Role of Cerium Oxide Nanoparticles as Diesel Additives in Combustion Efficiency Improvements and Emission Reduction

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Abstract: An experimental investigation is conducted to evaluate the performance, combustion, and emission characteristics of a diesel engine using cerium oxide (CeO_2) nanoparticles as diesel fuel additives. Using cetyl trimethyl ammonium bromide (CTAB) as the cationic surfactant and with ultrasonic vibration, CeO_2 nanoparticles at dosing levels of 50 and 100 mg/L can be stably and uniformly dispersed in diesel. Physically, the mixing of fuel blends with air is enhanced by microexplosion events because of the large surface-volume ratio and intensified thermal transmissibility of nanoparticles. Moreover, chemically, CeO_2 in diesel fuel plays a positive role in the dehydrogenation reaction at a high temperature because of its excellent redox ability. As a result, the addition of nano-CeO_2 to neat diesel (50 and 100 mg/L) leads to a gradual increase in the cylinder pressure compared with the reference neat diesel. Meanwhile, the ignition of fuel blends with a high dosing level of nano-CeO_2 occurs earlier by approximately 1.2° crank angle (CA) and 1.8°CA, respectively. At full load, the effective thermal efficiencies for nano-CeO_2 fuels are augmented by approximately 1.7 and 2.3%. Because of the distinctive merits of nano-CeO_2 in promoting fuel atomization and its favorable intrinsic catalytic effect, the level of harmful pollutants (such as HC, CO, NO_x, and soot) in exhaust gases is appreciably reduced to varying degrees. **DOI: 10.1061/(ASCE)EY.1943-7897.0000329.**

Author keywords: Cerium oxide; Nanoparticles; Diesel engine; Combustion; Emissions.

Introduction

Fossil fuels have played a critical role in the application of power engines. In particular, diesel engines, which have superior reliability, durability, sturdiness, and fuel economy compared with gasoline engines, have a great advantage in various fields such as transportation, electricity, marine, agricultural, and industrial applications. However, diesel engines produce harmful emissions, especially nitrogen oxides (NO_x), hydrocarbons (HC), and particulate matter (PM), which give birth to a variety of global environmental issues such as acid rain, greenhouse effects, and hazy weather. Therefore, stringent emission legislations are conducted to minimize pollutants.

To date, great efforts have been made to reduce emissions from diesel engines by three main approaches: (1) internal engine modification, (2) exhaust after-treatment, and (3) fuel adulteration. In this research, the fuel adulteration approach is adopted for convenience because it does not require any engine structure modification. A number of experimental investigations with a wide variety of metal additives have reported improvements in fuel properties, engine performances, and reduced emissions. The effects of calcium, barium, iron, and nickel naphthenates have been comprehensively studied, with the conclusion that calcium and barium can most efficiently reduce soot, both by suppressing soot formation and by enhancing soot oxidation (Miyamoto et al. 1987). Gürü et al. (2002) made a conclusion that the cetane number of diesel blend was enhanced, and the freezing point decreased by adding organic-based manganese drops to diesel.

Recently, solid nanoparticle additives have been considered to be advantageous catalysts that improve fuel properties. Efficient combustion and emission reduction for engines can be achieved through adding suitable levels of nano-additives to hydrocarbon fuels. Based on an experiment in a diesel engine using nano-aluminum diesel blends, Kao et al. (2008) reported that the brake specific fuel consumption and harmful emissions such as soot and NO_x were significantly reduced, and they demonstrated that metal oxide additives in a water/diesel emulsion acted as catalysts to activate the molecular bonds and promote clean burning and emissions, thanks to nano-sized aluminum. Yetter et al. (2009) found a shortened ignition delay and lower ignition temperature during combustion by adding nano-catalysts into hydrocarbon fuels. Basha and Anand (2011a) conducted experiments to investigate the performances of diesel blended with carbon nanotubes and observed a significant improvement in harmful pollutants and brake thermal efficiency because of the enhanced combustion characteristics of nano-sized solid particles, and they also observed the shortened evaporation time and ignition delay.

