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Experimental investigation of soot evolution in a turbulent non-premixed prevaporized toluene flame

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Abstract

The formation, growth, and oxidation of soot in turbulent prevaporized toluene diffusion flames stabilized on a jet-in-hot-coflow (JHC) burner are investigated in this study. Flame structure, local gas temperature as well as local soot volume fraction and primary soot particle diameter, are simultaneously detected by means of OH planar laser-induced fluorescence (PLIF), non-linear excitation regime two-line atomic fluorescence (nTLAF) of indium, and time-resolved (TiRe) laser-induced incandescence (LII), respectively. The collected data sets were used to generate joint statistics of soot properties and flame characteristics and provided new insights into the interaction of the OH layer and soot in turbulent flames. The interaction of OH and soot as a driving mechanism for soot oxidation is of particular interest as it has been proven to be challenging to model. Statistics of soot volume fraction and primary particle size in the OH layer are employed to gain deeper insights into the soot oxidation process. Mean soot volume fraction and primary soot particle size conditioned on temperature and OH signal intensity indicate that, due to differential diffusion of soot with respect to the chemical species, high soot volume fraction and primary soot particle diameter of up to 50 nm are present at low temperatures and low OH concentration. In the soot oxidation region, statistical analysis of the soot parameters disclose that clusters of high soot volume fraction mostly consist of large primary particles. Observations from instantaneous images and the presence of large primary particles inside the OH layer suggest that the oxidation is not sufficiently fast to burn the soot completely. © 2018 The Combustion Institute. Published by Elsevier Inc. All rights reserved.

Keywords: Soot in turbulent flames; Soot oxidation; Temperature; Soot particle diameter; Soot volume fraction

1. Introduction

Soot formation is an undesirable process in many practical combustion devices, such as diesel engines, direct injection gasoline engines, and gas turbines. Soot originates from fuel pyrolysis or combustion under rich conditions. It is well-known to have tremendous negative impacts on the environment and on human health [1,2]. Moreover, soot emission is an indicator of incomplete, and thus inefficient combustion [3]. Hence, a better understanding of the formation, growth, and oxidation of soot is required in order to optimize the

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thermal efficiency and meet increasingly stringent emission standards.

Most detailed experimental and numerical studies on soot have been performed in various laminar flame configurations yielding fundamental knowledge of the complex soot formation and its oxidation under well-defined conditions [4]. Although soot formation in turbulent flames is more relevant for practical combustion devices, only a limited number of studies have investigated the soot formation in turbulent flames. The strong interaction and highly non-linear correlation of soot formation and chemistry in turbulent flames requires spatially and temporally well-resolved simulations or experiments that simultaneously yield multiple quantities. Therefore, both simulation and experiment become extremely complex and costly.

Comprehensive datasets of two and three dimensional turbulent n-heptane flames have been obtained from direct numerical simulations (DNS) including chemistry of soot precursors and a higher-order statistical soot model [5,6]. These studies reveal high sensitivity of soot precursor concentration to the local dissipation rate, thereby highlighting the significant importance of the mixing field on the soot formation. In further studies, Attili et al. [7] observed the presence of soot at small mixture fractions (Z) below the stoichiometric mixture fraction (Z_{st}). It was demonstrated that these soot patches emerge through areas of local flame extinction, whereas soot crossing the burning OH layer was not obtained [7].

Recent advancement of laser diagnostics has enabled the simultaneous detection of soot and flame parameters with high spatial and temporal resolution. Park et al. [8] presented joint statistics of Z and f_v in the soot inception region by using a combined setup of laser-induced incandescence (LII), particle image velocimetry (PIV), and krypton laser-induced fluorescence (Kr-LIF). Their observations revealed that the peak soot volume fraction in the soot inception region occurs in the mixture fraction range of Z = 0.3–0.4, where the concentration of soot precursors is the highest [8].

