio was determined by calibrating the flow through the orifice as a function of the pressures and by direct measurement of the CO concentration in the flame using a gas chromatographer [13]. The two independent calibrations produced nearly identical results. The dilution ratio may be related to the pressure drop ΔP across the orifice empirically as

$$\log DR = a + \frac{b}{\log(c \bullet \Delta P)} , \qquad (1)$$

where for the current probe the parameters $a = 2.0659\pm0.10478$, $b = 0.70252\pm0.10583$, and $c = 0.093463\pm0.007039$ (mmH₂O)⁻¹. The range of pressure drop used was 50 to 150 mm H₂O, which gives a dilution ratio *DR* generally over 10³. The uncertainty of the calibration was used to quantify the uncertainty of the soot volume fraction data measured by the SMPS.

A standard SMPS system from TSI was used to classify the flame aerosols. The SMPS consists of a single stage inertial impactor with an orifice diameter of 0.071 cm, a 3077 charger and a 3080 classifier with a nano-Differential Mobility Analyzer (TSI 3085, nDMA) and a 3025A ultrafine Condensation Particle Counter (UCPC). The UCPC has a lower mobility size limit of 3 nm due to small activation efficiency at smaller sizes. The sample and sheath flows through the DMA were 1.5 and 15 L/min, respectively. Corrections for diffusion losses in the transfer lines and DMA were accounted for as a function of particle diameter using the AIM software.

Mobility measurements can overestimate the true size of particles smaller than 10 nm because of inherent limitation of the empirical Cunningham slip correction [34]. This correction does not account for (a) the transition from diffuse to specular scattering [35], and (b) the van der Waals gas-particle interactions, both effects are expected to be important for particles below 10 nm in diameter [34]. A nanoparticle transport theory [34-36], where these interactions are accounted for, gives a parameterized correlation between the mobility diameter $D_{p,SMPS}$ and true diameter D_p of a carbonaceous particle as [21]:

$$\frac{D_p}{D_{p,SMPS}} = \tanh\left(1.4566 + 0.010892D_{p,SMPS}\right) \times \left(1.0721 - \frac{0.4925}{D_{p,SMPS}}\right)$$
(2)

In the above equation, $D_{p,SMPS}$ has units of nm. The particles studied were assumed to be spherical. A previous study [16] used atomic force microscopy to investigate the morphology of nascent soot formed in lightly sooting premixed flames and showed that the particles are liquid-like and presumably exist as spherical or near spherical droplets in flames. All particle diameters presented hereafter are corrected according eq 2.

Computational Method

For the free-flow flame without the presence of flow divergence, we used the PREMIX code [30]. The BSS

flames studied here have a separation-to-diameter ratio = 1, which is appropriate for numerical solution using the pseudo one-dimensional formulation for counterflow reacting jets of Smooke et al. [29]. Here, we used a modified version of OPPDIF [37], accounting for flame gas radiation. Boundary conditions are specified as follows. At the burner exit (x = 0), the flow is a uniform plug flow with a mass flux nk_0^{c} given by the experimental cold flow velocity and reactant composition and a boundary temperature T_b equal to the measured burner temperature. The mass flux for the k^{th} species is $nk_0^{c}Y_{k,0}$, where $Y_{k,0}$ is mass fraction in unburned gas. These boundary conditions can be expressed as

$$F = \rho u/2 = n \delta_0/2 , \qquad (3)$$

$$G = \rho v/r = 0, \qquad (4)$$

$$\rho Y_k \left(u + V_k \right) = n \delta_0 Y_{k,0}, \tag{5}$$

where ρ is the mass density, u and v are the axial and radial convective velocities, respectively, and V_k is the axial diffusion velocity of species k. The stagnation plate, located at $x = H_p$, is treated as a no-slip wall, so u, v, and V_k are all 0. It has a temperature T_s equal to the measured plate temperature. The gradients for the mass fractions of all species were assumed to vanish at $x = H_p$ —an assumption expected to be valid so long as the free radical concentrations are negligibly small immediately below the stagnation surface, which is indeed the case here.

