Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Physicochemical and toxicological characteristics of particulate matter emitted from a non-road diesel engine: Comparative evaluation of biodiesel-diesel and butanol-diesel blends

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ARTICLE INFO

Article history: Received 2 August 2013 Received in revised form 12 November 2013 Accepted 14 November 2013 Available online 22 November 2013

Keywords: Diesel particles Physicochemical PAHs Particle oxidation

ABSTRACT

Combustion experiments were conducted to evaluate the effects of using blends of ultralow sulfur diesel (ULSD) with biodiesel or *n*-butanol on physicochemical and toxicological characteristics of particulate emissions from a non-road diesel engine. The results indicated that compared to ULSD, both the blended fuels could effectively reduce the particulate mass and elemental carbon emissions, with butanol being more effective than biodiesel. The proportion of organic carbon and volatile organic compounds in particles increased for both blended fuels. However, biodiesel blended fuels showed lower total particle-phase polycyclic aromatic hydrocarbons (PAHs) emissions. The total number emissions of particles ≤ 560 nm in diameter decreased gradually for the butanol blended fuels, but increased significantly for the biodiesel blended fuels. Both the blended fuels indicated lower soot ignition temperature and activation energy. All the particle extracts showed a decline in cell viability with the increased dose. However, the change in cell viability among test fuels is not statistically significant different with the exception of DB-4 (biodiesel-diesel blend containing 4% oxygen) used at 75% engine load.

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1. Introduction

Diesel particulate matter (DPM) originating from both mobile and stationary sources has been linked to a myriad of adverse health outcomes, ranging from cancer to cardiopulmonary disease, and an array of environmental problems, including urban air pollution and global climate change [1–3]. The physical characteristics and chemical constituents of DPM are important parameters in determining their potential influence on the environment and human health. DPM consists mostly of carbonaceous materials, which are often classified as elemental carbon (EC) and organic carbon (OC) [2,4]. The EC suspended in the atmosphere can effectively absorb solar radiation, thus affecting the energy redistribution and global climate [3], while some semi-volatile hydrocarbons including polycyclic aromatic hydrocarbons (PAHs) in the OC fraction are suspected human carcinogens [2,4]. DPM consists of fine particles (PM_{2.5}) including a high number of nanoparticles and ultrafine particles with their aerodynamic diameter <50 nm and 100 nm, respectively. As these particles are released from diesel engines, it is much easier for them to diffuse in the ambient air, and adsorb more hazardous substances because of high surface to volume ratio.

Exposure to these particles can pose a cancer risk as well as other short and long term health problems [2]. The development of oxygenated fuels to partly or totally replace petroleum diesel and thus to reduce DPM emissions is getting a high priority as a result of increased public concern about energy security and environmental pollution. Various oxygenates have been considered based on their availability, price, toxicity, safety and compatibility with diesel fuel, among which biodiesel and alcohols are the most widely investigated ones [5,6].

Biodiesel obtained via alcohol transesterification from vegetable oils is a promising alternative energy source. This biodiesel is renewable, nontoxic and readily biodegradable. It is also free of sulfur and aromatic compounds, and possesses a high cetane number, high flash point and also good lubricity performance [5,6]. It has been widely reported that substantial reduction in soot and particulate mass emissions can be obtained through the addition of biodiesel from various vegetable oils to diesel fuel [6-11]. Recently, Lin et al. [8], Tsai et al. [9], Surawski et al. [10] and Sukjit et al. [11] have investigated the effect of biodiesel-diesel blends on chemical composition of DPM, including EC, OC, volatile organic compounds (VOC) and PAHs emissions in the particulates. Apart from biodiesel, alcohols, mainly methanol and ethanol, have been widely studied as alternate fuels for reducing the smoke and the potentially carcinogenic carbonaceous soot particles emissions [7]. However, some practical difficulties prevent their use as fuels for diesel engines,







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^{0304-3894/\$ -} see front matter © 2013 Elsevier B.V. All rights reserved. http://dx.doi.org/10.1016/j.jhazmat.2013.11.033

for example, reduced lower heating value (LHV) compared to diesel fuel, miscibility and stability problems when blended with diesel fuel, low cetane number and poor lubricating properties [12,13]. Butanol has recently drawn particular attention as a renewable biofuel for diesel engines due to its higher heating value and cetane number, more miscibility with diesel, and less hydrophilic compared to methanol and ethanol [11–13]. These studies revealed the beneficial effects of using various blends of butanol with diesel fuel on carbon monoxide (CO), smoke and DPM emissions at various loads [12,13]. However, detailed investigations concerning the use of butanol-diesel blends in diesel engines and its effects on DPM emissions are still limited [11]. In addition, it is still not clear which chemical constituents of DPM are mainly responsible for its health effects, and how biodiesel-diesel or butanol-diesel blends affect particle toxicity.

The use of oxygenated fuels as a blending agent may not only reduce DPM emissions, but also may lead to a change in both the physical and chemical properties of the particulates and thus their toxicological characteristics. However, there is certainly a lack of in-depth investigations on the influence of biodiesel-diesel blends and butanol-diesel blends on these changes. The current study represents the first attempt of its kind to fully evaluate and compare the potential impact of biodiesel-diesel blends and butanol-diesel blends on physical, chemical and toxicological properties of the particulates emitted by a diesel engine, with the objective being to provide information for assessing if these fuel blends reduce health and environmental impacts or otherwise. It should be noted that previous studies on the influence of oxygenated fuels on DPM emissions were mainly conducted on on-road diesel engines. Although non-road diesel engines contribute a small fraction of the total number of diesel engines in operation, they account for a disproportionate fraction of the PM and NO_x emissions because they typically have minimal emissions control [4,8]. Moreover, the different operating conditions of non-road and on-road diesel engines result in different emission characteristics. This implies that the emission studies of on-road diesel engines cannot be used for understanding the emissions of non-road diesel engines [14].

