

A Progress Review on Soot Experiments and Modeling in the Engine Combustion Network (ECN)

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ABSTRACT

The 4th Workshop of the Engine Combustion Network (ECN) was held September 5-6, 2015 in Kyoto, Japan. This manuscript presents a summary of the progress in experiments and modeling among ECN contributors leading to a better understanding of soot formation under the ECN "Spray A" configuration and some parametric variants. Relevant published and unpublished work from prior ECN workshops is reviewed. Experiments measuring soot particle size and morphology, soot volume fraction (f_v), and transient soot mass have been conducted at various international institutions providing target data for improvements to computational models. Multiple modeling contributions using both the Reynolds Averaged Navier-Stokes (RANS) Equations approach and the Large-Eddy Simulation (LES) approach have been submitted. Among these, various chemical mechanisms, soot models, and turbulence-chemistry interaction (TCI) methodologies have been considered.

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INTRODUCTION

High-pressure, turbulent, compression-ignition sprays are among the most challenging research areas in combustion science encompassing numerous disciplines over a wide range of length and time scales. Topics of interest include internal nozzle flows with cavitation, multi-phase flows with liquid breakup, atomization, and vaporization in potentially trans- to super-critical regimes, turbulent mixing, gas-phase chemical kinetics, soot particle chemistry, aerosol dynamics, and radiation.

Given this broad range of disciplines, clearly it is advantageous to coordinate efforts among researchers toward a common set of conditions. Such a targeted effort will undoubtedly lead to more rapid progress in understanding the complexities of such systems and leveraging physics toward improved designs through computational fluid dynamics (CFD) models for engine sprays. With this purpose in mind, the Engine Combustion Network (ECN) was founded in 2010 as a means of coordinating collaborative experimental research efforts among international institutions to provide highly vetted targets for engine spray CFD model development and validation.

Prior experimental datasets compiled by ECN participants have been published on characterizing the various combustion vessel boundary conditions [1], injector geometry and hydraulic behavior [2,3,4], and spray development, vaporization, and combustion [5]. The present work provides a review of the experimental and simulation contributions from ECN participants on the topic of soot in high-pressure combusting spray flames.

EXPERIMENTAL CONTRIBUTIONS

High-pressure spray experiments targeting soot measurements have been conducted at Sandia National Labs (SNL, USA), IFP Energies Nouvelles (IFPEN, France), and Universitat Politècnica de Valencia (UPV, Spain). Soot collected at IFPEN was analyzed using Transmission Electron Microscopy (TEM) at Meiji University (MU, Japan). A brief description of the high-pressure combustion vessels and diagnostics used at the various institutions is provided below. The ECN “Spray A” condition (see Table 1) was the primary target of the experimental work; however, parametric variations in ambient temperature, density, and oxygen concentration were also considered. The fuel injectors belong to a family of nominally identical single-hole injectors supplied to the ECN by Bosch.

Experimental Vessels

Constant Volume Vessel

SNL and IFPEN use constant-volume, high-pressure, high-temperature spray vessels. The vessels achieve thermodynamic conditions relevant to modern compression ignition engines prior to liquid fuel injection via spark ignition of a combustible gas mixture. While the time needed for gas handling between fuel injections limits the experimental repetition rate to one injection every few minutes, the constant volume design can achieve temperature and

pressure conditions exceeding the current capabilities of higher repetition rate constant pressure designs. The design and operation of the SNL and IFPEN vessels have been described in detail previously [6,7,8]. For the experiments presented here, IFPEN and SNL used injectors #210678 and #210370, respectively, from the family of ECN Spray A injectors.

Constant Pressure Vessel

UPV conducted soot experiments in a constant pressure vessel, which allows for a higher experimental repetition rate, but is limited to a peak temperature of 1000 K. Constant pressure/temperature conditions are maintained by a flow of heated and pressurized gases. Reactants and/or products from each injection are flushed from the system by the flow of fresh gases permitting repeated injections at approximately 1 Hz. The constant pressure vessel was designed at the institute for combustion technology (ITV) of RWTH Aachen University and has been described in detail by Payri et al. [9]. Injector #210675 from the family of ECN Spray A injectors was employed at UPV.

Table 1. ECN Spray A Condition

Fuel	n-dodecane
Injection Pressure	150 MPa (1500 bar)
Orifice Diameter	90 μm (nominal)
Ambient Temperature	900 K
Ambient Density	22.8 kg/m^3
Ambient O ₂	15% vol.

