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Impacts of continuously regenerating trap and particle oxidation catalyst on the NO_2 and particulate matter emissions emitted from diesel engine

Zhihua Liu[1], Yunshan Ge[1,*], Jianwei Tan[1], Chao He[1], Asad Naeem Shah[1,2],
Yan Ding[3], Linxiao Yu[1], Wei Zhao[1]

1. National Lab of Auto Performance and Emission Test, School of Mechanical and Vehicular Engineering, Beijing Institute of Technology, Beijing 100081, China. E-mail: liuzhihua161@163.com
2. Department of Mechanical Engineering, University of Engineering and Technology, Lahore 54000, Pakistan
3. Chinese Research Academy of Environmental Sciences, Beijing 100012, China

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Abstract

Two continuously regenerating diesel particulate filter (CRDPF) with different configurations and one particles oxidation catalyst (POC) were employed to perform experiments in a controlled laboratory setting to evaluate their effects on NO_2, smoke and particle number emissions. The results showed that the application of the after-treatments increased the emission ratios of NO_2/NOx significantly. The results of smoke emissions and particle number (PN) emissions indicated that both CRDPFs had sufficient capacity to remove more than 90% of total particulate matter (PM) and more than 97% of solid particles. However, the POC was able to remove the organic components of total PM, and only partially to remove the carbonaceous particles with size less than 30 nm. The negligible effects of POC on larger particles were observed due to its honeycomb structure leads to an inadequate residence time to oxidize the solid particles or trap them. The particles removal efficiencies of CRDPFs had high degree of correlations with the emission ratio of NO_2/NOx. The PN emission results from two CRDPFs indicated that more NO_2 generating in diesel oxidation catalyst section could obtain the higher removal efficiency of solid particles. However this also increased the risk of NO_2 exposure in atmosphere.

Key words: continuously regenerating diesel particulate filter; particles oxidation catalyst; particle number; diesel engine; size distribution

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Introduction

Particulate matter and nitrogen oxide (NOx) emissions from diesel engines are being recognized as the pollutants having adverse effects on the environment as well as on human health. In epidemiological studies, close associations have been observed consistently and coherently between ambient concentrations of particulate matter and morbidity and mortality (Wichmann and Peters, 2000). The growing recognition of the harmful effects of diesel emissions on air quality and human health led the environment protect agency (EPA) to propose stricter regulations. To meet more and more stringent legislation, diesel manufacturers have made significant improvements in the technology of diesel engine design and control, but further reductions are still required to meet current or future legislation (Johnson, 2008). Diesel particulate filters (DPF) commonly known as soot filter is used to control the particulate matter (PM) pollutants emanated from diesel engine exhaust. Among the number of commercially available DPFs, continuously regenerating diesel particulate filter (CRDPF) has been proved to be very effective and promising technology for the abatement of PM emissions from the diesel engine exhaust, which normally consists of a diesel oxidation catalyst (DOC) followed by a ceramic particulate filter.

The CRDPF technology utilizes the NO_2 to combust the soot collected in a particulate filter (Copper and Thoss, 1989). NO_2 is a stronger oxidant than O_2 promoting low temperature oxidation of soot in the range 200–500°C, which has also been postulated to have a synergistic role with O_2 in the combustion of diesel soot (Ehrburger et al., 2002; Setiabudi et al., 2004). Typically, NO_2 is 5% to 15% of the total NOx (less than 50 ppm) in the diesel exhaust. However, oxidation catalysts like Pt could oxidize NO to NO_2 increasing NO_2 concentrations to 50% of the total NOx, in the temperature range of 300–350°C (Ehrburger et al., 2002; Marques et al., 2004). The CRDPF system has an additional advantage of containing DOC which is responsible not only for the conversion of NO to NO_2 but also for the oxidation of HC and CO to CO_2 and H_2O. Therefore, this system has the potential to remove PM, CO and HC emissions simultaneously (Copper and Thoss, 1989; Kittelson et al., 2006).

However, the increasing NO_2 in ambient air make

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* Corresponding author. E-mail: geyunshan@bit.edu.cn

significant contributions to urban air pollution, and playing a critical role in the atmospheric photochemistry that produces ground level ozone (Whitby, 1991). NO_2 can participates in many atmospheric reactions, such as oxidizing the organic compounds to be some potent pollutant species (Tang et al., 2004). Hong Kong EPA found the NO_2 annual average concentration nearby roadside is 10 times higher than the level in rural area (Hong Kong SAR, 2006).

The CRDPF requires regular active regeneration and periodical ash removal by external means to avoid blocking of the exhaust line (Lehtoranta et al., 2007, 2009), especially for those vehicles in developing countries, because they have relative high emission level and are always operated at low load and velocity for long periods of time. It might need an additional means of active regeneration (Karila et al., 2004). To avoid these blocking risks together with the complex regeneration and cleaning procedures, POC was developed which similar to CRDPF. The difference is that the DPF section of POC using honeycomb structure which traps no particles instead of alternately plugged channels with porous walls. This means that POC have no risk of clogging, ensuring trouble-free operation (Rens and Wilde, 2005).