Cerium oxide, with the desirable property of easy oxidation and reduction, has served as a popular catalyst. The amount of oxygen reversibly provided in and removed from the gas phase is called the oxygen storage capacity of cerium [S. Roger, "Cerium oxide nanoparticles as fuel additives," European Patent No. EP1587898 (2004)]. Cerium oxide nanoparticles possess a high specific surface area and can lead to high reactivity. Therefore, cerium oxide is widely used in a variety of fields such as oxygen sensitive materials, fuel batteries, and automobile exhaust purification.

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Note. This manuscript was submitted on February 2, 2015; approved on September 16, 2015; published online on December 16, 2015. Discussion period open until May 16, 2016; separate discussions must be submitted for individual papers. This paper is part of the ***Journal of Energy Engineering***, © ASCE, ISSN 0733-9402.

Jung et al. (2005) and Ribeiro et al. (2007) have critically reviewed the influence of cerium oxide additive on neat diesel PM emissions and the kinetic reaction of oxidation. They found that the addition of nano-CeO_2 to diesel caused a significant reduction in the number-counted size distributions and ignition temperature and an enhancement in the oxidation rate. An experiment was conducted by Sajith et al. (2010) in a single-cylinder diesel engine by adding ceria nanoparticles to jatropha biodiesel. A clear reduction of NO_x and HC and improvement in the brake thermal efficiency were observed. They concluded that nano-ceria in the base fuel acted as an oxygen buffer and offered high catalytic activity because of its higher surface to volume ratio. Rhodia company has explored a kind of ceria-based liquid fuel-borne catalyst which is called Eolys (Rhodia Group, France). Studies show that Eolys can facilitate the regeneration of diesel particulate filter (DPF) and eliminates most of soot (Lemaire 1999). Jiang et al. (2009) reported that Ce^{4+} and Ce^{3+} ions can be interconverted under special conditions because cerium oxide has a low redox potential between Ce^{4+} and Ce^{3+} ions (1.7 V) (Selvan et al. 2014) and can absorb oxygen in a high-oxygen atmosphere and donate oxygen in a low-oxygen atmosphere (Lawrence et al. 2011).

Studies show that nano-CeO_2 can play a particular role in catalytic combustion and gas purification through appropriate addition into hydrocarbon fuels. However, current research on the applications of nanoparticle additives in combustion engines is still in the preliminary stage. Given the potential properties of nano-CeO_2, this work aimed to investigate its effects on power performance, combustion characteristics, and emissions in a single-cylinder direct injection diesel engine using nano-CeO_2 fuel blends. The brake specific fuel consumption, heat release rate, and exhaust emissions are measured or calculated to perform an intuitive comparison between the modified fuel blends containing nanoscale CeO_2 particles and the reference neat diesel. Thus, the role of nano-scale particles with inherent catalytic activities in promoting fuel combustion and purifying emissions is synthetically expounded from both physical and chemical perspectives.

Preparation of Fuel Blend

The CeO_2 nanoparticles used in this work were produced through a chemical precipitation method. They were bought from Beijing Dekedaojin Technology in China. The nanoparticle material is 15–30 nm in size. Its surface and structure morphology were determined by scanning electron microscope (SEM) Model JSM-7001F (Japan Electron Optics Laboratory, Tokyo, Japan). The specific surface area of the nanometer CeO_2 sample is 30–50 m^2/g, and its morphology is shown in Fig. 1, with a uniformly spherical appearance and a dark yellow color.

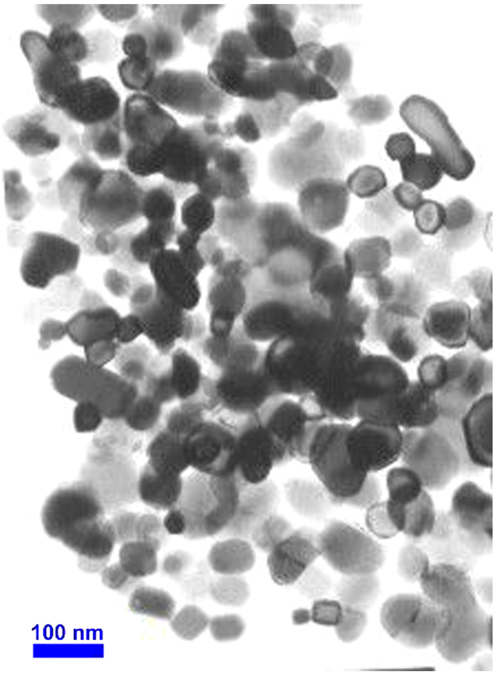

Fig. 1. SEM of CeO_2 (image by Qimin Wu)

