REVIEW ON CHARACTERIZATION OF NANO-PARTICLE EMISSIONS AND PM MORPHOLOGY FROM INTERNAL COMBUSTION ENGINES: PART 1

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ABSTRACT—This paper is review of the characterization of exhaust particles from state-of-the-art internal combustion engines. We primarily focus on identifying the physical and chemical properties of nano-particles, i.e., the concentration, size distribution, and particulate matter (PM) morphology. Stringent emissions regulations of the Euro 6 and the LEV III require a substantial reduction in the PM emissions from vehicles, and improvements in human health effects. Advances in powertrains with sophisticated engine control strategies and engine after-treatment technologies have significantly improved PM emission levels, motivating the development of new particle measurement instruments and chemical analysis procedures. In this paper, recent research trends are reviewed for physical and chemical PM characterization methods for gasoline and diesel fueled engines under various vehicle certification cycles and real-world driving conditions. The effects of engine technologies, fuels, and engine lubricant oils on exhaust PM morphology and compositions are also discussed.

KEY WORDS: Particulate matter, Gasoline direct injection, Engine lubricant, Filter regeneration, PM morphology

1. INTRODUCTION

The enforcement of more stringent vehicle emissions regulations, carbon dioxide (CO$_2$), and fuel economy (FE) standards has compelled automotive manufacturers to reduce pollutant emissions from vehicles and to develop energy-efficient internal combustion engines. The implementation of stricter emission standards in the transportation sector can make positive effects on improvement of urban air quality and human health (Denner, 2013; Schöppe et al., 2013).

The introduction of advanced vehicle emissions control technologies for gasoline, diesel, gas fuelled vehicles has resulted in a substantial reduction of hazardous air pollutants (HAPs), including both regulated and non-regulated harmful emissions. However, the steady growth of vehicle populations in metropolitan areas has resulted in frequent exceedances of the established environmental quality standards for ozone (O$_3$), particulate matter (PM), and nitrogen oxides (NOx) (Eastwood, 2008; Lu et al., 2012; Myung and Park, 2012; Rahman et al., 2013).

Many studies have shown that there are substantial differences in the fuel consumption and exhaust emissions between vehicle certification modes and on-road driving testing in the European Union (EU), therefore, a new driving cycle known as the worldwide-harmonized light-duty test cycle (WLTC) was developed to accurately reflect real-world vehicle driving patterns. Furthermore, a complementary emission test procedure was introduced that simulates real-world operating conditions, i.e., portable emissions measurement systems (PEMS) testing, which are considered to be the most promising measures for reducing actual gaseous tailpipe emissions from engines and vehicles (Kirchner et al., 2013; Johnson et al., 2011; Mock et al., 2013; Rakopoulos and Giakoumis, 2009).

There are significant opportunities for developing downsizing gasoline direct injection (GDI) engines and turbocharged diesel engines that meet global vehicle fuel economy and CO$_2$ emission standards. Regulated gaseous emissions, except particle number (PN) emissions, from GDI vehicles can be significantly reduced by using advanced three-way catalysts (TWCs) and stoichiometric operation (Arsie et al., 2013; Basshuysen, 2009; Hwang et al., 2012; Hassaneen et al., 2011; Momenimovahed et al., 2013). The use of a diesel particulate filter (DPF) with a diesel oxidation catalyst (DOC) in new diesel engine results in PM and PN emission levels that are comparable to those from port fuel injection (PFI) engines and much lower than those from GDI engines (Brijesh and Sreedhara, 2013; Eiglmeier et al., 2011; Giechaskiel et al., 2012, 2014; Wang et al., 2012). However, many studies have reported that considerable nano-particle emissions are produced by vehicles operating under transient running conditions at cold ambient temperatures and during particle regeneration periods. Several studies have also shown that

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PM formation from internal combustion engines can be substantially reduced via a combination of engine hardware modifications, precise air-fuel mixture preparation, and sophisticated engine control strategies during transient engine operations (Basshuysen, 2009; Fan et al., 2012; Kim et al., 2013b; Neufler et al., 2013; Ohm, 2013; Rakopoulos and Giakoumis, 2009; Zhao, 2010).