Two CRDPFs with different configurations and one POC were employed in this study to perform experiments from a diesel engine under controlled laboratory operating and dilution conditions. The primary objective of the current experiments aimed at investigating the contribution of NO_2 to total NOx after the test engine retrofitted with after-treatments and evaluating the performance between CRDPFs and POC for the decreasing of PM emission. The obtained results are of importance for understanding of the impaction of after-treatments on the emissions emitted from diesel engines and for estimating their contribution to the environment comprehensively.

1 Materials and methods

1.1 Experimental setting-up

The performance of the CRDPFs and POC were tested on engine test bench with emission measurement system (Fig. 1). The results of emissions, fuel consumption and relevant engine parameters were recorded in dynamometer control system. The emission measurements were performed at the upstream and downstream of after-treatments.

To decrease the particle concentrations and temperature to be tolerated by the instruments and to prevent condensation and nucleation, two stages of ejector diluter (ED) were used for sampling from raw exhaust stack. The dilution air of the first stage of ED diluter was preheated to 150°C and its probe was heated to 195°C to act as a hot dilution system, which will not only made the saturation ratio at low level, but also possible to vaporize those artificial nucleation particles by heating the sample gas to a temperature where they will be re-evaporated again (Burtscher, 2005).

1.2 Measurement and specifications of after-treatments

The SEMTECH-DS (Sensor, USA) which equipped a non-dispersive ultraviolet (NDUV) analyzer was used to measure nitric oxide (NO) and nitrogen dioxide (NO_2) separately, while the smoke opacity was measured with the help of an opacimeter AVL 439. An electrical low pressure impactor (ELPI, Dekati, Finland) was used for the investigation of particle number-size distributions downstream of the ejector dilution system. The dilution ratio of the dilution system was determined by using CO_2 as tracer gas.

The after-treatments employed in this study were recognized as CRDPF-K (SK Energy Korea Holding Co., Ltd.), CRDPF-C (SK energy China Holding Co., Ltd.), and

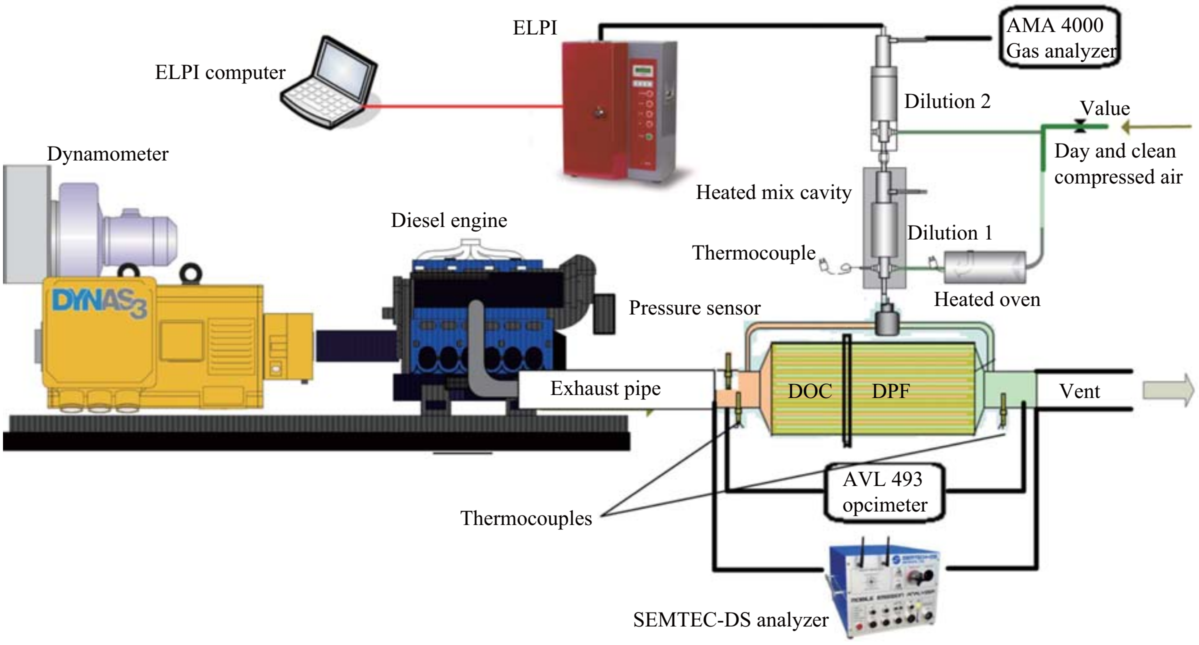

Fig. 1 Schematic diagram of experimental systems.

Table 1 Specifications of the CRDPF systems

Parameter	POC	CRDPF-K	CRDPF-C
DOC substrate	Cordierite	Cordierite	Metal
DOC cell density (cells/in^2)	400	400	600
DOC Pt. content (g/ft^3)	50	50	50
Diameter × Length (inch × inch)	6.5 × 5.5	8.5 × 3.5	8.5 × 3.5
DPF substrate	Cordierite	Cordierite	Cordierite
DPF cell density (cells/inch^2)	380	200	100
DPF Pt. content (g/ft^3)	35	35	35
Diameter × Length (inch × inch)	6.5 × 7.0	8.5 × 14.0	8.5 × 14.0

