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High NO₂/NO_x emissions downstream of the catalytic diesel particulate filter: An influencing factor study

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ABSTRACT

Diesel vehicles are responsible for most of the traffic-related nitrogen oxide (NO_x) emissions, including nitric oxide (NO) and nitrogen dioxide (NO₂). The use of after-treatment devices increases the risk of high NO₂/NO_x emissions from diesel engines. In order to investigate the factors influencing NO₂/NO_x emissions, an emission experiment was carried out on a high pressure common-rail, turbocharged diesel engine with a catalytic diesel particulate filter (CDPF). NO₂ was measured by a non-dispersive ultraviolet analyzer with raw exhaust sampling. The experimental results show that the NO₂/NO_x ratios downstream of the CDPF range around 20%–83%, which are significantly higher than those upstream of the CDPF. The exhaust temperature is a decisive factor influencing the NO₂/NO_x emissions. The maximum NO₂/NO_x emission appears at the exhaust temperature of 350°C. The space velocity, engine-out PM/NO_x ratio (mass based) and CO conversion ratio are secondary factors. At a constant exhaust temperature, the NO₂/NO_x emissions decreased with increasing space velocity and engine-out PM/NO_x ratio. When the CO conversion ratios range from 80% to 90%, the NO₂/NO_x emissions remain at a high level.

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Introduction

Nitrogen dioxide (NO₂) represents an important urban pollutant in most countries. Epidemiologic studies show strong evidence for an association between NO₂ exposure and adverse human health effects, especially respiratory morbidity (Wolfe and Patz, 2002; EPA US, 2008). Besides its toxicity to humans, NO₂ can also lead to increasing photochemical ozone production, which then impacts the air environment (Zielinska, 2005; Ma et al., 2013). Concerns over the health impacts of NO₂ have prompted legislation at a national and international level (EPA US, 2010). In Europe, the EU First Daughter Directive (99/30/EC) sets an annual mean limit of

40 μg/m³ and an hourly limit of 200 μg/m³ not to be exceeded on more than 18 occasions each year (Council Directive, 1999/30/EC, 1999).

With the enforcement of stringent emission standards, the nitrogen oxide (NO_x) concentration in ambient air showed a decreasing trend, however, the NO₂ emission was not alleviated, and even rose in some cities, which has led to the NO₂ fraction in NO_x (NO₂/NO_x) spiraling recently (Minoura and Ito, 2010; Mavroidis and Chaloulakou, 2011; Tian et al., 2011). Carslaw (2005) and Carslaw et al. (2007) undertook statistical analysis of roadside concentrations of NO₂ in London and indicated that the increased use of diesel particulate filters (DPFs) fitted to buses made an important contribution to the

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increasing trends in NO₂/NO_x emissions. A study reported that the NO₂/NO_x road traffic emission ratio had increased from 14% in 1992 to 23% in 2004 in Switzerland, and this trend had been caused by increasing primary NO₂ road traffic emissions (Hueglin et al., 2006). The primary NO₂ emissions from road traffic were assessed for ten case study locations across the European Union, and the primary NO₂ was predicted to increase further to an average of 32.0% in 2020 (Grice et al., 2009). The long-term trend in emission ratio of NO₂ to NO_x was analyzed in relation to traffic activities using ambient monitoring data in Seoul, and the authors suggested that the diesel particulate filter (DPF) or diesel oxidation catalyst (DOC) had direct influences on the primary NO₂ values at urban roadside sites (Shon et al., 2011).