Detailed experiments under well-defined conditions of a turbulent sooting ethylene flame have been provided by Köhler et al. [9,10]. Data of the flow field, gas temperature (T), f_v , OH as well as polycyclic aromatic hydrocarbon (PAH) distribution and local Z were obtained by means of laser diagnostics [9,10]. Gu et al. [11] have recently extended this comprehensive data set by simultaneous measurements of f_{ν} , primary particle size (d_p) , number density (N_p) , and T. Strong correlations were found for f_{ν} , d_p , and number density (N_p) , whereas these soot parameters exhibit moderate correlation with T. However, the interaction of soot and OH layers, which is particularly relevant for soot oxidation, has not been analyzed in these studies. Lee at al. [12] investigated the interaction of soot and OH in turbulent ethylene flames. It was found that the spatial overlap of soot and OH causes a reduction of f_{ν} [12]. Independent of the fuel, the same phenomenon has also been reported in turbulent methane flames by Franzelli et al. [13].

Although the strong interaction of soot and reaction surface, and its relevance for soot oxidation, have been reported for different fuels [12,13], quantitative analysis of the soot parameters (f_v and d_p) within the reaction zone is absent. Therefore, this study aims to quantitatively analyze the soot–flame interaction in a turbulent flame. Prevaporized toluene has been used as it is a simple aromatic compound and is a common component of diesel and gasoline surrogates [14,15]. While toluene has been frequently studied in laminar flames as a blending component where its dominant role on soot and PAH formation has been revealed, soot measurements in turbulent toluene flames are absent [16,17].

Simultaneous laser diagnostics were performed to measure OH, T, f_v , and d_p . Conditional data and joint statistics are presented to provide information on the soot–flame interaction.

2. Experimental setup

2.1. Burner configuration

The current burner setup is a modification of the well-established jet-in-hot-coflow burner (JHC) [18,19]. The fuel is introduced through a central insulated pipe with an inner diameter of 4.6 mm and a tapered end at the exit. The length-todiameter ratio of the fuel jet is more than 100 to ensure fully developed turbulent pipe flow at the exit. The fuel jet was surrounded by a coflow of gases produced by a lean, laminar nitrogen (N2)-diluted natural gas/air flame stabilized on a porous bed. The annular porous-bed burner with an inner diameter of 108 mm was located 25 mm upstream of the jet exit to minimize heat transfer to the central fuel jet while ensuring for a homogeneous coflow composition at the jet exit. An annular shielding flow of room air was issued from a 22 mm wide ring around the hot coflow.

The exit velocity of the shielding air flow was 0.4 m/s. The flow rates of natural gas, air, and N_2 were set by mass flow controllers to achieve oxygen mass fraction $Y_{O_2} = 0.083$ and a temperature T = 1544 K in the hot coflow stream with an exit velocity of 0.8 m/s. The fuel stream was a mixture of prevaporized toluene and N_2 as a carrier gas. The toluene mass fraction (Y_F) was kept at 0.25 and the Reynolds number at the fuel jet exit was 10,000.

Toluene was vaporized and mixed with the carrier gas in a controlled evaporator and mixer unit (CEM, Bronkhorst). After the CEM unit, the gas mixture was delivered to the burner via heated tubes. The temperature in the CEM and the tubes

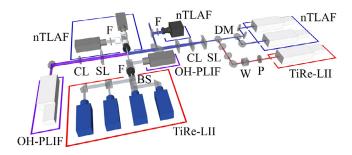


Fig. 1. Optical setup for simultaneous detection of temperature (nTLAF) and d_p as well as f_v (TiRe-LII). DM, dichroic mirror; W, wave-plate; P, polarizer; CL, cylindrical lens; SL, spherical lens; F, bandpass filter.

was maintained at 453 K to prevent fuel condensation.

2.2. Optical arrangement

Time-resolved (TiRe)-LII, OH planar laser-induced fluorescence (LIF), and non-linear two atomic fluorescence (nTLAF) are employed to simultaneously detect OH, T, f_v, and d_p. A schematic of the optical system is presented in Fig. 1.

The combined TiRe-LII and nTLAF setup was used in previous studies [11,20]. The optical system has been extended by the OH-PLIF diagnostic in the current study. The imaging sequence started with the nTLAF followed by the OH-PLIF and TiRe-LII with 1 μ s and 3 μ s delay, respectively, to avoid signal interference between the various systems. Manipulation of the soot particles by the LIF beams prior to the TiRe-LII measurements was determined to be negligible. For both LIF-techniques, the mean fluence is below 0.03 J/cm², hence far below typical laser fluence ovf 0.45 J/cm² where sublimation of soot becomes important [21].