Table 2 Specifications of the test engine

Parameter	Feature/Size
Engine type	Diesel, 4-stroke, 4-cylinders in- line
Air intake system	Inter-cooled, turbocharged
Fuel metering system	High pressure common rail
Displacement (L)	2.8
Bore × Stroke (mm)	93 × 10^2
Compression ratio	18.2:1
Max. Power (kW@r/min)	75/3600
Max. Torque (Nm@r/min)	250/1900

POC separately. The specifications are shown in Table 1. These units are made up of two chambers where the soot trapping/combustion step is preceded by the oxidation process in the inlet. The first chamber commonly is known as DOC which consists of a ceramic honeycomb substrate coated with Pt metal. The second chamber of CRDPF is wall-flow monoliths consist of channels with porous walls that are alternately plugged, which force exhaust gas to stream through these porous walls leaving the particles behind at the wall. Whereas, the second chamber of POC is tortuous honeycomb channels. This structure directs the exhaust gas to either follow the tortuous channels or to go through the substrate walls (Lehtoranta et al., 2009; Vakkilainen and Lylykangas, 2004).

During the tests, the back pressures were monitored continuously and the temperature of engine exhaust was monitored at the inlets and outlets of after treatments with k-type thermocouple.

1.3 Test engine and operating cycle

The tests were performed on a 4-stroke 4-cylinder turbocharged inter-cooled diesel engine. The specifications of the engine are listed in Table 2. The fuel used in this study was locally available commercial low sulfur (50 ppm) diesel.

The experiments were performed in accordance with the European steady-state cycle (ESC) test cycle. According to test cycle, the engine was run under a speed N_A = 2125 r/min at 100%, 75%, 50%, and 25% of full load for mode A100, A75, A50 and A25, respectively. Then it was operated under a speed N_B = 2660 r/min at 100%, 75%, 50%, and 25% of full load during the modes B100, B75, B50 and B25, respectively. Finally, it was run under a speed N_C = 3200 r/min at 100%, 75%, 50%, and 25% of full loads for mode C100, C75, C50 and C25, respectively. The idle speed of the given engine was 800 r/min.

2 Results and discussion

2.1 HC and CO emissions

The removal efficiencies of all the given after-treatments for HC and CO emissions were higher than 90% and 95% respectively over all the test models, which have been discussed in another article (Liu et al., 2011).

2.2 NO*x* emissions

NO*x* pollutants are lower at low load modes (Fig. 2). They tended to increase with the increasing of load level and, thus became maximal at full load modes for all the three speeds N_A, N_B and N_C of the ESC test. The maximum NO*x* pollutants at full load are attributed to the higher temperature developed in the combustion chamber of the cylinder. Figure 2 reflects the emissions of NO*x* pollutants with and without after-treatments, which indicates that there are no significant NO*x* reductions observed.

2.3 NO_2 emission

It is worth noting that the contribution of NO_2 to total NO*x* increased dramatically. Normally, the total NO*x* emissions emitted from diesel engine are dominated by NO (Sher, 1998) (Fig. 3a). Under most of engine modes the NO_2/NOx ratios upstream of after-treatments are below 10%, except under some light-duty and high speed modes it slightly above 10%. However, this situation was signif-

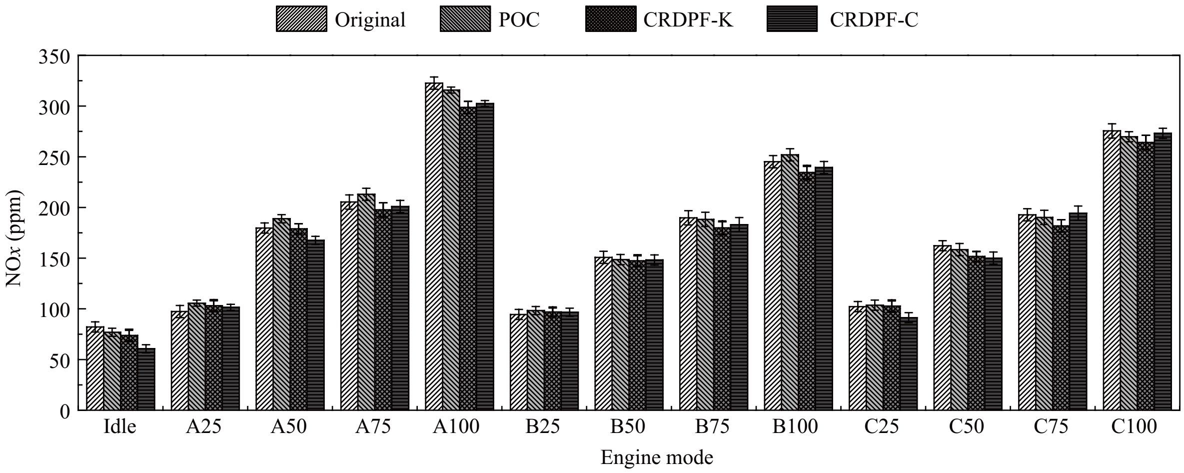

Fig. 2 NO*x* emissions concentrations after treatments at inlet and outlet. Engine modes refer to Section 1.3.