In general, nanoparticles have higher surface energy because of their higher surface area, so they tend to agglomerate and gather into large particles which are prone to deposition (Luo et al. 1995). Nanometer CeO_2 particles are difficult to disperse uniformly in diesel. However, combined chemical and physical methods can help to produce the stable and uniform suspensions. Surface modification is an effective and widely used chemical measure. Cetyl trimethyl ammonium bromide (CTAB), acting as a cationic surfactant, can form envelopes on the surfaces of particles to provide them with negative charges and hence suppress the sedimentation of nanoparticles. Moreover, the physical assistance of ultrasonic vibration is also necessary to form well-distributed and stable suspensions (Basha and Anand 2011b). In this study, nano-CeO_2 particles and the same weight of CTAB were weighed by a digital balance machine to a predefined dosing level of 50 mg/L and poured into the base diesel fuel, and then the fuel blends were vibrated by an ultrasound unit at a frequency of 40 kHz under 150 W working power for 1 h. The same procedure was performed to prepare the fuel blend with a dosing level of 100 mg/L. Then, the fuel blends for testing were obtained in light yellow. The labels for the two nanoparticle fuel blends, together with the reference neat diesel, and their specific compositions are briefly described in Table 1. It was observed that D50Ce and D100Ce samples can remain stable for more than 2 days without discernable sedimentation.

Experimental Setup and Procedure

The experiments were conducted using a single-cylinder, four stroke, naturally aspirated, air-cooled, direct injection, compression ignition engine. Its primary technical parameters are provided in Table 2. The test engine was coupled to a direct-current dynamometer for loading. The instrumentation system consists of a Horiba MEXA 7200D exhaust analysis system (HORIBA Advanced Techno, Kyoto, Japan), an AVL 415s smoke meter (AVL List Gmbh, Graz, Austria), and a Dewetron combustion analyzer (DEWETRON Gmbh, Austria). The Horiba MEXA 7200D exhaust gas analysis system consists of a group of analyzers for measuring nitrogen oxides (NO_x), carbon monoxide (CO), and total unburned hydrocarbons (HC). NO_x emissions were measured using a chemiluminescent analyzer CLA-755A, HC emissions were measured using a flame ionization detector FIA-725A, and CO emissions were determined using a nondispersive infrared analyzer AIA-721. The smoke level in the exhaust gas was measured using an AVL 415s filter type smoke meter. The fuel consumption was

Table 1. Compositions of Test Fuels

Label	Composition
Diesel	neat diesel
D50Ce	neat diesel + 50 mg/L CTAB + 50 mg/L CeO_2
D100Ce	neat diesel + 100 mg/L CTAB + 100 mg/L CeO_2

Table 2. Technical Parameters of Test Engine

Model	Naturally aspirated, four stroke, single-cylinder, air cooling, direct injection
Bore (mm)	86
Stroke (mm)	72
Compression ratio	18.5
Rated power/speed ($\mathrm{kW/r \cdot min^{-1}}$)	4.8/3,000
Combustion chamber	ω
Nozzle holes	5
Spray hole diameter (mm)	0.18
Spray cone angle	147°
Injection timing	16°CA (BTDC)
Injection pressure	20 MPa

Note: BTDC = before top dead center.

determined using a MCS-960 gravimetric fuel meter (Zhongcheng, Hongzhou, China), which consists of a measuring vessel filled with fuel suspended on a balance system. Fuel consumption values were then obtained by calculating the vessel's weight loss over time. A schematic representation of the experimental setup is shown in Fig. 2.