The nTLAF of indium is used to determine the local gas temperature [22]. A laser ablation technique was used to seed indium nanoparticles to the fuel stream [23]. Two dye lasers (Quantel, TDL 90) pumped by Nd:YAG lasers (Quantel, BrilliantB and YG-980) generated the Stokes and anti-Stokes beams to excite the indium. Both beams were formed into light sheets with a height of 16 mm and a thickness of 0.25 mm, focused on the jet centerline by a cylindrical lens with a focal length of $f = 1000 \,\mathrm{mm}$. In order to avoid interference, the anti-Stokes beam was emitted 120 ns after the Stokes beam. The corresponding signals were collected by a Nikon camera lens (f/1.2). Behind the lens, the incoming light was split by a dichroic mirror. The Stokes and anti-Stokes nT-LAF signals were collected by intensified CCD (ICCD) cameras (Princeton Instruments PiMax 4) equipped with custom-made narrow bandpass filters centered at 451.4 nm (FWHM = 1.32 nm) and $410.4 \,\text{nm}$ (FWHM = 1.08 nm), respectively [24]. The gating time of the cameras was set to 30 ns to suppress the background flame luminosity. To account for spatial and temporal fluctuations of the laser sheet, part of the light sheets was deflected to a quartz cuvette filled with a suspension of distilled water and titanium oxide nanoparticles. The Mie scattering from these nanoparticles was recorded by a CCD camera (MegaPlus II, ES 4020) to correct for vertical light sheet inhomogeneity on a shot-to-shot basis. The quantification of nT-LAF was described in detail previously [11]. Briefly, in order to obtain calibration factors, the nTLAF and thermocouple measurements were performed in the hot product stream of a well-characterized flat-flame burner on a daily basis. For uncertainty estimation, nTLAF measurements were performed in a Santoro reference flame and revelead a standard deviation in the temperature measurements of \pm 120 K [11]. In the turbulent flame, the atomic indium concentration is temporally and spatially fluctuating. In regions of low atomic indium concentration, the uncertainty of the nTLAF measurements is expected to increase. In order to reduce the uncertainty, regions with low nTLAF signal (anti-Stokes signal-to-noise ratio below 3) are excluded from the post-processing.

The fundamental (1064 nm) of a Nd:YAG laser (Quantel, BrilliantB) was used to heat up the soot particles for TiRe-LII measurements. The beam was formed to a collimated light sheet with 20 mm height and 0.25 mm width by a sheet optic. In order to avoid excessive sublimation of the soot particles, the laser fluence was set to approximately 0.3 J/cm². The light sheet was focused on the jet centerline by a cylindrical lens $(f = 1000 \,\mathrm{mm})$ to minimize the beam waist increase within the detection field. The radiation of the soot particles was collected by a Nikon f/1.4 camera lens equipped with a bandpass filter (435 nm, FWHM) = 48.8 nm). Beam splitters behind the camera lens deflected the soot radiation to four ICCD camera heads (pco, HSFC Pro) to record the LII signal decay in a series of four consecutive images. The first image was instantaneously recorded with the laser pulse, the following three were detected with a delay of 80 ns, 160 ns, and 240 ns. The gating time of all four cameras was 40 ns. The prompt LII image yields the local soot volume fraction.

The intensity ratios of the sequential images were compared to computed LII signal decay rates to extract the primary particle diameter. Detailed information on the LII model are provided in [25]. To compute the particle heat transfer towards the ambient gas, the local temperature from nT-LAF measurements is employed. The modeling assumption of non-aggregated, mono-disperse soot particles results in an overestimation of d_p by a factor of two [11]. The detection limit of the particle size is 5 nm [20]. TiRe-LII measurements in a reference Santoro flame revealed a measurement uncertainty of 20% [11]. For f_{ν} determination, LII measurements were calibrated against extinction measurements at 1064 nm in a Santoro flame. The precision of the f_v measurements is $\pm 10 \%$.

For qualitative OH-PLIF measurements, OH was excited on the line $Q_1(8)$ by a 283.58 nm laser beam generated in a dye laser (LambdaPhysics, ScanMate) pumped by the second harmonic of a Nd:YAG laser (Quantel, Brilliant B). The beam was formed to a 0.3 mm wide and 15 mm high light sheet that was focused on the beam centerline by a cylindrical lens (f = 750 mm) and overlapped with the LII and nTLAF laser sheets. The OH-LIF signal was reflected by a dichroic mirror and imaged by a catadioptric f/1.2 UV camera lens equipped with a narrow bandpass filter (310 nm, FWHM = 10 nm) on an ICCD camera (Princeton Instruments, PI-MAX).