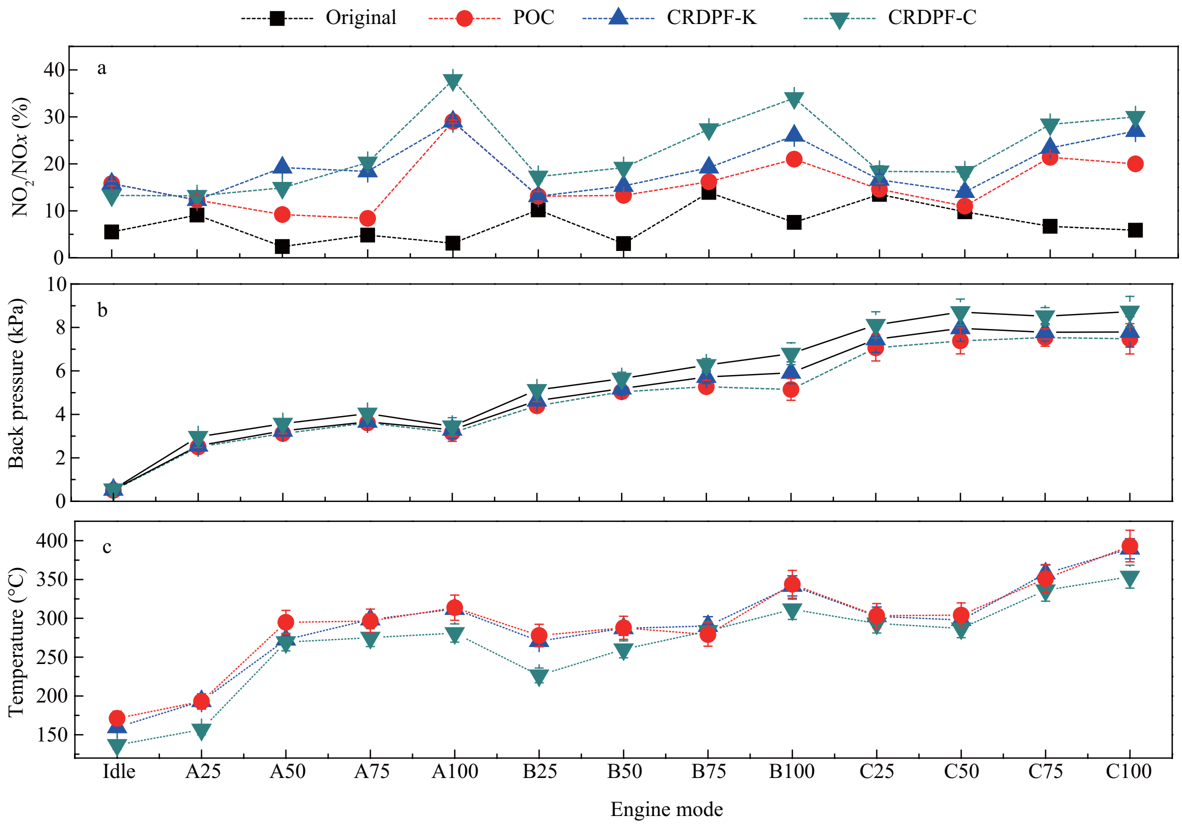

Fig. 3 The ratio of NO_2/NOx in the exhaust after treatments inlet and outlet (a), back pressure at three after treatments upstream (b), and temperature at three after treatment downstream (c). POC: particle oxidation catalyst; CRDPE: continuously regenerating diesel particulate filter.

icantly changed with the application of after-treatments. As can be seen (Fig. 3), the ratios of NO_2 to NOx at the outlet of the after-treatments are significantly higher than the original exhaust. Especially under high load modes, the concentration of NO_2 downstream of the CRDPF-K, CRDPF-C and POC constituted approximately 30%–40%, 25%–30% and 20%–30% of the total NOx, respectively. During the whole test, CRDPF-C has the highest ratio of NO_2 to NOx while POC has the lowest. The differences, on one hand, would be caused by the different DOC cell densities of the three after-treatments. On the other hand, the temperature inside the after-treatments also could affect the oxidation of NO to NO_2 (Fig. 3c).

The NO oxidation is the thermodynamic equilibrium limitation, NO_2 decomposing to O_2 and NO at temperatures higher than 350°C, depending on the oxygen partial pressure and the presence of a catalyst (Majewski et al., 1995).

Above information indicate that the superfluous NO_2 will pass through the after-treatments and emit into the atmospherical circumstance. The direct primary emission of NO_2 is a significant concern due to NO_2 has a higher toxicity as compared to NO and would lead to increased photochemical ozone production (Carslaw and Beevers, 2004). Some previous studies shown NO_2 constituted one-third of the total NOx (Shorter et al., 2008) or even up to 40%–50% of the total NOx depending on the drive cycle (Ayala et al., 2002; Tang et al., 2004) after the test engine retrofitted with CRDPF. A large scale study conducted by Danish Road Safety and Transport Agency showed that primary NO_2 increased from approximately 5% without a CRDPF to 15%–20% by volume with a CRDPF (Danish Road Safety and Transport Agency, 2002).

2.4 Smoke emissions

As the results of the smoke meter measurement from the attenuation of visible light in the measuring chamber, the smoke density value is in effect the result of elemental carbon (black smoke) also those sulfates and soluble organic fraction (SOF) attached on its surface and hydrocarbon vapor (blue smoke). In other word, the measurement of the AVL 439 opacimeter indicates the emission level of total PM.