Soot Diagnostics

Planar Laser-Induced Incandescence (PLII)

IFPEN used single-shot PLII calibrated with point-based, helium-neon (HeNe) laser line-of-sight extinction measurements to quantify the soot volume fraction (f_v) in the central plane of high-pressure combusting sprays [10]. PLII images were acquired at different timings after the start of injection (ASOI) to assess the temporal evolution of soot formation. Several PLII images were also acquired at the same time ASOI in repeated experiments during the quasi-steady period of the spray to generate an ensemble average representation of the quasi-steady f_v . Parametric variations around the nominal Spray A condition in ambient temperature and oxygen concentration were considered to assess their effects and provide additional target data for engine spray combustion modeling efforts.

Diffused Back-Illumination Extinction Imaging (DBIEI)

Time-resolved images of soot optical thickness (KL) in high-pressure spray flames were acquired at Sandia National Labs (SNL) and Universitat Politècnica de Valencia (UPV) using a diffused, back-illumination, extinction imaging (DBIEI) diagnostic.

The optical setup consists of a high-intensity, diffused illumination source and a high-speed camera equipped with the necessary band-pass and neutral density filters. The original diffused illumination source used at SNL [2,11] followed the design of

Ghandi and Heim [12], and this configuration was also implemented in the DBIEI measurements presented here from UPV; however, a new optical configuration characterized by a much larger region of spatial uniformity and more diffused rays at the imaging plane has been developed at SNL as described in Westlye et al. [13]. The improved optical setup reduces perceived attenuation due to beam steering. Results from SNL using both the original and improved illumination setup are included here.

As a line-of-sight diagnostic, DBIEI maps of KL must be tomographically reconstructed to obtain the dimensional extinction coefficient, K . The dimensional extinction coefficient can then be related to the radial distribution of f_v at the central spray plane through calculations using the Rayleigh-Debye-Gans approximation for fractal aggregates (RDG-FA) as described in Manin et al. [11]. Tomographic reconstruction of projected data from a non-axisymmetric source can be accomplished using the inverse Radon transform. The accuracy of the reconstruction is greatest when projected data are available from multiple viewing angles. Because only a single view of the spray event is available in these DBIEI experiments, the additional projected views are generated by weighted interpolation. The uncertainty introduced into the f_v profiles because of this interpolation is an inherent weakness of the DBIEI diagnostic.

On the other hand, KL is proportional to the path-integrated mass of particles in the attenuating medium. Thus, the DBIEI diagnostic can provide a more accurate measure of the total soot mass than a planar diagnostic such as PLII. A determination of total soot mass from f_v measurements by PLII requires an assumption of axisymmetry followed by integration of the revolved f_v distribution.

With a sufficiently long injection duration, the turbulent combustive sprays achieve quasi-steady behavior. By time-averaging the temporally resolved KL images over the quasi-steady period and subsequently ensemble averaging data from repeated experiments, an axisymmetric map of the KL field is obtained, which can be tomographically reconstructed to yield K , and subsequently f_v , using the inverse Radon transform mentioned above or the simpler inverse Abel transform. For the SNL experiments presented here, quasi-steady soot measurements were performed with a 6-ms injection duration. Time averaging began after the spray head passed outside the field of view at approximately 2.5 ms and continued until just prior to the end of injection at 5.8 ms.

Finally, Manin et al. [11] demonstrated the use of two different LED wavelengths in an alternating fashion to extract information about the optical properties of the spray flame soot. Specifically, the dispersion exponent can be derived from the ratio of extinction measured at two different wavelengths. The dispersion exponent can then be related to the carbon-to-hydrogen ratio (C/H), which provides some indication of the soot maturity. We must add that for incident wavelengths shorter than about 450 nm, molecular absorption by large polycyclic aromatic hydrocarbons (PAH) may be a source of uncertainty in such measurements.

Particle Sampling and Transmission Electron Microscope (TEM) Analysis

Soot particles formed during high-pressure spray combustion were collected on TEM grids by IFPEN and shipped to MU for TEM analysis [14]. During sampling, the probe was positioned slightly off the main jet flow axis such that only half of the jet impinged onto the probe, while the other half of the spray was minimally disturbed. The particle-laden flow entered the probe through a 1-mm diameter hole, and soot particles were deposited onto the TEM grid. The restricted flow passage ensures flame quenching and also protects the grid against excessive heating [15, 16].