The combustion attributes of the diesel engine were obtained using the pressure transducer mounted on the cylinder head, and the crank angle encoder was used to record the engine crank angle. The pressure signals collected from the piezoelectric pressure sensor were given as an input to the data acquisition system through a signal conditioning unit. The cylinder pressures were measured with an accuracy within ±1% of the full scale. For each operating mode, 200 consecutive cycles of cylinder pressure data with a sampling rate of 0.2° crank angle (CA) were collected and averaged using a computer program. The heat release rate can be calculated based on the first law of thermodynamics via the cylinder pressure data. The ignition delay and other characteristic parameters can be obtained by analyzing the heat release regularity.

The engine was started with neat diesel and then warmed up. The warming up period ended when the lubricant temperature was stabilized. Then, the fuel consumption, cylinder pressure, exhaust emissions, and smoke opacity were measured. A similar procedure was repeated for the other two nano-fuel blends. For nano-diesel fuel, the tests were conducted by starting and warming up the engine with neat diesel and then switching to nano-fuels. At the end of the test, the engine was run with neat diesel to flush out the fuel blends containing nano-$\mathrm{CeO_2}$ particles from the fuel lines and the injection system. To ensure the accuracy of the measured values, the gas analyzers were calibrated before each measurement using reference gases. The experiments were performed at a constant engine speed of 3,000 revolutions per minute (rpm) by varying the loads. Tests on each fuel were repeated thrice, and the average of the measurements was noted at a steady state and under identical conditions. The repeatability of all the results was within 2%.

Results and Discussion

The power performance, combustion, and emission characteristics of the diesel engine using neat diesel and nano-$\mathrm{CeO_2}$-blended diesel fuels (D50Ce and D100Ce) were investigated. Based on the combustion data, the cylinder pressure and heat release rate are plotted against the crank angle. The performance attributes such as brake thermal efficiency, brake specific fuel consumption, and the emission characteristics of $\mathrm{NO_x}$, HC, CO, and smoke are plotted against brake mean effective pressure (BMEP).

Analysis of Combustion Process

The variation in the cylinder pressure and the heat release rate with respect to the crank angle for D50Ce and D100Ce fuel blends and the reference diesel fuel at the rated operating mode ($p_{me} = 0.46$ MPa, $n = 3{,}000$ rpm) are shown in Fig. 3. The addition of nano-$\mathrm{CeO_2}$ to the diesel (50 and 100 mg/L) leads to a gradual