The application of the after-treatments greatly influenced the smoke emissions as depicted in Fig. 4. Here, the reduction ratio (RR) is defined as the ratio of emission under downstream of the after-treatments divide under upstream. As can be seen, the RRs of smoke across the CRDPF-C are more than 95.5% at all modes of the ESC test, except idle mode where the smoke RR falls to 83.3%. Regarding of the CRDPF-K system, the smoke RRs vary from 81.7% to 95.8% at various engine modes. The smoke RRs of the POC fluctuate from 40% to 70%.

The POC used honeycomb structure rather than alternately plugged channels with porous walls to decrease the risk of clogging. The back pressures (Fig. 3b) indicate that the POC have the lowest back pressure over all the test models. However, this also increased the possibility of the solid particle escaping from the after-treatment for the shorter residence time. In fact, the RRs of the POC majorly depend on the oxidation of the SOF in the DOC section. This is also confirmed by the results of particle number

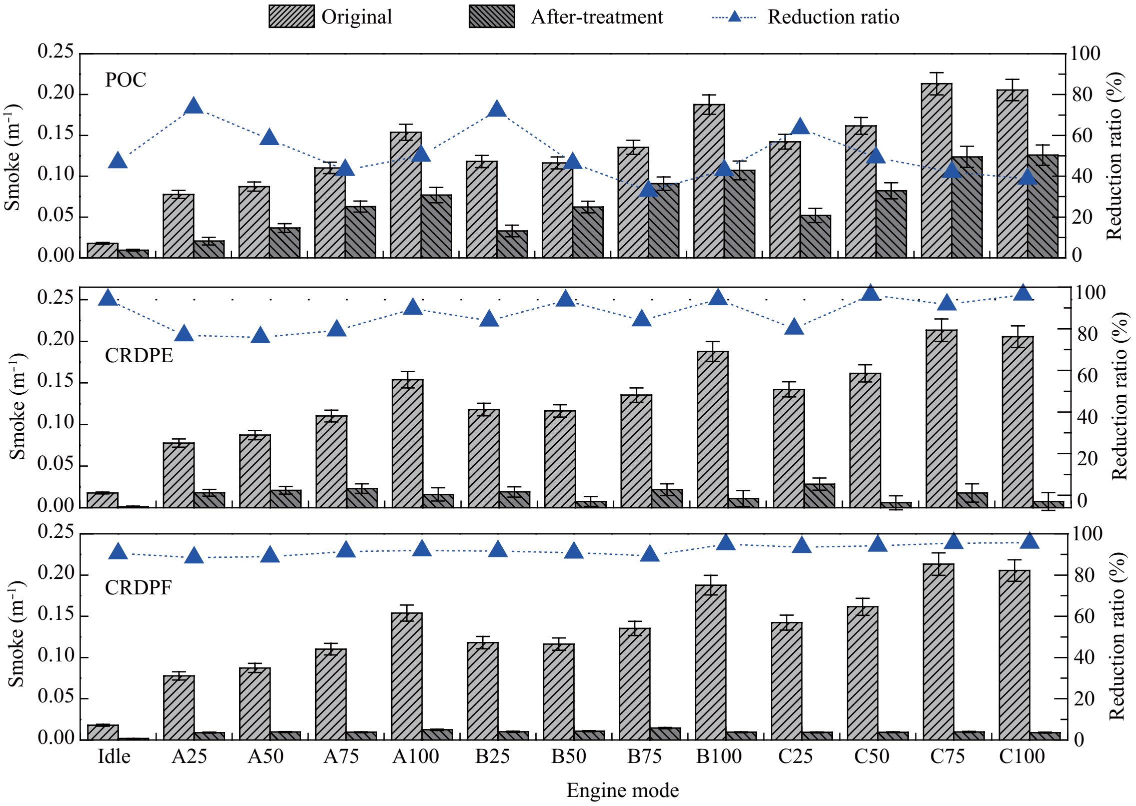

Fig. 4 Smoke emissions and the impact of POC, CRDPF-K and CRDPF-C on their reduction. CRDPF: continuously regenerating diesel particulate filter; POC: particles oxidation catalyst.

emission which will be addressed later in this article.

2.5 Particle number emissions

The ELPI in combination with aerodynamic size classification and the electrical detection was used to determine the particle number concentrations for the particles with size in the range from 7 nm to 10 µm. The particles in the size range 0.5–10 µm (corresponding to the stage 8–12 of EPLI) typically were recognized as artifacts which previously deposited on the internal surface of exhaust pipe and re-entrainment as agglomerate particles hence will not be considered in the following analysis.

Particular number (PN) emission will be introduced into Euro 5 and Euro 6 regulation in addition to the mass-based limits. The comparison of total PN emission from the given engine under ESC 13 models with and without after-treatments (Fig. 5) indicate that the CRDPFs decreased PN concentrations dramatically but the influence of the POC on PN emissions are negligible. It is worth pointing out that the first stage of the dilution system used in the study is a hot dilution, which is able to keep the saturation ratio at a low level and vaporize the nucleation particles by heating the sample gas to a temperature where they would be evaporate again. In this case, the measured PN emissions during the tests are only solid particles.

