



Full Length Article

Characterization of soot from diesel-CNG dual-fuel combustion in a CI engine



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HIGHLIGHTS

- Exhaust soot from diesel/CNG dual-fuel combustion analyzed using HRTEM, TGA, Raman and DRIFTS.
- Soot reactivity increased significantly with increasing CNG substitution rate.
- CNG soot showed larger variation in particle sizes and aggregate sizes.
- CNG soot appears immature due to insufficient time for development (reduced combustion duration).

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ABSTRACT

The physico-chemical characterization of exhaust soot is very important in the design and operation of suitable after-treatment systems. The nanostructure and reactivity of soot depends strongly on the initial fuel identity and synthesis conditions. This paper presents the chemical composition, oxidation reactivity and nanostructural characteristics of particulate matter (PM) produced by a diesel engine operating with diesel/compressed natural gas (CNG) dual-fuel combustion. Raw, undiluted soot samples from pure diesel, 40% CNG and 70% CNG (energy-based substitution rate) combustion were collected from the exhaust pipe. Engine operation conditions were held at 1200 RPM and 20 mg/cycle baseline load. For dual-fuel operation, split diesel injection (two injections) was used as the pilot. CNG was injected into the intake manifold. First, soot oxidation reactivity was characterized using thermogravimetric analysis (TGA). Transmission electron microscopy (TEM) was then used to determine the diameter of the spherules, and the morphology of agglomerates. Raman spectroscopy was employed to determine the graphitic nature of the soot. Finally, diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS) was used to identify surface functional groups of soot samples. It was found that soot reactivity increased with increasing CNG content. TEM images revealed a higher variation in particle diameter with increasing CNG substitution. High resolution TEM (HRTEM) images showed that CNG70 soot displayed features of immature soot particles. Raman spectroscopy results showed that soot reactivity does not depend on the initial graphitic structure. DRIFTS showed increased concentration of aliphatic and oxygen functional groups, which explained the higher reactivity. This enhanced reactivity could also be due to more active sites available in CNG soot, as well as due to CNG soot being immature. Under this test condition and engine configuration, it can be concluded that the use of CNG affects the morphology and nanostructure of PM, and hence the oxidation reactivity of the soot.

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1. Introduction

Dual-fuel combustion, burning compressed natural gas (CNG) and diesel, is an attractive alternative to diesel because CNG is

clean-burning, abundant, and has been shown to decrease particulate matter (PM) emissions. CNG is inexpensive, has low greenhouse gas emissions, and yields lower NO_x and CO₂ emissions [1,2]. CNG consumption has risen since 1995 due to tax incentives, increased CNG supply, and falling prices [3].

Performance and emissions of diesel/CNG dual-fuel combustion have been extensively studied. It has been shown that dual-fuel combustion can achieve efficiencies similar to that of diesel [4].

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Nomenclature

ATDC	after top dead center	HRTEM	high resolution transmission electron microscopy
BTDC	before top dead center	ICE	internal combustion engine
BTE	brake thermal efficiency	NO _x	nitrogen oxides
CAD	crank angle degree	PB	paper blackening
CI	compression ignition	PM	particulate matter
CNG	compressed natural gas	RPM	revolutions per minute
CO	carbon monoxide	TEM	transmission electron microscopy
DRIFTS	diffuse reflectance infrared fourier transform spectroscopy	TGA	thermo-gravimetric analysis
ECM	engine control module	THC	total hydro-carbons
ECU	engine control unit	UHC	unburned hydro-carbons
EGT	exhaust gas temperature	Φ	equivalence ratio
FSN	filter smoke number	Λ	lambda (1/Φ)

Unburned hydrocarbons (UHC) and carbon monoxide (CO) emissions are typically higher compared to pure diesel, however, significantly decreased soot emissions have been reported [5–8].