There are some issues with applying the OPPDIF formulation to this particular problem which should be noted briefly. It is assumed that there is no axial pressure gradient. This is justified since the flow is a low Mach number deflagration in a boundary layer [38]. Furthermore, the flames originally studied in [29, 38] have inlet flow velocities sufficiently high, so buoyancy can be neglected. The agreement between the simulation results and experimental measurements shown later suggests that buoyancy is again negligible for the flames studied here.

Numerical simulations used windward differencing, multicomponent transport and thermal diffusion. Heat release rates and transport properties were calculated using Sandia CHEMKIN [39] and TRANSPORT [40]. The reaction kinetic model used was USC-Mech II [41], which has 111 species and 784 reactions. The reaction model was developed for $H_2/CO/C_1$ -C4 hydrocarbon combustion through a series of kinetic modeling studies over the last decade [42-49]. Adaptive mesh resolution was used and it was found that the flame is sufficiently resolved with roughly 200 grid points.

Modifications to the OPPDIF code include mainly radiation correction [50]. Assuming that the gas is

optically thin, an additional radiative heat loss term q_r to the energy equation

$$q_r = 4\kappa_p \sigma \left(T^4 - T_0^4\right) \tag{6}$$

was added into the energy conservation equation. In eq 6, T is the local temperature, T_0 is the ambient temperature, σ is the Stefan-Boltzmann constant, and κ_p is the Planck mean absorption coefficient of the gas. Since a fluid particle in the flame will see mostly either the stagnation plate or the burner surface, T_0 was taken to be the average of these two temperatures. The temperature-dependent κ_p values for H₂O and CO₂ were taken from [51], while CO was assumed to have a κ_p 10 times lower than CO₂ [52]. κ_p for soot was estimated, according to [51], as

$$\kappa_p = 3.83 \,[\mathrm{cm}^{-1}\mathrm{K}^{-1}]C_0 T f_v \,, \tag{7}$$

where f_{ν} is the local soot volume fraction and C_0 is determined as [51]

$$C_0 = 36\pi \frac{nk}{\left(n^2 - k^2 + 2\right)^2 + 4\left(nk\right)^2} \,. \tag{8}$$

In eq. 8, (n + ik) is the complex refractive index of soot, assumed to be 1.57 + 0.56*i* [53]. As will be shown later, the maximum soot volume fraction obtained for the lightly sooting flame is roughly 10⁻⁸ and the maximum flame temperature is $T_{f,max} = 1837$ K. Assuming that soot temperature, *T* in Eq. (7), is the same as the gas temperature, we obtained a mean absorption coefficient two orders of magnitude lower than that for gas radiating species (CO₂ and H₂O). Hence, only radiation loss by gas phase species is considered in the simulations.

Results and discussion

Centerline temperature was determined as a function of distance from the burner surface, $H (0 < H < H_p)$ for several stagnation-surface (probe) positions, ranging from $H_p = 0.55$ to 1.2 cm, as shown in 3. In addition, the temperatures at the burner and stagnation surfaces, $T_b =$ 473 ± 20 K and $T_s = 511\pm20$ K, were independently measured and are also included in Figure 3. The data were corrected for thermocouple radiation. As seen, each position yields a different flame, even though the boundary temperatures are almost the same. As the stagnation surface approaches the burner surface, the peak flame temperature becomes lower, due to an enhanced heat transfer rate to the stagnation plate. For the six temperature profiles shown, the preheat zone up to H =0.15 cm is however identical, indicating that the heat



Figure 3. Measured centerline temperature profiles (symbols, radiation corrected) for several separation distances (H_p) between the stagnation and burner surfaces. Lines are fits to data.

transfer rates to the burner surface is not affected significantly by the burner-to-stagnation separations.

The stagnation surface allows the flame boundary conditions to be accurately defined. With the measured burner and stagnation surface temperatures $(T_b \text{ and } T_s)$ and the mass fluxes of the unburned gas mixture used as input boundary conditions, we solved the detailed flame structures for the underlying stagnation, reacting flow problem using the OPPDIF code. The computed temperature profiles are shown as lines in Figure 4 and compared to the radiation-corrected flame temperature data. As seen, the agreement between experiment and model is very good for all six H_p values tested. This agreement is encouraging as the flame structure can be calculated with relative ease and high accuracy. The base model can be used as a foundation for advanced soot models with a high level of confidence that experiments and models may be compared directly.