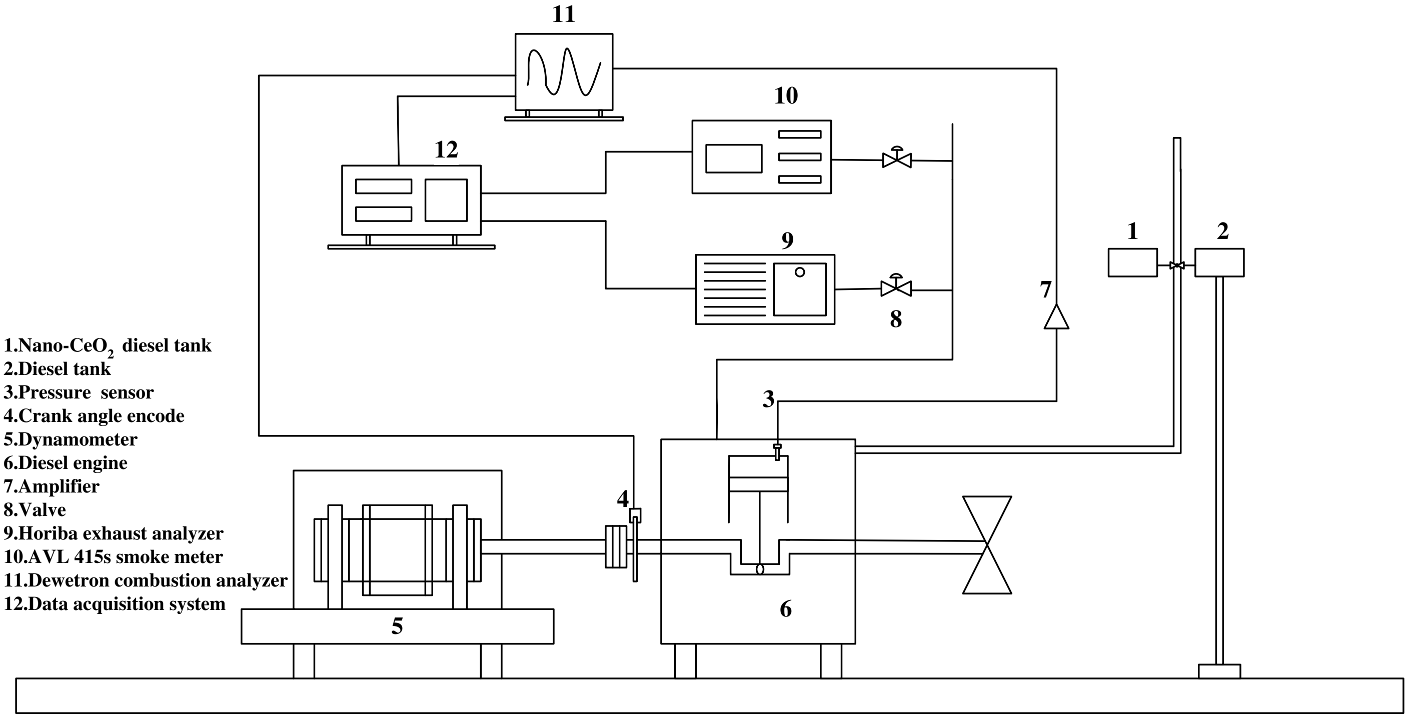

Fig. 2. Schematic representation of experimental setup

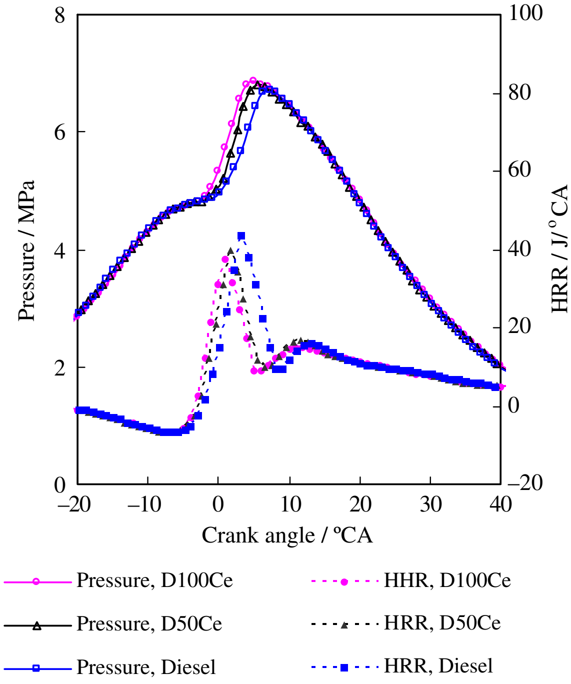

Fig. 3. Cylinder pressure and heat release rate with crank angle

increase in cylinder pressure because of enhancement of the mixing process of fuel with air and its intrinsic catalytic combustion performance. The small size and large surface-volume ratio of nano-CeO_2 particles intensify the thermal conductivity of diesel. Moreover, diesel has a much lower boiling temperature than that of nanoparticles. At a high temperature in the cylinder, when fuel drops become super-heated and swollen, the nanoparticles form layers. Subsequently, a drastic vaporization event, which is called a microexplosion (Ganesh and Gowrishankar 2011; Basha and Anand 2014) (Fig. 4), takes place. This microexplosion promotes the secondary atomization of fuel droplets toward ultrafine granularities and thus shortens the physical preparation period before ignition. In addition to the favorable physical attributes of nanoparticles, CeO_2 in diesel fuel plays a positive role in dehydrogenation at high temperatures because of its excellent redox ability (Lawrence et al. 2011). Classically, diesel hydrocarbons are in the form of H─C covalent bonds. Cerium oxide, as an efficient catalyst, can release oxygen and promote dehydrogenation on the particle surface to form active radicals such as OH^-, which can facilitate the spontaneous combustion of diesel fuel. As a result, the cylinder pressure for D50Ce and D100Ce are augmented compared with the values for neat diesel.