Spatial matching and calibration of all images were achieved by imaging a target in the light sheet plane. The resulting field of view had a height of 14 mm and the in–plane spatial resolution was determined to be 0.0887 mm/pixel. All images were corrected for vignetting, chip inhomogeneities and background flame luminosity. For all techniques, interference of measurement signal and flame luminosity was carefully checked and determined to be below the detection limit. Hence, background correction was performed with mean luminosity images.

3. Mean flame structure and soot distribution

The mean distributions of f_v , d_p , T, and OH averaged from 499 single images at various heights above the jet exit plane are shown in Fig. 2.

As nTLAF is limited to $T \ge 800$ K, only data above 800 K are included in the calculation of the mean temperature. For nTLAF and TiRe-LII, the beam propagates from the left to the right side of the images (- $20 \text{ mm} \le x \le 20 \text{ mm}$). The asymmetic soot distribution in the range of $60 \text{ mm} \le y \le 210 \text{ mm}$ is a result of beam steering [26]. In order to minimize errors induced by beam steering, statistical data are obtained from the half of the image on the beam entrance side. The direction of the OH-PLIF beam is from right to left. The radial asymmetry of the OH-PLIF signal also occurs at height

 $60 \,\mathrm{mm} \le x \le 210 \,\mathrm{mm}$ and it is predominantly attributed to absorption of laser energy by soot and PAH.

The mean images of OH-PLIF and T indicate a thin, distinct flame structure slightly lifted from the fuel nozzle. At the outer radial locations up to $y \le 80$ mm, OH from the hot coflow is detected. Consistent with findings in [18], this observation reveals that the influence of the coflow diminishes at approximately y = 80 mm. Hereafter, the flame develops to a non-premixed air-fuel flame. Strong interference of OH-PLIF and nTLAF signal with PAH-LIF is observed at the jet centerline at a height of $40 \text{ mm} \le y \le 100 \text{ mm}$.

The first measurable soot was detected at $y = 70 \,\mathrm{mm}$. The mean images of f_v and d_p reveal the characteristic regions of soot formation $(70 \,\mathrm{mm} \le y \le 170 \,\mathrm{mm})$, soot transport by turbulence $(195 \,\mathrm{mm} \le y \le 290 \,\mathrm{mm})$, and soot oxidation $(320 \,\mathrm{mm} \le y \le 350 \,\mathrm{mm})$ that have been also reported previously [12,13].

Upstream of y = 130 mm, d_p is below the detection limit of 5 nm. Further downstream, both f_v and d_p increase in the soot formation region. Meanwhile, a core of low temperature is apparent at the jet centerline. A comparison of f_v , d_p , T, and OH images indicates that the formation of soot is confined to a narrow layer on the fuel rich side of OH with elevated temperatures.

For $y \ge 195$ mm, the soot is transported by the turbulent flow field resulting in a wide radial distribution of soot. Within 245 mm $\le y \le 290$ mm, f_v and d_p remain almost constant. The peaks of f_v and d_p are observed on the jet centerline and decrease with increasing distance from the centerline. In contrast, the temperature and OH signal are lowest on the jet centerline and increase towards the outer region.

In the range from y = 315 mm to 350 mm, the mean f_v and d_p are significantly reduced and the distribution is narrowed towards the centerline due to enhanced soot oxidation.

In order to provide further information on the flame structure, a one-dimensional calculation is performed in physical space with unity Lewis numbers using FlameMaster [27] and the kinetic mechanism presented in [14]. As shown by Attili et al. [28], imposing unity Lewis numbers is the most appropriate choice to capture the structure of turbulent non-premixed flames. The oxidizer is air and the fuel stream consists of an N₂-toluene mixture ($Y_F = 0.25$). According to [29], a characteristic strain rate is approximated based on the inverse of the global residence time to be 25 s⁻¹.