As we discussed earlier, the vertical error bars of Figure 4 represent the uncertainties from the emissivity of the thermocouple coating, which ranges from 0.3 to 0.6 and is by far the greatest uncertainty among factors considered. The horizontal error bars indicate positioning accuracy. The computed temperatures generally fall within the error bars of the data, except for spatial positions close to the stagnation surface, where the model gave slightly larger flame temperature values. This discrepancy can be explained by the finite elasticity of the fine thermocouple wire and the lower drag force acting on the thermocouple as the flow velocities near the plate fall off, resulting in a positional uncertainty of the thermocouple near the sampling plate relative to that upstream where the axial flow velocity is much larger.



Figure 4. Comparison of experimental (symbols, radiationcorrect) and calculated (lines) centerline temperature profiles at several separation distances between the burner and stagnation surfaces.

The difference may also be caused by soot radiation to an extent, which was not considered here as discussed earlier. This effect can be accounted for when a joint simulation is carried out in which both soot formation and the radiative losses from soot are considered.

To understand the cause for the discrepancy, sensitivity analyses were conducted, varying (a) the cold gas velocity $(v_0 \times / \div 1.2)$, (b) the stagnation surface temperature (T_s +/-100 K), and burner surface temperature $(T_b+/-100K)$, as shown in Figure 5. These are the most important boundary conditions for the flames. Note the extent to which each of these parameters is varied is larger than the true experimental uncertainty. For example, the mass flow rates metered is accurate to within 1 to 2% and the uncertainties of T_s and T_b are well within 20 K. In any case, the impact of these uncertainties is shown to be small, and in most cases, the change in the absolute temperature is well within the experimental spatial resolution of the fine-wire thermocouple and is certainly smaller than the uncertainty caused by the emissivity of the thermocouple coating. Hence, the detailed structures of BSS flames are generally not sensitive to perturbation or uncertainty of the flow and temperature boundary conditions. They are, however, sensitive to the distance of separation between the burner and stagnation surfaces H_p .

To understand the influences of the stagnation plate and the tubular probe on the flame, we plot the temperature profiles for $H_p = 1.0$ cm in Figure 6, and compare them with the profile measured for the same flame without the presence of a probe. These temperature profiles were determined along the centerline of the flame. As seen, the tubular probe yields basically the same temperature profile as the stagnation plate, though the temperature values are somewhat higher than those with stagnation plate. This is expected since the tubular probe has a smaller surface area than the stagnation plate, and hence a smaller heat loss into the probe. In addition, the tubular probe allows for heat transfer in the radial direction perpendicular to the probe tube.

On the other hand, the temperature profile behaves quite different with and without the probe. For the "free" flame, i.e., the flame free of probe perturbation, the top panel of Figure 6 shows that the rise in the temperature in the preheat zone is somewhat slower in the "free" flame and that the maximum flame temperature is lower (by about 50 K) than the flames with the tubular probe or stagnation surface. These results are consistent with the laminar flame theory. With the probes, the flow is locally or globally stretched. With the Lewis number of the flame being close to but smaller than unity and for the current positively stretched flame, the flame speed increases as the result of flow stagnation [54]. This leads to a larger temperature gradient in the preheat zone and a somewhat higher maximum flame temperature.

An important result that Figure 6 demonstrates is the degree of flame temperature perturbation and the needs to quantify this perturbation and account for it in numerical modeling studies. Likewise, the flow stagnation also



Figure 5. Sensitivities computed for the flame temperature with respect to (a) cold gas velocity, (b) stagnation plate temperature, and (c) burner temperature. Symbols are experimental data.

causes drastic differences in the fluid velocity and hence the convective time (or roughly the reaction time) allowed for soot nucleation and mass and size growth. The top panel of Figure 7 shows that for $H_p = 1.2$ cm the axial velocity drops to zero at the stagnation surface, whereas the velocity stays constant for the "free" flame. For particles, we need to consider the thermophoretic velocity due to the temperature gradient that push the particles towards the stagnation surface. This velocity is given by [55]

$$v_T = \left(1 - \frac{6\Omega_{avg}^{(1,2)*}}{5\Omega_{avg}^{(1,1)*}}\right) \frac{\lambda}{Nk} \frac{d\ln T}{dx} , \qquad (9)$$

where the $\Omega_{avg}^{(1,i)^*}$ is the reduced collision integral and taken here in its hard-sphere and diffuse scattering limits as $\Omega_{avg}^{(1,1)^*} = 1 + \pi/8$ and $\Omega_{avg}^{(1,2)^*} = 1 + 5\pi/48$, λ the thermal conductivity, N the gas number density, and k the Boltzmann constant. The total particle velocity may be calculated by adding v_T to the convective velocity v. The combined velocity is above zero in the stagnation plate