The smoke emission in the preceding paragraph indicat-

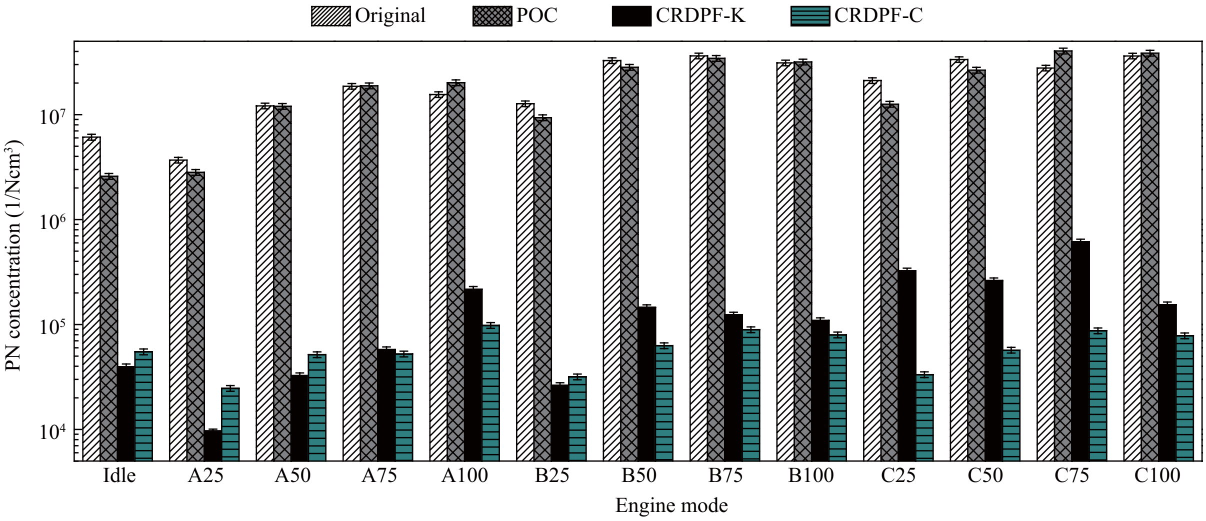

Fig. 5 Comparison of total particle numbers (PN) from test engine at 13 test models with and without after-treatments.

ed that the POC is able to reduce 40% to 70% of total PM emission, however the reductions for solid particles under some models are so slightly that can be negligible. By comparison, the CRDPF-K reduced more than 85% of total PM and more than 97% of solid particles and the CRDPF-C reduced more than 95% of total PM and more than 99% of solid particles under most of the test models except idle.

The comparison of brake specific (BS) PN emissions generated from the test engine under the whole ESC cycle with and without after-treatments (Fig. 6) indicate that the solid PN emission were decreased by 99.3% and 99.8% with the application of the CRDPF-K and the CRDPF-C respectively. This finding is in good agreement with those of other studies reported that particle trapping efficiency of the CRDPF system was about 99% (Holmen and Ayala, 2002; Lanni et al., 2001). However, the reduction of BS solid particle number emission caused by the POC under the ESC cycle was 24%. The DPF section of the POC used honeycomb structure to decrease the risk of clogging, consequently, it also reduced the removal efficiency of solid particles for unable to trap them.

2.6 Particle size distributions

The number size distributions are weighted according to the weight factors listed on emission regulation. The ELPI equipped with a filter stage which means the cutoff point of its lower particle is 7 nm. The number concentrations were corrected with dilution ratio. It is important to note that these distributions are shown with the aerodynamic diameter.

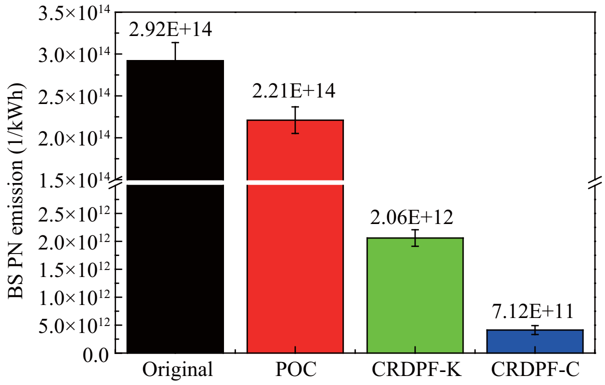

Fig. 6 Comparison of brake specific (BS) PN emissions from test engine at ESC test with and without after-treatments.

Figure 7 illustrates the comparison of the weighted number-size distribution of particles emitted from the test engine with and without after-treatments under the ESC cycle. In order to display clearly, the graph was separated into two due to the PN concentrations collected from the CRDPF-K and the CRDPF-C were two or three orders of magnitude less than from engine without after-treatments. As can be seen, the differences between the different after-treatments are not so much to a change in size distribution but to a change in the quantity of emitted particles. The number mean diameter of the number size distributions is quite stable at about 60 nm aerodynamic diameter.

The comparison of the PN size distribution between the test engine without after-treatment and with POC demonstrate that POC was able to reduce approximately 60% of the number concentration of the particles with size less than 30 nm. However, for the larger particles, there is no substantial removal efficiency observed due to the honeycomb structure of the POC without enough residence time to oxidize or trap them.