MU investigated the samples using high-resolution transmission electron microscopy (HR-TEM: JEM-2100F, operating voltage: 200 keV, point resolution: 0.19 nm). The TEM grids were interrogated without any thermal or chemical pre-treatment to avoid altering properties that may be characteristic of soot particles produced in Diesel flames. More than 25 TEM images were taken at five different locations on each TEM grid using a magnification of 20,000 $\times$. The time between the sampling and the TEM investigation was slightly longer than one month; however, Aizawa et al. [16] reported that such a delay does not impact the measured results.

MODELING CONTRIBUTIONS

Computational Fluid Dynamic (CFD) modeling of soot formation and oxidation in high-pressure combustive sprays was performed by Argonne National Laboratory (ANL, USA), Politecnico di Milano (PM, Italy), ETH Zurich (ETHZ, Switzerland), the University of Wisconsin (UW, USA), and the University of New South Wales (UNSW, Australia). A brief description of the different approaches is provided below.

ANL

ANL used the commercial software “Converge” to perform simulations using a dynamic structure large-eddy simulation (LES) model. The calculations use the traditional Lagrangian parcel Eulerian-fluid approach incorporating models for spray injection, atomization and breakup, turbulence, droplet collision, and coalescence. The Favre-averaged Navier-Stokes equations in conjunction with the LES-based turbulence model describe the gas-phase flow field. The dynamic structure model was shown to most accurately capture the experimental mixture fraction relative to other sub-grid scale models [17], and the initial turbulence levels were set based on experimental measurements. The chemical kinetic model for n-dodecane combustion includes 105 species and 420 reactions [18], while combustion is treated with a well-mixed model. Soot was modeled using the methodology of Hiroyasu [19] with acetylene (C_2H_2) as the soot precursor. Additional details are available in Refs. [20,21,22,23].

Politecnico di Milano, Italy (PM)

PM used the opensource CFD platform OpenFOAM®, together with a set of internally developed solvers and libraries referred to as Lib-ICE [24]. The gas phase is described by the unsteady Reynolds-averaged Navier-Stokes (URANS) formulation. The mass

momentum, and energy equations are solved for a compressible, multi-component flow using the second-order, unstructured finite-volume method supporting polyhedral cells. The standard k - ϵ model was used for turbulence. In order to match the experimental vapor penetration, the constant $C_{\epsilon 1}$ was changed from 1.44 to 1.55. This practice is quite common in simulation of gas jets and is referred to as the "round jet correction". Assigning a value of 1.55 to $C_{\epsilon 1}$ ensures a very good matching between computed and experimental vapor penetration data in a wide range of operating conditions. Concerning initial turbulence data, turbulence intensity and the integral length were set to 0.25 m/s and 1 mm, respectively. Such data are considered representative of constant volume vessel conditions after the pre-burn event. Pressure and velocity equations are coupled by the pressure-implicit method for pressure-linked equations (PIMPLE) algorithm. The discrete droplet method (DDM) is used to compute the evolution of the liquid fuel spray. Jet and droplet breakup are computed by the KHRT model, which accounts for both Kelvin-Helmholtz (KH) and Rayleigh-Taylor (RT) instabilities. The turbulence-chemistry interaction is modeled with the multiple representative interactive flamelet (mRIF) model, using a skeletal mechanism with 54 species and 269 reactions [25] for the n -dodecane chemistry. Soot inception, coagulation, surface growth and oxidation are described as in Moss et al. [26] with acetylene (C_2H_2) as the main precursor.

ETH Zurich, Switzerland (ETHZ)

ETHZ used the commercial CFD software STAR-CD v4.22 coupled with an in-house combustion code. A RANS approach was employed with the standard k - ϵ model. A round-jet correction was applied to this model by modifying the constant $C_{\epsilon 1}$ to 1.52 to capture the penetration length as closely as possible. Initial conditions for turbulence were based on a turbulent fluctuation and length scale of 1 m/s and 0.1 mm respectively, representative of near-quiescent conditions before injection. To incorporate the liquid-phase fuel spray, a Lagrangian approach was used with the blob model for atomization and the Reitz-Diwakar [27] model for droplet secondary breakup. Turbulence-chemistry interaction was accounted for by the conditional moment closure (CMC) approach, previously applied for simulation of various high-pressure spray test rigs [28,29,30,31,32]. For soot modeling, the semi-empirical two-equation model of Leung et al. [33] was employed within the CMC framework. The model considers inception (from C_2H_2), surface growth, oxidation (by O_2 and OH) and coagulation. For Spray A, the simplified n -dodecane mechanism from Luo et al. [18] was applied using in-situ adaptive tabulation (ISAT) [34] to reduce computational effort. An extended CMC model was employed to account for multiple injections [31].