In order to decrease the levels of harmful pollutants, after-treatment systems are widely used in diesel engines. Wall-flow DPFs are considered to be the key technology to detoxify diesel exhaust (Mayer et al., 2000; Johnson, 2009). To avoid an increase of the exhaust back pressure and realize the regeneration of DPF, a catalyst is usually employed to convert nitric oxide (NO) to NO₂, which is a powerful oxidizing agent supporting the oxidation of soot (Kim et al., 2010; Shrivastava et al., 2010). The catalytic DPF (CDPF) increases the risk of high NO₂/NO_x emissions of diesel engines. The real-world NO and NO₂ emissions of modern vehicles were studied and the results indicated that the NO₂/NO_x emissions of Euro III cars or Euro IV vehicles without DPF were lower than those of Euro IV vehicles with DPF, reaching the maximum proportions of 35%–70% (Alvarez et al., 2008). The research of Heeb et al. (2010) showed that the DPFs with high oxidation potential induced NO₂ formation up to 3.3 ± 0.7 g/kWh, whereas low oxidation potential DPFs reduced the NO₂ emissions. The after-treatments did not exhibit significant impacts on the conversion of NO_x, however, the emission ratios of NO₂/NO_x were significantly increased for an engine equipped with two CDPFs and one particle oxidation catalyst (POC) (Liu et al., 2012). Furthermore, NO₂/NO_x ratio variation affects the conversion efficiency and the catalyst chosen in the selective catalytic reduction (SCR) system that follows the filter to meet the Euro IV diesel engine emission standard (Liu et al., 2010; Colombo et al., 2012).

Although many researchers have reported the phenomenon of high NO₂/NO_x emissions by CDPFs, the formation mechanism is seldom studied. The origins of NO₂-production were investigated by the after-treatment industry (Spruk et al., 2010; Czerwinski et al., 2013), who paid attention to the parameters of CDPF. High content of Pt in the coating and low space velocity were identified as the reason for NO₂ increase. However, the role of engine-out parameters in the increase of NO₂/NO_x emissions has barely been considered.

In this paper, an experiment on a diesel engine with CDPF was carried out and the influencing factors on NO₂/NO_x emissions were investigated, such as exhaust temperatures, space velocity, carbon monoxide (CO) conversion ratio, and engine-out particulate matter (PM) emissions. The work should be beneficial to the further optimization potential of diesel engine–CDPF system developments to reduce NO₂/NO_x emissions.

1. Materials and methods

1.1. Experimental setup

The test engine is a direct injection, high pressure common-rail, turbocharged diesel engine (6DF3, FAW-WDEW, Wuxi, China), whose characteristics are shown in Table 1. The specifications of the CDPF are shown in Table 2. The engine with CDPF was tested on an engine test bench based on an AC dynamometer (HT350, Schenck, Darmstadt, Hessen, Germany) with emission measurement system (Fig. 1). A gas analyzer (SEMTECH-DS, Sensor, Saline, Michigan, USA) equipped with a non-dispersive ultraviolet (NDUV) analyzer was used to measure NO and NO₂, as well as carbon monoxide (CO) and hydrocarbons (HC). An electrical low pressure impactor (ELPI, Dekati, Kangasala, Finland) was used for the investigation of particulate matter (PM). To prevent particle condensation and nucleation, two stages of ejector diluter (ED) upstream ELPI were used for sampling from the raw exhaust stack (Burtscher, 2005). The fuel used in this study was locally available commercial low sulfur (50 ppm) diesel.

1.2. Test methods

Because bag sampling has the potential to convert NO to NO₂, the direct sampling of raw exhaust was employed to measure NO₂ (Genese et al., 2006). The exhaust pollutant, temperature and pressure measurements were performed upstream and downstream of the after-treatment device. The results of emissions, fuel consumption and relevant engine parameters were recorded by the dynamometer control system. The stationary operation points (OPs), so called step-tests, were

Table 1 – Specifications of the test engine.

Parameter	Feature/size
Engine type	4-stroke, 6-cylinder, in-line
Bore (mm) × stroke (mm)	107 × 125
Displacement (L)	6.7
Compression ratio	16.8
Fuel system	High pressure common rail (BOSCH)
Max. power (kW at 2300 r/min)	147
Max. torque (N·m at 1400 r/min)	760

Table 2 – Specification of catalytic diesel particulate filter (CDPF).

Parameter	Feature/size
DOC substrate	Cordierite
DOC cell density (cells/in ²)	400
DOC Pt content (g/ft ³)	50
DOC diameter (mm) × length (mm)	215.9 × 88.9
DPF substrate	Cordierite
DPF cell density (cells/in ²)	200
DPF Pt content (g/ft ³)	35
DPF diameter (mm) × length (mm)	215.9 × 355.6
DOC: diesel oxidation catalyst.	

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