Fig. 3 illustrates the profiles of T, dimer production rate as a measure for soot formation, naphthalene (A2) as a representative soot precursor, and OH mass fraction in the mixture fraction space. It is evident that soot precursors are present at $Z \ge Z_{st}$ with the maximum mass fraction at Z = 0.4 and temperatures of $1220 \text{ K} \le T \le 1600 \text{ K}$. The dimer

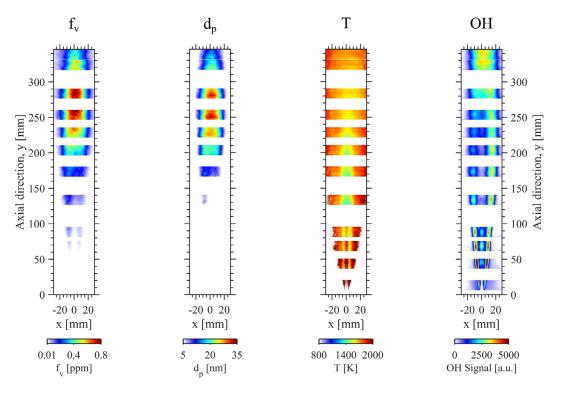


Fig. 2. Averaged distribution of f_v , d_p , T, and OH at various heights above the burner.

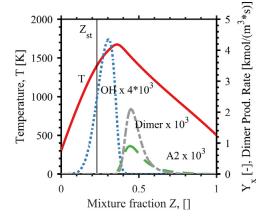


Fig. 3. Profiles of T, $Y_{\rm OH}$, Y_{A2} , and dimer production rate obtained from flamelet simulation. The black vertical line indicates Z_{st} . Fuel side: $Y_F = 0.25$ balanced with N_2 , $T_F = 500$ K; Oxidizer side: $Y_{\rm O_2} = 0.233$ balanced with N_2 , $T_{\rm O_2} = 298$ K. Strain rate is $25 \, {\rm s}^{-1}$.

production rate follows the same trend, whereas the peak rate is obtained at slightly higher Z. In contrast, OH is solely present in a narrow range at high temperatures and around Z_{st} . This characteristic makes OH a good marker of the flame region. The profiles indicate that OH and soot do not over-

lap. Additional flamelet computations, with different strain rates in the range expected in the turbulent flame, revealed that the flame structure observed in Fig. 3 is not affected (not shown).

4. Statistical analysis of soot-OH interaction in turbulent flame

Previous experimental studies of turbulent sooting flames have qualitatively reported the spatial overlap of OH and soot [12,13]. The simultaneous detection of reaction zone location and quantitative soot characteristics enables a detailed analysis of soot parameters in the OH layer. To illustrate the soot–OH interaction, Fig. 4 depicts instantaneous representative images of f_{ν} , d_{p} , and T from the soot formation, mixing-dominated, and oxidation zone that were described in Section 3. The iso-contours of two OH signal intensities are plotted as gray and black lines in the instantaneous images.

In the soot formation region (165 mm $\le y \le$ 180 mm), soot is concentrated in small segregated clusters. Regions without soot are particularly present in low-temperature regions around the jet centerline. The temperature increases towards the OH layer. Soot clusters with f_v of up to 1.5 ppm and d_p up to 40 nm are found close to the OH layer

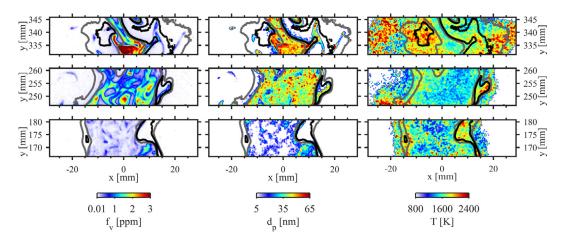


Fig. 4. Instantaneous images of f_v , d_p , and T overlaid with OH iso-contours (2000 counts: gray line, 5000 counts: black line).

on the fuel rich side, whereas soot is absent in the OH layer and on the lean side.

At 245 mm $\leq y \leq 260$ mm, the local f_v and d_p are significantly higher compared to the formation zone, exceeding values of 2.5 ppm and 60 nm, respectively. The soot patterns follow the flow and are stretched and wrapped by turbulent vortices resulting in wide distribution and high spatial gradients of f_{ν} . Soot is also present in areas of lower temperatures ($T \le 1500 \,\mathrm{K}$). This is due to the very low diffusivity of soot particles compared to that of temperature. This differential diffusion promotes a significant transport of soot in the temperature space [6]. Although the soot is still predominantly distributed in elongated structures in the rich side of the flame, discrete ligaments of soot are present in the OH layer and even on the lean side of the non-premixed flame.