University of Wisconsin, USA (UW)

UW used the open source code KIVA3V release 2 [35] to provide the CFD platform for the simulations. The generalized RNG k - ϵ turbulence model proposed by Wang and Reitz [36] was applied to improve the prediction of fuel mixture distribution under non-reacting condition. The spray breakup process was predicted by the KH-RT instability model [37, 38]. A reduced n -dodecane-PAH

mechanism with 100 species and 432 reactions was used to predict the n -dodecane combustion and PAH formation processes [39] using a well-mixed model. The multi-step soot model developed by Vishwanathan and Reitz [40] was applied to predict the soot formation and oxidation processes. In this modeling approach, soot particle inception occurs with the formation of pyrene ($C_{16}H_{10}$), then the soot particle undergoes the C_2H_2 assisted surface growth, coagulation, and soot is oxidized by O_2 and OH radicals and PAH condensation on the soot particle surface. More details about this modeling approach can be found in [40, 41].

University of New South Wales, Australia (UNSW)

UNSW employed the commercial software "Fluent" to perform simulations using the transported probability density function (tPDF) method [42] with the interaction by exchange with the mean (IEM) mixing model. The unsteady RANS equations coupled with the realizable k - ϵ turbulence model were used to provide turbulence information to the tPDF solver. The $C_{\epsilon 1}$ constant from the turbulence model has been augmented from 1.44 to 1.52 to match the experimental vapor penetration. The field has been initialized as quiescent with a turbulent velocity and integral length scale of 0.1 m/s and 2 mm, respectively. These initial values were found to have a minor influence on the spray development. A reduced n -dodecane chemical mechanism including 54 species and 269 reactions [25] was used. The soot formation model is identical to that of ETHZ using an acetylene-based two-equation model [33], which accounts for simultaneous soot particle inception, surface growth, coagulation and oxidation by O_2 and OH. Additional details are available in [43, 44]. For selected test cases, simulations have also been performed with the well-mixed model for chemical reaction rates - neglecting turbulent fluctuations - to directly study the influence of TCI.

RESULTS AND DISCUSSION

To support the purposes of the ECN, the present work reports on experimental and modeling results that have been prepared for prior ECN workshops. New results not yet available in the literature are presented and discussed along with those previously published.

The results presented below are organized as follows. First, recognizing the strong coupling between mixing and ignition processes with soot formation, we present a comparison of experimental and simulated vapor penetration, ignition delay times, and lift-off length. As a necessary prerequisite for optical soot measurements, we then present TEM results relating to soot morphology and the determination of parameters required to relate extinction measurements to soot volume fraction. Of particular importance is the finding that the primary particle diameter of soot particles formed in high pressure spray flames is significantly smaller than those observed in atmospheric pressure flames. Following the discussion of the TEM data, we present experimental and modeling results for the early soot formation transients. Quasi-steady soot results are then presented for Spray A and a few parametric variants. Lastly, we present experimental and modeled results for transient soot formation at the Spray A condition with multiple injections.

Penetration, Ignition Delay, and Lift-off Length for Model Validation

There is significant utility in time-resolved measurements of transient processes in high-pressure combustions sprays as validation targets for CFD models. For example, before trusting any comparison of modeled soot results to experiments, the models should demonstrate that they can accurately capture the experimentally determined vapor penetration and ignition delay time. For long injection duration cases, a quasi-steady lift-off length is also established providing yet another relevant model validation target. This section briefly compares the simulated vapor penetration, ignition delay time, and quasi-steady lift-off length against experimental data available from the ECN community.

Vapor Penetration

Pickett and coworkers [45] highlighted the strong relationship between spray development and global mixing. As such, jet penetration can be used as a metric to evaluate the mixing fields across modeling predictions and compare them to the experimental results. The experimental vapor penetration under the non-reacting Spray A condition is presented in Fig. 1 along with the simulated results. The experimental curve (solid black) represents the average of several injections carried out with nozzle #210370 at SNL and nozzle #210675 at CMT. These two Spray A nozzles have similar orifice diameters and discharge (C_d) and area contraction (C_a) coefficients and were the targets of the modeling efforts presented in this work. The standard deviation of these two datasets is too small to be resolved on this figure; however, for reference, the grey shaded region represents the standard deviation from the mean of all non-reacting Spray A vapor penetration data available on the ECN data archive. This includes data from five different institutions using five different nozzles. It is noted that the average penetration of #210370 and #210675 falls on the upper bound of the grey shaded region, which is consistent with the slightly larger orifice diameters of #210370 and #210675 relative to the other Spray A nozzles.

