Available online at www.sciencedirect.com

ScienceDirect

Proceedings of the Combustion Institute 35 (2015) 3079–3086

Proceedings of the Combustion Institute

www.elsevier.com/locate/proci

In-cylinder soot precursor growth in a low-temperature combustion diesel engine: Laser-induced fluorescence of polycyclic aromatic hydrocarbons

C.A.J. Leermakers [a], M.P.B. Musculus [b,*]

[a] *Department of Mechanical Engineering, Eindhoven University of Technology, P.O. Box 513, GEM-N 1.21, 5600 MB Eindhoven, The Netherlands*

[b] *Combustion Research Facility, Sandia National Laboratories, P.O. Box 969, MS 9053, Livermore, CA 94551, USA*

Available online 19 July 2014

Abstract

The growth of poly-cyclic aromatic hydrocarbon (PAH) soot precursors are observed using a two-laser technique combining laser-induced fluorescence (LIF) of PAH with laser-induced incandescence (LII) of soot in a diesel engine under low-temperature combustion (LTC) conditions. The broad mixture distributions and slowed chemical kinetics of LTC "stretch out" soot-formation processes in both space and time, thereby facilitating their study. Imaging PAH–LIF from pulsed-laser excitation at three discrete wavelengths (266, 532, and 633 nm) reveals the temporal growth of PAH molecules, while soot-LII from a 1064-nm pulsed laser indicates inception to soot. The distribution of PAH–LIF also grows spatially within the combustion chamber before soot-LII is first detected. The PAH–LIF signals have broad spectra, much like LII, but typically with spectral profile that is inconsistent with laser-heated soot. Quantitative natural-emission spectroscopy also shows a broad emission spectrum, presumably from PAH chemiluminescence, temporally coinciding with of the PAH–LIF.

Keywords: Polycyclic aromatic hydrocarbons; Particulate matter; Laser-induced fluorescence; Laser-induced incandescence; Diesel low-temperature combustion

1. Introduction

Strategies to reduce pollutant emissions from diesel engines while maintaining fuel efficiency are of interest for meeting environmental regulations and market demands. Low-temperature combustion (LTC) is a class of in-cylinder strategies that uses dilute mixtures and enhanced pre-mixing to maintain or even improve fuel efficiency while reducing pollutant emissions, including soot [1]. In addition to its practical utility, the broad mixture distributions and slowed chemical-kinetics of LTC conditions "stretch out"

* Corresponding author. Fax: +1 (925) 294 1004.
E-mail address: mpmuscu@sandia.gov (M.P.B. Musculus).

http://dx.doi.org/10.1016/j.proci.2014.06.101
1540-7489/

soot-formation processes in both space and time, thereby facilitating their study.

Poly-cyclic aromatic hydrocarbons (PAH) are an important class of soot precursors [2]. Smaller PAH with one to five rings may be either fuel-borne [3] or combustion-generated. Larger PAH with more than five rings are almost entirely due to combustion-induced formation and/or growth. In addition to toxicity/carcinogenicity [4], PAH formation is undesirable because PAH contribute to particulate matter (soot) [5].

PAH growth during combustion can be observed to some degree through spectroscopic techniques. The absorption/emission spectra of PAH molecules shift to longer wavelengths with increasing number of rings [6,7]. Individual PAH spectra are broadband and overlap with each other, however, so specific PAH species generally cannot be isolated spectrally.

One technique to probe the absorption redshift due to PAH growth is laser-induced fluorescence (LIF). If soot is present, the laser-induced emission can also include significant laser-induced incandescence (LII) of soot, which is also spectrally broad. One way to discriminate soot-LII from PAH–LIF is to use a second laser pulse at a wavelength that yields only soot-LII. The Nd:YAG laser fundamental at 1064 nm should yield only soot-LII, because even very large PAH do not display single-photon absorption at near-infrared wavelengths [8,9]. Comparisons of the two signals, acquired virtually simultaneously, thus allow discrimination of PAH-only regions from regions with both PAH and soot.

This two-laser LIF-LII approach for diesel combustion follows previous work [10], using only 532-nm PAH–LIF. Here, the PAH–LIF is extended over a broader excitation wavelength range, adding 266 and 633 nm, to study the spatio-temporal evolution of both small and very large PAH. Excitation at 266 nm probes small PAH that do not absorb at 532 nm, while very large PAH could conceivably absorb at 633 nm and thus fluoresce upon 633-nm excitation. Such absorption, if present, would confound soot extinction techniques that use 633-nm (i.e. HeNe) laser light [11]. Both the soot-LII and the PAH–LIF signals are also spectrally resolved to further discriminate LIF from LII, and quantitative chemiluminescence spectra provide further insight into PAH growth.

2. Experiment set-up

2.1. Optical engine and operating conditions

The optical engine is a single-cylinder Bowditch-piston version of a Cummins N14 heavy-duty diesel (bore 139.7 mm, stroke 152.4 mm, 2.34 L). Table 1 shows operating conditions and some engine specifications (see [12,13] for more engine details).

Table 1
Engine specifications and operating conditions.

Compression ratio	11.22:1
Common-rail injector	Cummins XPI
Nozzle hole arrangement	8 × 140 μm × 152°
Fuel rail pressure	1000 bar
Command start of injection	352 CAD
Command injection duration	2.5 ms
TDC motored density	16.6 kg/m^3
TDC motored temperature	975 K
Intake pressure	177 kPa (abs)
Intake temperature	155 °C
(Simulated 16:1 intake pressure)	116 kPa (abs)
(Simulated EGR rates)	30–59%
Air excess ratio	2.39 at 21% O_2

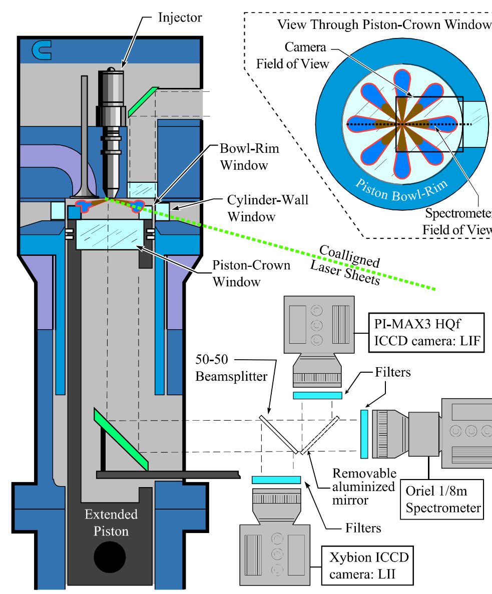

Fig. 1. Schematic of the single-cylinder engine, laser configuration, and optical detector system. The camera and spectrometer field-of-view are shown in the upper right corner.

In the experiment schematic of Fig. 1, a flat UV-grade fused-silica piston-crown window provides imaging access from below to the open, right-cylindrical bowl (diameter 97.8 mm). A 30-mm-wide curved window matching the piston bowl-wall contour and a flat rectangular cylinder-wall window provide laser access into the piston bowl, even near top-dead center.

A target load of 6 bar gross indicated mean-effective pressure (gIMEP) is investigated at 1200 rpm with a single fuel injection per cycle. The fuel is *n*-heptane, selected for its low fluorescence compared to pump diesel fuel to avoid interference with the PAH–LIF and soot-LII signals. It also has essentially zero fuel-borne PAH, meaning