At the highest axial position, the soot appears in confined regions with $f_v \ge 3$ ppm and $d_p \ge 60$ nm in the zone between the OH layers where the local temperature is low. Similar to observations in the mixing dominated area, soot patterns with $5 \le d_p \le 35$ nm penetrate the OH layer. The soot is mostly oxidized in the OH layer, nevertheless, small soot clusters are found on the lean side of the OH layer.

The presence of soot in the OH layer is assessed by joint probability density functions (PDF) of f_v and OH signal intensity as well as d_p and OH signal intensity presented in Fig. 5. The mean of f_v and d_p conditioned on OH are also included in the plots. The samples are obtained from 499 single images and are spatially averaged over 0.45 mm \times 0.45 mm.

Figure 5 shows that, in the formation region, soot is present at locations where the OH signal is in the range of background noise, whereas soot is absent at high OH signal intensities. This finding is consistent with the mean images and the flamelet simulation in Fig. 3. The conditional means of f_v

and of d_p reveal that, soot in this region is still formed of primary particle of rather small size. With increasing axial distance, f_v and d_p increase remarkably. In areas of low OH signal, f_v fluctuates by more than an order of magnitude from values below 0.1 ppm to above 1 ppm, and d_n ranges from the detection limit up to diameters of approximately 60 nm. It is worth noting that the mean of f_{ν} conditioned on OH does not coincide with the most probable value [30]. This is a characteristic feature of highly intermittent fields. The probability of soot and OH overlap is also significantly increased compared to the soot formation zone. Due to soot oxidation in the OH layer, the mean f_{ν} and d_p conditioned on the OH gradually decrease as the OH signal increases. In the soot oxidation region, the probability of $f_v \ge 0.2$ ppm is considerably reduced compared to the mixing dominated zone. Meanwhile, the PDF of d_p indicates the presence of primary particles with d_p in the range of 25 nm to 50 nm at low OH signal intensities. Compared to the mixing dominated region, the conditional mean of f_v and d_p increases further at low OH signals, hence upstream of the reaction zone. Moreover, the joint PDF of d_p and OH demonstrates that in addition to small particles, soot particles of significant diameter as large as $d_p \ge 35 \,\mathrm{nm}$ are present in the OH layer.

The simultaneously detected local gas temperature provides further information regarding the local interaction of soot and flame. As indium is oxidized under high temperatures and the presence of oxygen, the nTLAF measurements yield the temperature on the fuel rich side. The mean values of f_v and d_p conditioned on temperature and OH signal intensity in the soot oxidation region are shown in Fig. 6.

Figure 6 shows that at the region of OH signal below 2000 and temperature below 1600 K, f_v is most likely to be higher than 1.5 ppm. Meanwhile,

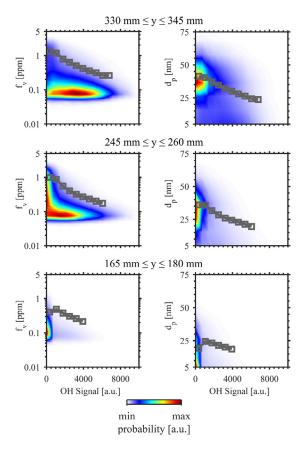


Fig. 5. Joint PDFs of f_v and OH as well as d_p and OH at three locations. The gray line represents the mean of f_v and d_p conditioned on OH signal.

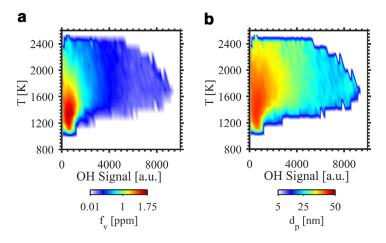


Fig. 6. Mean of f_v (a) and d_p (b) conditioned on T and OH in the soot oxidation region (330 mm $\leq y \leq$ 345 mm).