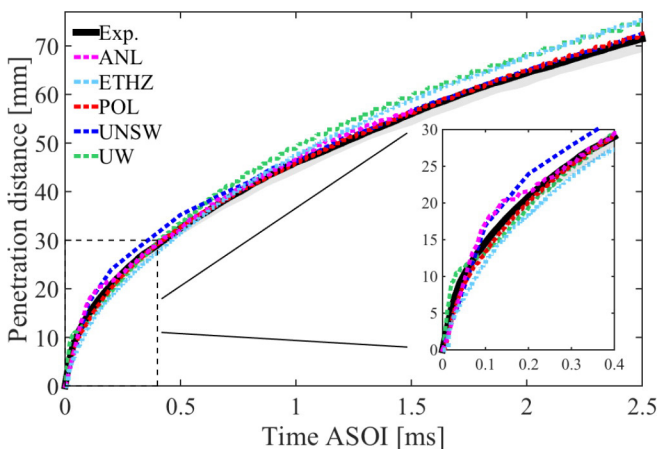


Figure 1. Experimental and modeled non-reacting vapor penetration of an n-dodecane spray issuing from a 90- μ m (nominal) orifice at 1500 bar (150 MPa). Ambient conditions: 900 K, 22.8 kg/m³, 0% O₂.

At timings prior to ignition for the reacting Spray A case (i.e., < 0.4 ms ASOI) the ETH and POL models most accurately reproduce the experimental penetration data (see Fig. 1 inset), with the UW and

ANL models initially penetrating too quickly before relaxing to the experimental data at 0.05 ms and 0.2 ms, respectively. With the exception of the UW result, which shows greater penetration than the experiment after approximately 0.5 ms, the models all appear to reproduce the experimental non-reacting vapor penetration at later times relevant to soot formation at the Spray A condition. We note, however, that the ETHZ vapor penetration diverges slightly from the experimental data near 1.1 ms, which is after soot formation begins at the Spray A condition.

Ignition Delay

Four institutions participating in the ECN have measured experimental ignition delay times at the Spray A baseline condition. For the parametric variants in temperature at 750 K, 800 K, and 850 K, two institutions have contributed data. At 1000 K and above, only SNL has contributed ignition delay data. The experimental and simulated ignition delay times as a function of ambient temperature at an ambient density of 22.8 kg/m³ and an ambient oxygen concentration of 15% are presented in Fig. 2. The standard error is included for datasets combined across institutions where applicable. At ambient temperatures of 1000 K and above, ignition delay times are highly repeatable and the standard error cannot be resolved in the figure. The ignition delay values at the Spray A condition are also tabulated in the figure for reference and the experimental value includes the standard error. Note that the ANL ignition delay time at 900 K is the average of five LES realizations and the standard error from these simulations is included in the table as well. Pei et al. [23] provides more detail regarding the variation among different LES realizations.

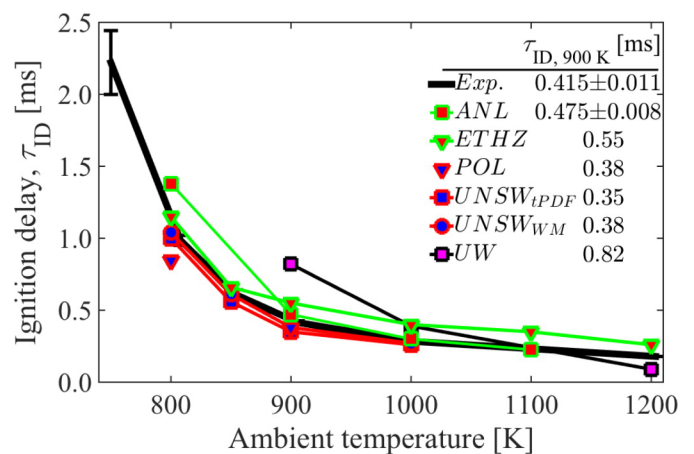


Figure 2. Experimental and simulated ignition delay times for Spray A and its parametric variants in ambient temperature.

The ANL, UNSW, and POL simulations best reproduce the experimental trends and quantities for ignition delay, with only a slight difference between the tPDF and well-mixed models of UNSW; however, it should be noted that the chemical mechanism implemented in the POL and UNSW simulations was specifically tuned by Yao et al. [25] to ECN spray combustion data. A consistent over-prediction of ignition delays is seen for the ETHZ results at 900 K and above using the Luo et al. 106-species chemistry. This systematic over-prediction was also found in perfect-stirred reactor (constant-volume) simulations compared to shock tube data. The UW