Figure 6. Radiation-corrected, centerline temperature profiles measured in the BSS flame configuration, a tubular probe, and in a free flame. The vertical line indicates position of plate or probe. Symbols are experimental data, lines are fits to data.

case, but it is still substantially smaller than that of the "free" flame at the same distance from the burner surface. This velocity difference is directly translated into drastically different convective (or reaction) time. The convective time diverges in theory with the use of the stagnation plate. If the thermophoretic velocity is considered, the total time is finite but it exceeds 50 ms, whereas the total convective time for the "free" flame is about 20 ms at a distance of 1.2 cm above the burner surface, as shown in the bottom panel of Figure 7. These results demonstrate that the "free" flame assumption is incompatible with the experiment. This assumption leads to an over-estimated surface growth rates because the temperature is artificially high as the gas approaches the probe, and an under-estimation of particle-particle coagulation because of a shorter reaction time provided for it.

To illustrate the impact of the free-flame assumption further, we present the mole fraction profiles of major species computed for a BSS flame with $H_p = 0.8$ cm (solid lines). The species solutions for a comparable burnerstabilized "free" flame (dotted lines) are also shown for comparison. As seen, the variations of the mole fractions as a function of the distance from the burner surface are similar to those of free-flame for the first 0.5 cm, but the concentration of some species, especially H₂, undergo rapid changes in their concentration just before reaching the stagnation surface. These variations are caused by the rapid temperature changes accompanied by an increase in the reaction time near the same surface that shifts the chemical equilibrium of the burned gas. The free flame assumption is seen to be generally adequate for predicting major species in a flame with probe intrusion (in the



Figure 7. Top panel: typical velocity profiles computed with H_p =1.2 cm and compared to that of a free flame, bottom panel: convective time computed for a fluid particle as a function of height above the burner surface, comparing the free flame and the companion stagnation flame.

postflame region), but this observation does not extend to minor species, particularly those important to soot nucleation.

Figure 9 shows the benzene mole fractions computed for a range of H_p values. The "free" flame results are also shown for comparison. If a computational species probe is embedded on the stagnation surface facing the incoming gas flow, the "measurement" would yield a set of species concentrations given by the symbols in Figure 9. As seen, these "measured" values are not directly comparable with the result of a "free" flame simulation. For benzene, the "measured" species mole fraction rises at large *H* values than that of a "free" flame—an observation consistent with the traditional approach that shifts the computed species profile downstream to match the experimental profile. Here, this shift amounts to 0.25



Distance from Burner Surface, *H*(cm)

Figure 8. Typical main species profiles computed for a burner-stagnation surface separation of $H_p = 0.8$ cm (solid lines) and for a "free" flame (dotted lines).

to 0.3 cm, which is consistent with observations made in an earlier study [13]. More importantly, the benzene mole fraction for the BSS flames levels off at a value higher than the "free" flame prediction.

We note that these comparisons should be applicable, at least qualitatively, to the tubular probe. In terms of the flow field and temperature variations, a flame with a tubular probe intrusion is bound between the limits of a burner-stabilized, "free" flame and a burner-stabilized stagnation flame. For this reason, the free flame assumption employed in previous numerical simulations of soot formation [2, 21] must have resulted in local flame conditions not comparable with the experiment. As we discussed earlier, the free flame assumption overpredicts the temperature, yet the reaction time is underpredicted as compared to the experiment. This discrepancy causes the surface reaction being promoted artificially and particleparticle coagulation suppressed.