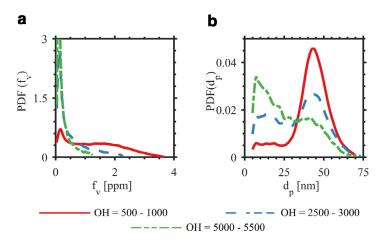


Fig. 7. PDFs of f_v (a) and d_p (b) conditioned on OH signal obtained at 330 mm $\leq v \leq 345$ mm.

 d_p of most soot particles is higher than 30 nm. Considering that nTLAF yields the temperature on the fuel rich side, the accumulation of soot occurs on the fuel rich side of the OH layer. This characteristic has been already reported by Pitsch et al. [31] and has been also observed by Attili et al. [7], where it was shown that, due to the differential diffusion of soot with respect to the chemical species, a strong relative movement of the soot with respect to the chemical species, temperature and mixture fraction exists resulting in an accumulation of soot on the fuel rich side. The strong effect of the differential diffusion between soot, temperature, and chemical species in the turbulence regimes also explains the much wider range of temperature at which soot is observed in the turbulent flames during the experiment compared to the laminar cal-

Figure 6 reveals that the overlap of OH and soot occurs in a wide temperature range, where f_v and d_p gradually decrease with increasing OH signal intensity due to soot oxidation. The conditional mean of d_p indicates the appearance of soot primary particles with $d_p \ge 15$ nm at high values of the OH signal. Although the flame was non-smoking, the joint PDFs and the pockets of soot on the lean side of the OH layer in Fig. 4 suggest that the soot oxidation rate is not sufficient to completely burn the soot transported by turbulent fluctuations through the flame. Different to the DNS stduy conducted by Attili et al. [7], where soot only emerged through locally extinguished flame patches, the soot might cross the OH layer before being completely oxidized.

To analyze the distribution of f_v and d_p on the fuel rich side and inside the OH layer, Fig. 7 presents the PDFs of f_v and d_p conditioned on three different OH signal ranges.

As the overlap of soot and OH increases, the reduction of soot is apparent in the PDFs of f_{ν}

(Fig. 7a). With increasing OH signal, the distribution of f_v narrows and a distinct peak is obtained at $f_v = 0.25$ ppm. The distribution of d_p in Fig. 7b demonstrates that mostly particles with $d_p \ge 25$ nm are present on the fuel rich side of the OH layer, where OH concentration is small. With increasing soot-OH overlap, a bimodal distribution of d_p is observed, where the occurrence of large d_p decreases and more small primary particles are present. The physical mechanism causing the bimodality cannot be conclusively clarified. With increasing OH signal, the soot oxidation shifts the peak ratio towards small primary particles with d_p of approximately 10 nm. Considering the slow reduction of larger primary particles, the rate for soot oxidation might be also rather slow. Noteworthy is, that this observation is vastly different to the typically used models which assume that soot is rapidly oxidized with the abundance of OH and O_2 [7].

5. Conclusions

Simultaneous, laser-based measurements of f_{ν} , d_p , T, and OH have been performed in a turbulent prevaporized N₂ diluted toluene non-premixed flame with a jet Reynolds number of 10,000 and Z_{st} = 0.23 at various axial positions. The soot-flame interaction has been quantitatively analyzed in the three characteristic regions of the soot formation and oxidation process. In the turbulent flame, soot is formed at locations of high temperature in the absence of OH on the fuel side which is in good agreement with one-dimensional flamelet simulations. With increasing distance from the burner, f_{ν} and d_p are increased and soot is present in a wide temperature range. In the soot oxidation region, clusters of high f_{ν} and d_{p} are present on the fuel rich side. The driving mechanism for the accumulation of soot on the rich side is assumed to be the differential diffusion of soot. It was found that the clusters of high f_v mostly consist of large primary particles. Inside the OH layer, a bimodal distribution of small and large d_p reveals a limited oxidation of soot particles. This finding and observations of soot clusters on the lean side of the OH layer suggest that soot is not completely oxidized in the OH layer.