Because of the distinctive merits of nano-CeO_2 in promoting fuel atomization and favorable catalytic function in the dehydrogenation reaction, both physical and chemical preparations are effectively boosted before ignition. The combustion characteristic parameters are summarized in Table 3. A diesel fuel blend with a high dosing level of nano-CeO_2 can cause earlier ignition, which also can be sufficiently supported by the excellent flammability of nano-fuels on a hot plate (Tyagi et al. 2008; Basha and Anand 2011c). The ignition delays are, respectively, shortened by approximately 1.2°CA and 1.8°CA for D50Ce and D100Ce. As usual, for the same working condition, a longer ignition delay for the reference neat diesel indicates a higher heat release peak, as illustrated in Fig. 3.

Economic Performance

The variations in effective thermal efficiencies and brake specific fuel consumption for neat diesel, D50Ce, and D100Ce fuels are shown in Fig. 5. An increase in thermal efficiency and a slight decline in specific fuel consumption for nano-CeO_2 fuels compared with neat diesel are observed. As stated previously, with the addition of nano-CeO_2, the fuel evaporation time and the physical delay is favorably lessened because of the microexplosion effects. In addition, the intensified thermal transmissibility of CeO_2 nanoparticles accelerates the chain reactions at the following combustion stage. CeO_2 acts as a catalyst and produces plentiful oxygen for combustion, which causes the fuel to burn rapidly and completely. Therefore, the effective thermal efficiencies for D50Ce and D100Ce fuels are improved by approximately 1.7 and 2.3% at full load compared with neat diesel. Correspondingly, the brake specific consumption is reduced with increased dosing levels of nanoparticles.

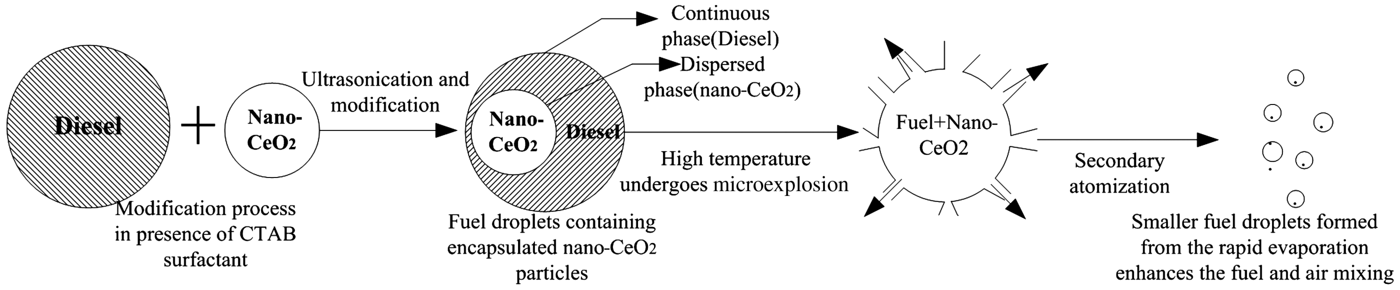

Fig. 4. Microexplosion and secondary atomization for nano-CeO_2 fuel blend

Table 3. Combustion Characteristic Parameters

Dosing level of nanoparticles (mg/L)	Ignition timing (°CA)	Peak pressure (MPa)	Location of peak pressure (°CA)	Heat release peak (J/°CA)	Location of heat release peak (°CA)
0	−5.8	6.702	7.2	42.08	3.2
50	−7.0	6.816	5.8	39.71	1.6
100	−7.6	6.881	5.0	37.42	0.8