2. Experimental

2.1. Test engine and fuels

The schematic of the experimental system is shown in Supplementary Fig. S1. Experiments were carried out on a single cylinder, naturally aspirated, four-stroke, direct-injection diesel engine (L70AE, Yanmar Corporation) connected to a 4.5 kW generator. The diesel engine has a capacity of 296 cm³ with bore and stroke of 78 mm \times 62 mm, a fixed speed of 3000 rpm (revolutions per min). The fuels used include ULSD with less than 50 ppm (parts per million) by weight of sulfur, palm oil methyl ester (PME) as biodiesel, and anhydrous n-butanol of 99.8% purity (Sigma-Aldrich). The PME used in this study was obtained from palm oil-based biodiesel plant in Malaysia operated by Vance Bioenergy, and the fuel properties are provided by the biodiesel supplier. Major properties of the tested fuels are shown in Supplementary Table S1. Biodiesel-diesel blends and butanol-diesel blends having the same oxygen concentrations of 1%, 2%, and 4% by weight were used for this study, which were designated as DB-1, DB-2, DB-4, and DBu-1, DBu-2, DBu-4, respectively.

2.2. Particulate sampling and testing

A two-stage Dekati mini-diluter (DI-2000, Dekati Ltd) was used for diluting the exhaust gas for sampling. The diluter provides primary dilution in the range of 8:1 to 6:1, depending on the engine operating conditions, while the secondary dilution system provides a further dilution of 8:1. The actual dilution ratio was evaluated based on measured CO_2 concentrations in the raw exhaust, in the background air and in the diluted exhaust. The CO_2 concentration was measured with a non-dispersive infrared analyzer (MRU Vario Plus, Germany,±0.5% accuracy).

The first stage diluter was only used to cool the sampling gas temperature below 52 °C for particulate sampling. PM with aerodynamic diameter $\leq 2.5 \,\mu$ m (PM_{2.5}) was collected on pre-combusted (650 °C for 12 h) 47 mm quartz fiber filters (Whatman, USA) by using two Mini-Vol low volume particulate samplers (Air metrics Ltd; $5 \,\text{Lmin}^{-1}$ flow rate), respectively. Before and after sampling, the filters were allowed to equilibrate in a humidity-controlled chamber at constant temperature and humidity (22 ± 3 °C, $35 \pm 8\%$ RH), and weighed using a microbalance (Sartorius MC5, accuracy of $\pm 1 \,\mu$ g) for quantifying total PM_{2.5} mass emissions.

After taking the weight, the filters were closed in glass petri dishes and stored under refrigeration at -20 °C for subsequent chemical analysis. A thermal/optical carbon aerosol analyzer (Sunset Labs, Forest Grove, Oregon, USA) was used to quantify EC and OC emissions according to the NIOSH 5040 reference method [15].

A thermogravimetric analyzer (TA instruments, SDT Q600) was used to investigate the volatile organic fraction (VOF) in particles and the oxidation property of soot. The TGA heating program is the same as used in [16]. The mass loss in the argon environment was taken as the mass of the volatile organic fraction (VOF). The remaining part (soot) was continually heated at an air environment to investigate soot oxidation reactivity. The ignition temperature is determined as the temperature at which the rate of mass loss at the air environment reaches the maximum. While the activation energy for the soot was estimated using a modified form of the Arrhenius expression, as suggested in Stratakis and Stamatelos [17]. A similar approach had been used in earlier studies reported in the literature [11,16,18].

The number concentration and size distributions of volatile and non-volatile particles in the secondary dilution stage were measured by a fast mobility particle sizer (FMPS, Model 3091, TSI Incorporated, USA) for particles in the size range of 5.6 to 560 nm. In this setup, two diluters were used in series, with the first stage being heated by a surface heater to 190°C in order to minimize thermophoretic deposition. During the non-volatile particle number emission experiments, a thermodenuder (TD, Dekati Ltd) was placed in-line between the secondary stage diluter and the FMPS. In the TD, the volatile and semi-volatile compounds of the particles are vaporized by heating the sample aerosol, and then are gradually cooled and adsorbed into active charcoal so that they do not re-condense onto the remaining solid particles. The temperature in the heating section of the TD was maintained at 265 °C. The TD diffusion losses were estimated using the method of Lu et al. [18], and the diffusion loss-corrected particle size distributions are presented in this article.

2.3. Particle-phase PAHs

Particle-phase PAHs were extracted from the quartz filter samples in 50/50 hexane/acetone (HPLC-grade, Fisher Scientific) mixture using a closed vessel microwave-assisted extraction system (MLS-1200 mega, Milestone, Italy) in accordance with the US EPA method 3546 [19]. The particle extracts were first filtered, concentrated to 5 mL using a rotary evaporator (Buchi, Switzerland), and finally dried under a gentle stream of nitrogen. Each of the eluents was divided into two equal parts, with one part for subsequent PAHs analysis, and the other part for toxicological analysis. More details of this method including the chemical analysis of 16 US priority PAHs by gas chromatography/mass spectrometry (GC/MS) have been discussed in our previous publication [20]. The total-PAH Download English Version:

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