Acknowledgments

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References

- [1] A. Seaton, D. Godden, W. MacNee, K. Donaldson, Lancet 345 (8943) (1995) 176–178.
- [2] M. Heal, P. Kumar, R. Harrison, Chem. Soc. Rev. 41 (19) (2012) 6606–6630.
- [3] D.O. Lignell, J.H. Chen, P.J. Smith, Combust. Flame 155 (1) (2008) 316–333.
- [4] H. Wang, Proc. Combust. Inst. 33 (1) (2011) 41-67.
- [5] F. Bisetti, G. Blanquart, M.E. Mueller, H. Pitsch, Combust. Flame 159 (1) (2012) 317–335.
- [6] A. Attili, F. Bisetti, M.E. Mueller, H. Pitsch, Combust. Flame 161 (7) (2014) 1849–1865.
- [7] A. Attili, F. Bisetti, M.E. Mueller, H. Pitsch, Proc. Combust. Inst. 35 (2) (2015) 1215–1223.
- [8] O. Park, R.A. Burns, O.R. Buxton, N.T. Clemens, Proc. Combust. Inst. 36 (1) (2017) 899–907.
- [9] M. Köhler, K.P. Geigle, W. Meier, B.M. Crosland, K.A. Thomson, G.J. Smallwood, *Appl. Phys. B* 104 (2) (2011) 409–425.
- [10] M. Köhler, K.-P. Geigle, T. Blacha, P. Gerlinger, W. Meier, Combust. Flame 159 (8) (2012) 2620–2635.

- [11] D. Gu, Z. Sun, B.B. Dally, P.R. Medwell, Z.T. Alwahabi, G.J. Nathan, *Combust. Flame* 179 (2017) 33–50.
- [12] S.-Y. Lee, S.R. Turns, R.J. Santoro, Combust. Flame 156 (12) (2009) 2264–2275.
- [13] B. Franzelli, P. Scouflaire, S. Candel, *Proc. Combust. Inst.* 35 (2) (2015) 1921–1929.
- [14] L. Cai, H. Pitsch, Combust. Flame 162 (5) (2015) 1623–1637.
- [15] X. Liu, H. Wang, X. Wang, Z. Zheng, M. Yao, Appl. Energy 189 (2017) 187–200.
- [16] S. Park, Y. Wang, S.H. Chung, S.M. Sarathy, Combust. Flame 178 (2017) 46–60.
- [17] B. Choi, S. Choi, S. Chung, *Proc. Combust. Inst.* 33 (1) (2011) 609–616.
- [18] P.R. Medwell, P.A. Kalt, B.B. Dally, Combust. Flame 148 (1) (2007) 48–61.
- [19] J. Ye, P.R. Medwell, B.B. Dally, M.J. Evans, Combust. Flame 171 (2016) 173–184.
- [20] K.K. Foo, Z. Sun, P.R. Medwell, Z.T. Alwahabi, B.B. Dally, G.J. Nathan, *Combust. Flame* 181 (2017) 270–282.
- [21] R. Vander Wal, T. Ticich, A. Stephens, Appl. Phys. B 67 (1) (1998) 115–123.
- [22] P.R. Medwell, Q.N. Chan, P.A.M. Kalt, Z.T. Alwahabi, B.B. Dally, G.J. Nathan, *Appl. Opt.* 48 (6) (2009) 1237–1248.
- [23] P.R. Medwell, Q.N. Chan, B.B. Dally, S. Mahmoud, G.F. Metha, G.J. Nathan, *Appl. Phys. B* 107 (3) (2012) 665–668.
- [24] D. Gu, Z. Sun, G.J. Nathan, P.R. Medwell, Z.T. Alwahabi, B.B. Dally, *Combust. Flame* 167 (2016) 481–493.
- [25] Z. Sun, D. Gu, G. Nathan, Z. Alwahabi, B. Dally, Proc. Combust. Inst. 35 (3) (2015a) 3673–3680.
- [26] Z.W. Sun, Z.T. Alwahabi, D.H. Gu, S.M. Mahmoud, G.J. Nathan, B.B. Dally, *Appl. Phys. B* 119 (4) (2015b) 731–743.
- [27] H. Pitsch, A C++ Computer Program for 0D Combustion and 1D Laminar Flame Calculations, *Tech*nical Report, RWTH Aachen University, 1998.
- [28] A. Attili, F. Bisetti, M.E. Mueller, H. Pitsch, Combust. Flame 166 (2016) 192–202.
- [29] S.R. Turns, F.H. Myhr, *Combust. Flame* 87 (3) (1991) 319–335.
- [30] F. Bisetti, A. Attili, H. Pitsch, *Philos. Trans. R. Soc.* A 372 (2022) (2014) 20130324.
- [31] H. Pitsch, E. Riesmeier, N. Peters, Combust. Sci. Technol. 158 (2000) 389–406.