With the use of combined stagnation surface and sampling probe or the BSS flame approach discussed here, this fundamental difficulty is removed, especially considering the close agreement of the experimental and simulated temperature profiles seen in Figure 4. In principle, each sampling position gives a specific burnerto-stagnation separation or a unique BSS flame, even though the unburned-gas boundary conditions are identical among the flames tested. In comparison to the traditional approach that uses the free flame assumption (e.g., using the PREMIX) to simulate the flame, the BSS flame approach requires a separate stagnation flame simulation (e.g., using the OPPDIF) for each sampling position or H_p value. This is somewhat more time consuming than the traditional approach, but truly a minor disadvantage, considering that the free flame assumption



Figure 9. Benzene mole fraction profiles computed for several burner-to-stagnation surface distances (dotted lines) and for a burner-stabilized, "free" flame (dashed line). The symbols represent the computationally measured benzene mole fractions at the stagnation surfaces/sampling points.

is fundamentally incorrect for any probe based experiments. Moreover the BSS flame approach can reduce also the experimental effort and uncertainty. As we discussed earlier, the entire temperature profile may be computed accurately if the boundary temperatures T_b and T_s are determined reliably and used as the boundary conditions for the simulation. Hence, the spatiallyresolved temperature probing, like those presented in Figure 3, can be carried out for a significantly fewer number of spatial positions, or these measurements may be eliminated completely, if the boundary temperatures and the mass fluxes of the unburned gases are determined accurately.

We note that ideally a rigorous prediction of soot formation in the BSS flames should be made by simultaneous solution of the governing and particle transport equations. If this is not possible, a decoupled approach may be used in which soot-free flame chemistry is solved first followed by postprocessing aerosol dynamics using the flame chemistry solution.

We examined the detailed soot PSDFs collected using the tubular probe and the BSS flame approach. Since the temperature profiles are found to be similar between the two cases (see, Figure 6), we do not expect the PSDFs to be drastically different. Figure 10 shows the PSDFs at several burner-to-stagnation separations. As seen, the two probes yield very similar results, qualitatively and quantitatively. Keeping in mind that the PSDFs are sensitive to the H_p value, the discrepancy of the two probes may well be attributed to limitations in the positioning accuracy, even for the largest discrepancy observed at $H_p = 0.8$ cm.



Figure 10. PSDFs measured using the BSS flame method (symbols and solid lines) and the tubular probe (dotted lines).



Figure 11. Soot volume fraction $(D_p > 2.5 \text{ nm})$ observed, comparing the BSS flame method with the tubular probe approach. Symbols are experimental data, lines are fits to data.



Figure 12. Soot particle number densities (particle diameter $D_p > 2.5$ nm) observed, comparing the BSS flame method with the tubular probe approach. Symbols are experimental data, lines are fits to data.

Likewise, the soot volume fraction and number density (for particle diameter $D_p > 2.5$ nm) probed are well within the uncertainty of each other between the two probes, as seen in Figures 11 and 12. This again indicates that the tubular probe behaves similar to a stagnation surface. Hence the experimental data reported in earlier studies [2, 3, 5, 9, 13, 16] is better simulated as BSS flames than simple burner-stabilized flames free of downstream flow divergence and heat loss. In these studies, the burner temperatures (T_b) were carefully measured and reported. Unfortunately, the probe temperature (T_s) was not available, but based on the current study, we recommend a value of $T_s = 500\pm100$ K.

Conclusions and Summary

We presented the burner-stabilized stagnation (BSS) flame technique to probe soot formation in laminar premixed flames. The basic premise of this technique is that since a sample probe inherently perturbs the flame, it is better to let the probe to perturb the flame in a way that can be quantitatively described using a one-dimensional reacting flow code. We demonstrate that a circular plate acting as the stagnation surface (commonly referred to as the flame stabilizer) over a premixed, burner-stabilized flame allows the flame flow field and temperature to be more accurately described. The probe, embedded in the stagnation surface, draws the flame sample, dulutes it and brings the aerosol into a scanning mobility particle size for PSDF quantification. Like the earlier tubular probe approach, the BSS flame approach can resolve the evolution of PSDFs spatially, but each spatial distance results in a unique flame, which may be simulated accurately using a counterflow or stagnation reacting flow code (e.g., OPPDIF). Hence, the BSS flame approach enables direct and rigorous comparisons between the mobility size measurements and numerical simulationsa condition that has not been accomplished in the past. also shows that the previous PSDF Analysis measurements using tubular probe sampling produces flame conditions that are better simulated with a stagnation flow code than a traditional burner stabilized flame free of effects due to the sample probe.

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