PM has been a serious concern for human health due to its direct and broad impact on the respiratory organs, as well as contributing to the global warming issue. Diesel exhaust gas is a major contributor to combustion-derived particulate-matter air pollution. As such, PM emission standards are continually evolving and becoming more stringent globally. The most common after-treatment method for soot reduction is to employ a diesel particulate filter (DPF) to trap soot particles. However, the design and effectiveness of these filters depend on soot properties. Thus, soot characterization has been the subject of several recent studies. Diesel soot generally takes the form of larger aggregates composed principally of smaller spherical particles called spherules and possessing characteristic structural properties. The nanostructure of soot depends strongly on the initial fuel identity and synthesis conditions, such as burning temperature, residence time, fuel properties, and fuel/oxygen ratio; the nanostructure in turn affects the oxidation reactivity of the soot [9–14]. Therefore, characterizing the physicochemical characteristics of soot is important and can provide crucial information to improve the design and operation of after-treatment systems. Common methods used to analyze soot structure and chemical properties are transmission electron microscopy (TEM) [15–21], thermogravimetric analysis (TGA) [16,19–21], Raman spectroscopy [19,21,22], and diffuse reflectance infrared fourier transform spectroscopy (DRIFTS) [9,11,21,22]. Although soot morphology and nanostructure have been widely studied for other alternative fuels such as crude vegetable oil [9] and biodiesels [10,11], there are no studies that have compared PM from diesel and diesel/CNG dual-fuel combustion.

In this study, we compare soot samples from pure diesel, 40% CNG, and 70% CNG (energy-based substitution rate) collected from the exhaust pipe. First, soot oxidation reactivity was characterized using TGA. TEM was then used to determine the diameter of the spherules and the morphology of agglomerates. Raman spectroscopy was employed to determine the graphitic nature of the soot, and finally, DRIFTS was used to identify surface functional groups of soot samples.

2. Experimental setup and methodology

2.1. Engine parameters

Experiments were carried out in an AVL 5402 single-cylinder diesel engine (displacement = 0.5 L; compression ratio = 17.1). The engine was modified to run diesel/CNG dual-fuel mode by adding a Solaris CNG injection system, which was used to inject CNG

into the intake manifold (port injection). Experiments were performed using No. 2 diesel and chemically pure methane was used to emulate CNG. The Solaris Diesel V4 control program was used to adjust the mass of CNG injected.

ETAS INCA v7 was used to control the electronic control module. In-cylinder pressure was measured using a Kistler pressure transducer, and NO_x, UHC, CO, and filter smoke number (FSN) were measured using Horiba emissions analyzers. More details can be found in [4].

The CNG substitution was defined as the percentage of heat energy from CNG available in the cylinder. For example, 70% CNG indicates that 70% of the energy input for the given engine condition was provided by CNG. Under a load equivalent to 20 mg/cycle diesel (medium load), a CNG flow rate of 15 L/min is equivalent to 70% CNG substitution. Test conditions are summarized in Table 1.

2.2. PM sampling

The PM sampling system was designed to allow simultaneous collection of raw PM on TEM grids, quartz fiber filter papers, and on stainless steel filters. The TEM grids (400 mesh Au) were inserted into the manifold through a Swagelok tee. The key advantage is that soot is directly collected in the aerosol phase onto TEM grids, bypassing filter collection and re-dispersal upon a TEM grid. Such processes cause agglomeration of soot aggregates and obscure aggregate recognition. For better high-resolution TEM (HRTEM) analysis, sampling was done on lacey carbon grids. PM was simultaneously collected on quartz (Pall Tissuquartz filters) and stainless steel filters using a vacuum pump system. PM collected on TEM grids was subjected to TEM, the quartz filters were used for Raman spectroscopy, and the PM was carefully scraped from the stainless steel filters for DRIFTS and TGA analysis.

2.3. Diagnostic techniques

2.3.1. TGA

A TA Instruments Q50 was used to perform TGA. The sample was loaded in an alumina crucible and placed inside a furnace, where the temperature was increased following a user-defined program (Table 2) and the weight loss was continuously recorded.

Table 1
Test conditions (1200 RPM, 20 mg/cycle load).

Fuel	Diesel injection strategy
#2 Diesel	5 mg @ 12 BTDC; 15 mg @ 4 BTDC
CNG40	3 mg @ 12 BTDC; 9 mg @ 4 BTDC
CNG70	2 mg @ 12 BTDC; 4 mg @ 4 BTDC

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