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Determination of soot onset and background particulate levels in a spark-ignition engine

Mitchell D. Hageman [a], Stephen S. Sakai [a,*], David A. Rothamer [b]

[a] *University of Wisconsin-Madison, Department of Mechanical Engineering, 1500 Engineering Dr., RM 140, Madison, WI 53706, USA*

[b] *University of Wisconsin-Madison, Department of Mechanical Engineering, 1500 Engineering Dr., RM 127, Madison, WI 53706, USA*

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Abstract

An experimental method utilizing premixing of the air and fuel well upstream of the intake port in conjunction with careful exhaust sample conditioning was developed to study the onset of sooting in spark-ignition engines. By fully premixing the air–fuel mixture upstream of the intake port, it was possible to create a fully premixed and pre-vaporized (PMPV) fuel–air charge and to eliminate liquid fuel in-cylinder. A baseline particle size distribution (PSD) was demonstrated using multiple fuels including gasoline, methanol, and E100. The baseline particulate level was shown to be insensitive to fuel composition and equivalence ratio below a critical value of the equivalence ratio or C/O ratio (Φ_c or C/O$_c$), indicating that the baseline PSD is not the result of fuel carbon, but instead is the result of other particulate sources, e.g., engine oil and engine wear particles. With the baseline particle levels established the PMPV technique was applied to determine the local enrichment needed to generate significant soot above the baseline particle size distribution. The critical equivalence ratio for onset of sooting under PMPV operation was determined to be between 1.345 and 1.349 (C/O$_c$ = 0.461–0.462). The results, interpreted based on the literature on laboratory premixed flames, indicate that soot first forms by the walls at the end of combustion or slightly after during the expansion stroke. The results give for the first time an indication of the level of local-enrichment that would be expected to cause a significant increase in soot production in spark-ignition direct-injection engines operating on gasoline at moderate engine load and speed conditions.

Keywords: SIDI; Soot; Gasoline; Threshold; Premixed

1. Introduction

The desire for higher fuel efficiency has driven the increased use of spark-ignition direct-injection (SIDI) engines in automotive applications over the past two decades. Estimates indicate that up to 60% of the light-duty automotive market in the United States will utilize SIDI engines by

* Corresponding author.

E-mail addresses: mhageman@wisc.edu (M.D. Hageman), ssakai@wisc.edu (S.S. Sakai), rothamer@wisc.edu (D.A. Rothamer).

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2016 [1]. One drawback of SIDI engines is their higher particulate matter (PM) number emissions compared to port-fuel injection (PFI) engines, both of which produce a large fraction of their PM emissions in the ultrafine (mobility diameter, D_m, < 100 nm) size range. Epidemiological studies indicate that nanoparticles can be more damaging to human health than micron-sized particles of the same composition [2]. Results from a range of studies have shown the negative impacts of ultrafine aerosols, including increased occurrences of cardiovascular events and reduced lung function [2–4], not to mention the possible carcinogenic nature of PM [5]. Concerns regarding the health effects of nanometer-sized particles emitted from vehicle exhaust have resulted in particulate number (PN) regulations in Europe for spark-ignition direct-injection engines [6,7] beginning in September of 2014, and stricter particulate mass regulations in the United States beginning in 2017 [1]. Current designs of both SIDI and PFI engines may have difficulty meeting these and future tighter limits [8].

The particulate formation and oxidation processes in SIDI engines have been less well studied than these processes in diesel engines. The majority of SIDI PM engine studies in the literature are applied studies that investigate the influence of engine operating parameters and engine design parameters on engine-out or vehicle tailpipe particle number and particle size distributions, see for example [8–11]. In these studies fundamental variables are not isolated and many thermodynamic and mixture properties are simultaneously changing. This makes it difficult to interpret results. From these studies, the root cause for the higher particle number emissions from SIDI engines has been identified as the higher likelihood for mixture inhomogeneity and fuel wetting of in-cylinder surfaces [11]. The level of mixture inhomogeneity and liquid fuel impingement is exacerbated at high engine loads, under vehicle accelerations and decelerations, and at cold start conditions [11]. Data from vehicle drive-cycle tests indicates that the vast majority of PN emissions are due to these events during the drive-cycle. The primary factor increasing PN emissions for these conditions is local and global fuel enrichment leading to conditions favorable for PM formation. One of the fundamental issues not addressed in the literature is the level of local enrichment needed to form a significant number of soot particles (here soot is taken to mean carbonaceous solid particles derived from the fuel and PM refers to all engine out particles) and how this depends on local thermodynamic conditions and fuel chemistry.

Premixed flame studies have provided a wealth of knowledge on soot formation and its dependence on fundamental variables: temperature (T), pressure (P), fuel structure, fuel-oxidizer ratio, and residence time [12]. A significant body of work has focused on the determination of critical equivalence ratios, Φ_c or Ψ_c (CO_2 or CO based [13], respectively), or critical carbon-to-oxygen ratios (C/O_c) where the onset (threshold) of sooting is observed (see for example [12–20] and references therein), typically by visual identification of soot luminosity. All of the critical sooting parameters are trying to measure similar things, here data will be discussed in terms of Φ_c and C/O_c. For oxygenated fuels the calculation of C/O includes the oxygen in the air and in the fuel. Previous work has demonstrated that increasing flame T results in an increase in Φ_c for $T > 1700$ K in atmospheric pressure flames. Böhm et al. [19] demonstrated that for $T < 1500$ K, C/O_c increases in ethylene and benzene flames. This results in an optimal T for soot formation where C/O_c and Φ_c are minimized. For C/O exceeding C/O_c the soot volume fraction produced in the flame is maximum at approximately the same optimal temperature. The increase in C/O_c with increasing T has often been stated as being due to a competition between precursor formation by fuel pyrolysis and oxidation of those precursors by the hydroxyl radical (OH) [13–15,21]. Markatou et al. [16] argued that this was not likely the case but instead that two factors: the concentration of acetylene, and the growth of polycyclic aromatic hydrocarbons (PAHs) control the appearance of soot. The formation of PAHs is limited with increasing T due to the increase in reverse reaction rates.

The critical C/O has also been shown to decrease with increasing pressure in premixed ethylene flames [17]. The influence of pressure on C/O_c is most prominent in the T range corresponding to the minimum C/O_c, i.e., 1500–1700 K. The influence at higher temperatures is much less pronounced. Above the threshold for sooting onset the soot volume fraction produced was seen to increase as P^2 up to a pressure of 10 bar and was seen to increase proportional to P for $P > 10$ bar [22].

The current work builds on previous measurements in premixed laminar flames to develop an experimental methodology utilizing premixed pre-vaporized (PMPV) fueling of a single-cylinder SIDI engine to isolate the chemical sooting tendency of the fuel and its dependence on thermodynamic conditions from physical property differences that influence mixture formation. This methodology is applied to determine a background particulate size distribution for the conditions of interest that is independent of fuel type. With knowledge of the background particle levels (due to oil, engine wear particles, etc.), a sooting threshold behavior is demonstrated, above which PN rapidly increases. It is shown that below this critical level of enrichment, particulate formation ceases to depend on Φ or fuel type. The concept