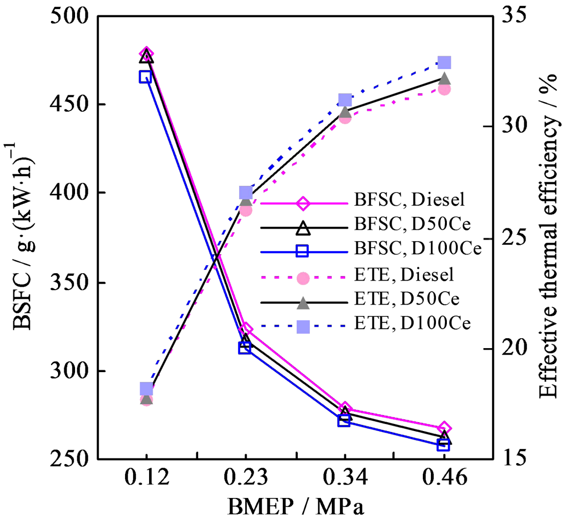

Fig. 5. Variation in brake specific fuel consumption and effective thermal efficiency

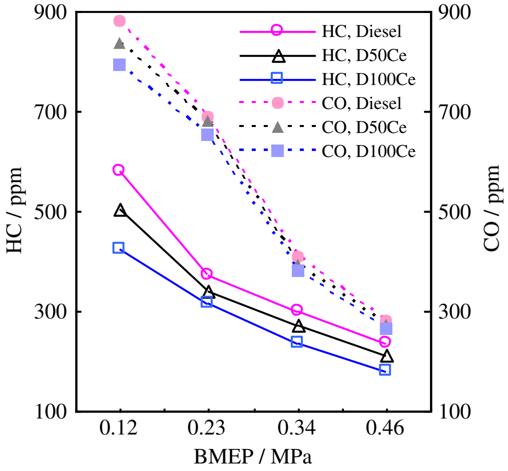

Fig. 6. Variations of HC and CO emissions

Emission Features

HC and CO emissions at the rated operating mode are compared in Fig. 6. Clearly, for D50Ce and D100Ce, both HC and CO emissions are decreased to some degree compared with neat diesel. Cerium oxide can be beneficially transformed from the stoichiometric CeO_2 (+4) valance state to the Ce_2O_3 (+3) state via a relatively low-energy reaction. Cerium oxide supplies the oxygen for the oxidation of hydrocarbons and is converted to cerous oxide (Ce_2O_3) as follows (Sajith et al. 2010):

$$(2x+y)CeO_2 + C_xH_y \rightarrow \frac{x+y}{2}Ce_2O_3 + \frac{x}{2}CO_2 + \frac{y}{2}H_2O \quad (1)$$

In addition, cerium ions can actively lower the activation energy for breaking H─C bonds and accelerate the dehydrogenation reaction at high temperatures. Owing to the catalytic role of nano-CeO_2, fuel-air mixtures tend to burn earlier. Such a reduction in combustion delay has been related to the decrease in HC emissions (Monyem et al. 2001). The average decrease in HC emissions compared with the reference fuel were 15.1% and 18.4% for D50Ce and D100Ce, respectively.

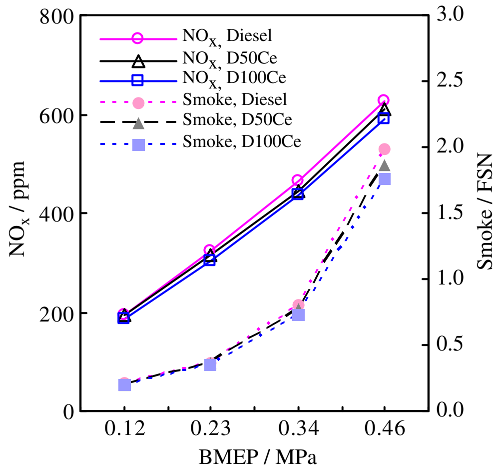

Fig. 7. Variations of NO_x and smoke emissions

As shown in Fig. 6, higher CO emissions are observed for neat diesel than for nano-CeO_2 fuels. For neat diesel, when there is not enough oxygen to convert all carbon into CO_2, some fuels are not burned, and some carbon ends up as carbon monoxide. Carbon monoxide is an intermediate product formed during combustion. Poor mixing, local rich regions, and incomplete combustion can also result in CO emissions (Selvan et al. 2014). Because of its advantageous oxygen storage and release capacity, nano-CeO_2 in diesel fuel acts as an oxygen buffer and donates more oxygen for carbon to convert into CO_2. The oxygen vacancy defects (OVDs) of ceria can absorb CO molecules to directly form CO_2 or form the intermediate bidentate carbonate, which can also oxidize CO to CO_2 (Jiang et al. 2009). The corresponding reaction principle is presented in Eq. (2). Moreover, the secondary atomization because of the microexplosion of nano-CeO_2 fuel blends lowers the probability of fuel-rich zone formation, which is usually related to CO emissions (Lapuerta et al. 2008). In summary, CO emissions for D50Ce and D100Ce are reduced by 3.4 and 6.5%, respectively, compared with neat diesel