It is of great concern to note that, with the application of the CRDPF, the PN concentrations downstream of the after-treatment have high degree of correlations with the ratio of NO_2/NOx. The relationship between the NO_2/NOx ratios and the PN concentrations from the CRDPF-K and the CRDPF-C are plotted (Fig. 8). When the NO_2/NOx ratios of the CRDPF-K are higher than the CRDPF-C (see two shaded models in Fig. 8), the PN concentrations downstream of the CRDPF-K are lower than the CRDPF-C, and vice versa. This indicates that more NO_2 is good at the removal of solid particles but also increase the risk of emitting NO_2 into atmospheric environment. That will be a critical issue to find a trade-off when the CRDPF is under development.

In spite of the usage of 50 ppm sulfur content diesel in this research, particle removal efficiencies of the CRDPF systems are remarkably high. Although the two CRDPFs showed slightly difference in their trapping efficiencies, they played a significant role in the reduction of both nuclei

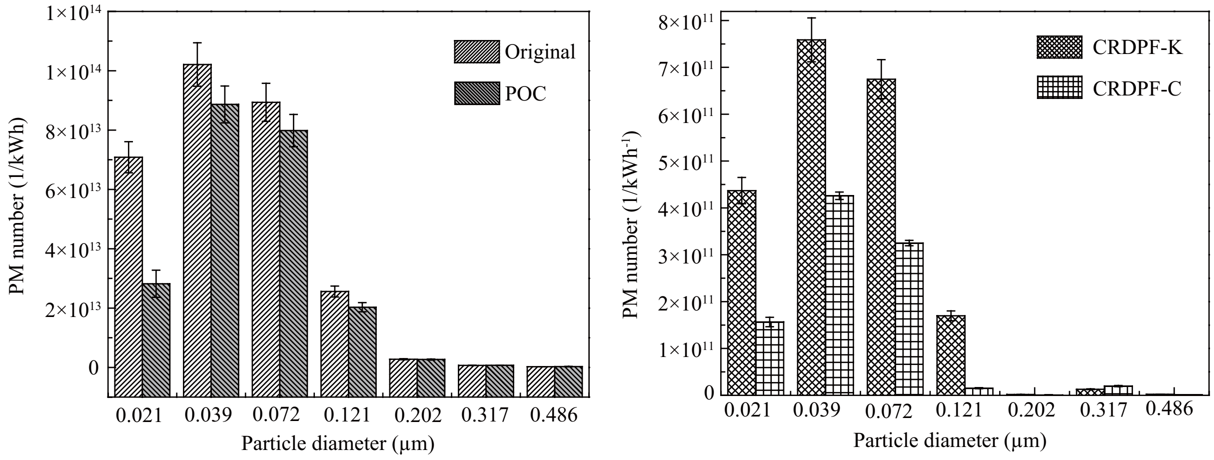

Fig. 7 Weighted number-size distribution of the particles emitted from test engine under ESC cycle with and without after-treatments (due to the PN emissions from CRDPF-K and CRDPF-C were much lower than those from original and POC, the graph was separated into two to display more clearly).

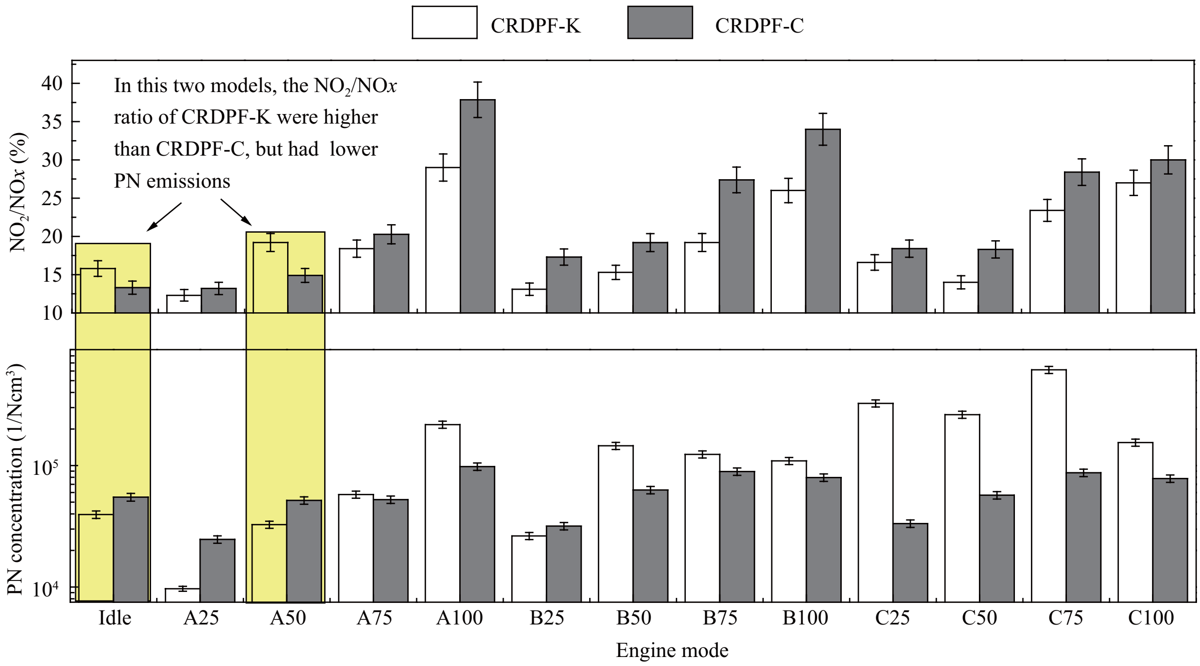

Fig. 8 Relationship between the NO_2/NOx ratios and the PN concentrations from the CRDPF-K and the CRDPF-C.

mode as well as accumulation mode particles. However, the POC does not reduce the carbonaceous particles emitted from the test engine significantly.