Carbonaceous PM particles from internal combustion engines are composed of various harmful substances, and their particle size distributions, morphologies, and chemical compositions have been characterized in detail (Barone et al., 2012; Gaddam and Vander Wal, 2013; Paul et al., 2013). The nanostructure of PM particles depends on various engine operating conditions and after-treatment devices, which has motivated the development of microscopic evaluation techniques, i.e., scanning electron microscopy (SEM) and transmission electron microscopy (TEM) (Burtscher, 2005; Bzdek et al., 2012; Eastwood, 2008; Maricq, 2007; Lee et al., 2013a). Recently, there has been growing concern regarding engine oil-derived PM emissions, which are associated with increasing vehicle mileage accumulation and the deterioration of after-treatment performance. Combustion byproducts of engine oil containing various additives cause soot and ash deposits in the combustion chamber and after-treatment devices, and deteriorate the durability of the after-treatment systems. The inhalation of ultrafine particles of mixed heavy metal elements has adverse human health effects (Brandenberger et al., 2005; Eastwood, 2008; Jung et al., 2003; La Rocca et al., 2013; Liati and Eggenschwiler, 2010; Shim et al., 2013).

The EU has proposed a solid particle PN (#/km) limit in Euro 6b and Euro 6c, which will be enacted in September 2014, for the type-approval of GDI light-duty vehicles (LDVs) of $6.0 \times 10^5$-6.0 $\times 10^4$ (#/km) from 2014 to 2017 and $6.0 \times 10^3$ (#/km) from 2017, respectively. The PN limit in Euro 5b of $6.0 \times 10^3$ (#/km) for compression ignition (CI) LDVs has already been implemented as in September 2011. The test procedure for the PM mass (mg/km) emissions limit has been defined in United Nations Regulation 83 as 4.5 mg/km for both GDI and diesel passenger vehicles in the Euro 5b standard. The PN (#/kWh) limits in Euro 6 for type-approval heavy-duty (HD) diesel engines were implemented in December 2012 and are as follows: $6.0 \times 10^3$ (#/kWh) for the worldwide heavy-duty transient cycle (WHHTC) and $8.0 \times 10^3$ (#/kWh) for the worldwide heavy-duty steady-state cycle (WHHSC). The PM mass (mg/kWh) limit in Euro 6 was 10 mg/kWh for both CI (WHHSC and WHHTC) and SI (WHHTC) heavy-duty engines (Delphi, 2013; Fraidl et al., 2012; Ko et al., 2012; Mamakos et al., 2012a; Myung and Park, 2012).

The California Air Resources Board (CARB) proposed the LEV III standards and a phase-in schedule for particulate emissions of 3.0 mg/mile as of 2017 MY and 1.0 mg/mile as of 2025-2028 MY for passenger cars, light-duty trucks (LDTs) and medium-duty passenger vehicles. The current PM limit is 10 mg/mile for passenger vehicles, and the PN (#/mile) standards have now been eliminated from the proposal. Gasoline particulate filters (GPFs) are very effective measures for reducing both the PM mass and the PN concentration: CARB expects that almost all GDI vehicles with GPFs will clear the 1.0 mg/mile PM standard of the LEV III PM regulations very easily (CARB, 2010; Delphi, 2013; Mamakos et al., 2011; Myung and Park, 2012; Richter et al., 2012; Zhang and McMahon, 2012).

In this study, we review the particle formation and reduction mechanism in internal combustion engines to provide comprehensive measures for meeting stringent particle number and mass regulations worldwide. We also review trends for the physical characteristics, i.e., the size distribution and morphology, and the chemical composition of exhaust carbonaceous soot particles, as well as the properties of various automotive fuels, lubricant compositions, and after-treatment systems.

2. NANO-PARTICLE EMISSIONS FROM INTERNAL COMBUSTION ENGINES

2.1. Characterization of Particulate Matter

In June 2012, the World Health Organization (WHO) International Agency for Research on Cancer (IARC) reclassified diesel exhaust emissions as ‘carcinogenic to humans’, which triggered controversy among automotive manufacturers. The primary claims made by industries are that PM/NOx components have been reduced through substantial improvements in combustion and after-treatment technologies of new diesel fuelled vehicles, which when combined with the use of DPF, deNOx systems and ultra-low sulfur diesel (ULSD) fuel, produce emissions under the Euro 5 standards (Borge, 2013). However, significant levels of PM/NOx emissions exhausted from old diesel vehicles on the road can aggravate human health and urban air quality. Additionally, concerns persist on how effectively all of these technologies that meet Euro 6 legislative standards work for reducing off-cycle emissions in real-world vehicles.