$$2CeO_2 + CO \rightarrow Ce_2O_3 + CO_2 \quad (2)$$

The NO_x and smoke emission characteristics of D50Ce and D100Ce fuel blends and the base diesel fuel are illustrated in Fig. 7. Because of its high thermal stability, Ce_2O_3 formed from the reaction of hydrocarbon and soot remains active after enhancing the initial combustion and is reoxidized to CeO_2 through the reduction of nitrogen oxide (Sajith et al. 2010). NO and NO_2 can be reduced to N_2 through chemical Eqs. (3) and (4) (Bozek et al. 2011a, b). Thus, NO_x emissions can be inhibited to some extent, and meanwhile, CeO_2 can be regenerated. Overall, the NO_x emissions of nano-CeO_2 fuel blends are clearly lower than those of neat diesel, with average reductions of 2.4 and 5.4%, respectively

$$2Ce_2O_3 + NO \rightarrow 4CeO_2 + N_2 \quad (3)$$

$$4Ce_2O_3 + 2NO_2 \rightarrow 8CeO_2 + N_2 \quad (4)$$

Soot is usually formed because of the thermal decomposition of fuel at a high temperature under oxygen deficit conditions. The secondary atomization role for nano-CeO_2 fuel blends promotes sufficient mixing of fuel with air and thus can effectively abate fuel-rich zones, reducing the generation of C_2H_4, the precursor of PM, generated during combustion. Moreover, owing to the excellent oxygen storage capacity of cerium oxide, it can allow oxygen to facilitate the further oxidation of soot. Chemical Eq. (5) for soot burning is as follows. The soot formed in the cylinder can be partly oxidized into CO_2 through the catalytic reaction of CeO_2

$$4CeO_2 + C\,(\text{soot}) \rightarrow 2Ce_2O_3 + CO_2 \qquad (5)$$

In summary, nano-CeO_2 can act as a smoke suppression additive. The smoke emissions are decreased by approximately 4.0 and 7.8% for D50Ce and D100Ce, respectively. Although the addition of nano-CeO_2 to diesel will result in the presence of cerium oxides in PM, a trace amount of CeO_2 has little effect on PM emissions. The catalytic activity of CeO_2 on soot oxidation can introduce new ideas regarding the internal online regeneration of DPF, as suggested by studies of Jung et al. (2005) and Valentine et al. (2000).

Conclusions

The combustion and emission characteristics of a diesel engine using neat diesel and nano-CeO_2-blended fuels were investigated to understand the effects of ceria nanoparticles as a fuel additive in diesel. Because of both the favorable fuel atomization and the beneficial catalytic activities of nano-CeO_2, adding it to neat diesel (50 and 100 mg/L) leads to a gradual increase in cylinder pressure compared with the base diesel fuel. The ignition delays of D50Ce and D100Ce are shortened by approximately 1.2°CA and 1.8°CA, respectively. At full load, the effective thermal efficiencies for D50Ce and D100Ce fuels are augmented by approximately 1.7 and 2.3%. Because of the desirable oxygen storage capacity of CeO_2 and its active role in promoting fuel atomization, the levels of harmful pollutants such as HC, CO, NO_x, and soot are decreased to varying degrees.

Overall, the nano-CeO_2 particles can act as a highly efficient fuel catalyst. Adding these nanoparticles into diesel could achieve efficient combustion and energy reduction in diesel engines.

Acknowledgments

The authors wish to express their appreciation for the funds from the National Natural Science Foundation of China (No. 51376083 and No. 51376095) and the Priority Academic Program Development of Jiangsu Higher Education Institutions (PAPD), which supported this study.

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