3 Conclusions

Two CRDPFs and one POC after-treatment systems were employed to investigate their impacts on the emission ratio of NO_2/NOx and RR of particle emissions emitted from a diesel engine. The CRDPFs did not exhibit any significant impacts on the conversion of NOx. However, the emission ratios of NO_2/NOx were significantly increased after the given engine was retrofitted with the after-treatments. Especially under high load modes, NO_2 concentration of the CRDPF-K, the CRDPF-C and the POC constituted approximately 30% to 40%, 25% to 30% and 20% to 30% of the total NOx, respectively.

PM emissions from the given diesel engine were greatly reduced by the application of the after-treatments. The results of smoke emissions and PN emissions indicated that the CRDPF-K and the CRDPF-C both had sufficient capacities to remove more than 90% of total PM and more than 97% of solid particles. However the POC was only able to remove the organic components.

The DPF section of the POC using honeycomb structure rather than alternately plugged channels with porous walls to decrease the risk of clogging; consequently, it also reduced the removal efficiency of carbonaceous particles for the inadequate residence time. The PN size distribution demonstrated that the POC was able to remove partial of the particles with size less than 30 nm and had negligible effects to larger particles.

The particle removal efficiencies of the CRDPFs had high degree of correlations with the emission ratio of NO_2/NOx. The PN emission results from two CRDPF indicated that the more NO_2 generated in the DOC section the higher particles removal efficiencies were observed, however which also increase the risk of NO_2 exposure in the atmosphere. It is very important find a trade-off of particle removal efficiency and the potential of NO_2 exposure when developing and evaluating the performance of CRDPF.

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Appendix A. Supplementary data

Supplementary figures associated with this article can be found in the online version.

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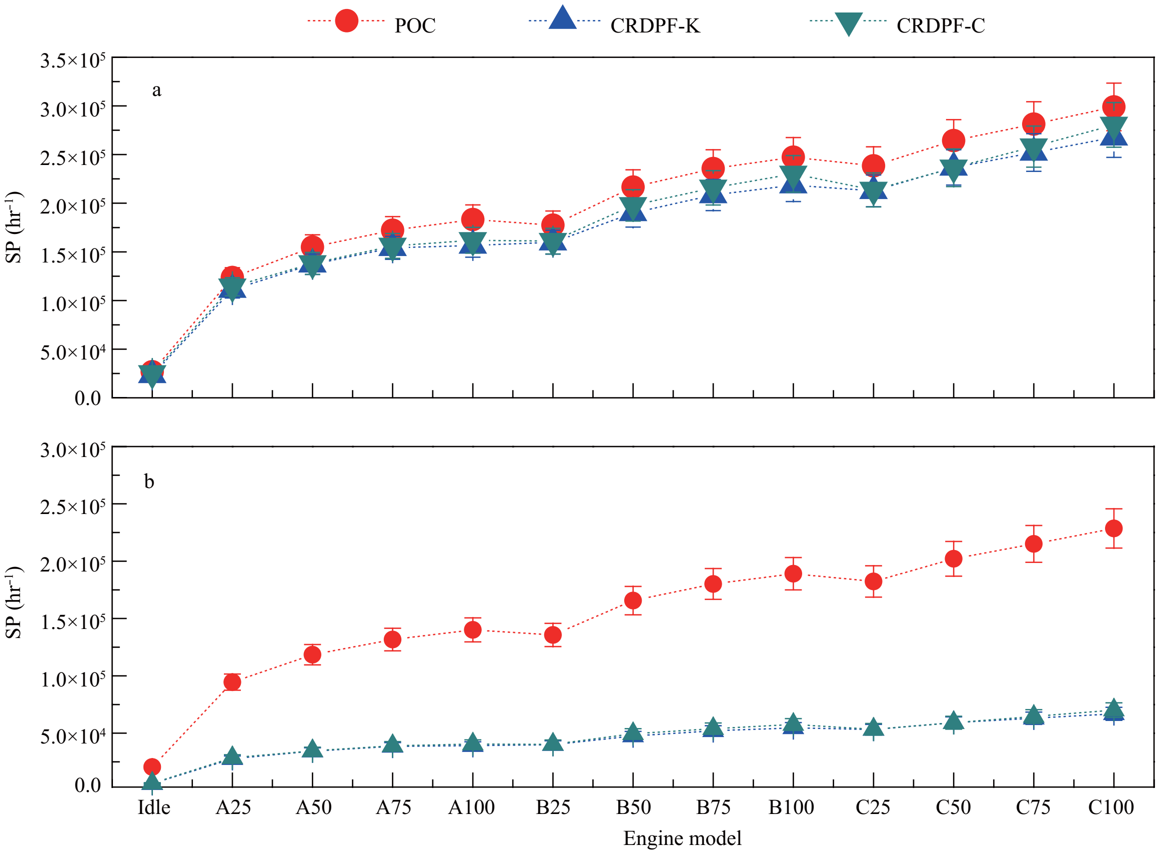
Appendix material



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