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Review of Vehicular Emissions Trends

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ABSTRACT

This review paper summarizes major developments in vehicular emissions regulations and technologies from 2014. The paper starts with the key regulatory advancements in the field, including newly proposed Non-Road Mobile Machinery regulations for 2019-20 in Europe, and the continuing developments towards real driving emissions (RDE) standards. An expert panel in India proposed a roadmap through 2025 for clean fuels and tailpipe regulations. LD (light duty) and HD (heavy-duty) engine technology continues showing marked improvements in engine efficiency. Key developments are summarized for gasoline and diesel engines to meet both the emerging NOx and GHG regulations. HD engines are demonstrating more than 50% brake thermal efficiency using methods that can reasonably be commercialized. Next, NOx control technologies are summarized, including SCR (selective catalytic reduction), lean NOx traps, and combination systems. Emphasis is on durability and control. Diesel PM (particulate matter) reduction findings are evolving around the behavior of the soot cake and PM sensors. Gasoline particulates are further described and gasoline particulate filter regeneration is now better understood. Oxidation catalysts mainly involve developments towards stubborn problems, like sulfur tolerance, low-temperature performance with exhaust with high hydrocarbon and CO, and methane oxidation. Finally, the paper discusses some key developments in gasoline gaseous emission control, focusing on meeting new regulatory requirements in the US, durability, and on lean burn gasoline emissions control.

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INTRODUCTION

Progress in emissions and fuel consumption reductions over the last 15 years or so have been very impressive, and it is continuing.

Since before the deliberations on LEVII (Low Emission Vehicle II) tailpipe standards in 1997 to the start of implementation of LEVIII standards in 2015, the automotive industry has faced a total tightening of emissions standards of about 97%. On-road heavy-duty (HD) trucks and non-road (NR) equipment faced similar reductions over the same time period. The challenges are continuing as the LEVIII and US Tier 3 implementation proceeds through 2025, the EU shifts to an emphasis on real-driving emissions (RDE), and the California HD and European NR sectors tighten further.

Similarly, since about 2000 light-duty (LD) greenhouse gas (GHG) emissions have dropped about 25-30%, with further reductions being required on the order of 25-30% through 2021. HD engine fuel consumption has dropped about 10% over the last 15-20 years with further reductions coming, perhaps another 10% through 2025.

These two trends have put a tremendous burden on the vehicle industry, with many experts proclaiming that these continuing challenges are among the greatest the industry has ever faced. In response, the technical community has been very active, with hundreds of papers and presentations on emissions control, and another thousand or more on engine technology. A high-level review of these developments from the last year is presented here. As in previous years (<u>1</u>), this review paper covers representative regulatory, engine technology, and emission control technology developments reported in 2014 and is not intended to be comprehensive. The review covers LD, HD, and NR applications fueled by natural gas, gasoline, and diesel fuel.

REGULATIONS

Regulatory activity in 2014 that will affect vehicular emissions technologies were a mix of proposals that were developed in previous years and some finalization and development efforts. This section will touch upon the following key regulatory initiatives from 2014:

- Non-Road Mobile Machinery (NRMM) Stage V proposal. This will harmonize the EU with Switzerland, and implements a particle number (PN) regulation.
- Light-Duty Initiatives: Real-Driving Emissions (RDE), WLTP (World-Harmonized Light-Duty Test Procedure) for Euro 6c (2017), and finalization of US Tier 3.
- Indian fuels and emissions roadmap: Bharat IV nationwide by April 2017.
- China air quality initiatives.
- Other California HD NOx program.

Non-Road Mobile Machinery - Stage V

The proposed Stage V regulation introduces a limit on the number of particles (PN) of 1×10^{12} /kW-hr complementing a particulate mass limit of 15 mg/kW-hr for land-based engines between 19 and 560 kW. This aligns with the Swiss standard which was the first one ever to introduce a PN limit in the NRMM sector. There is however no PN limit for engines >560 kW in the main category but a PN standard applies to all inland waterway engines above 300 kW.

The NOx limit for the main engine category remains unchanged compared with Stage IV at 400 mg/kW-hr but engines >560 kW have now to meet 3.5 g/kW-hr. Emissions durability periods vary by engine category. In the land-based engine category, the durability requirement is 8000 hours for engines >37 kW. In the main category, Stage V requirements will become effective on 1 January 2018 for new engines designs and 1 January 2019 for all engines. Engines in the power band 56-130 kW which benefit from a 1-year delay. Inland waterway engines between 300 and 1000 kW implement one-year later while those >1000 kW and railway engines have a 2-year delay (i.e. 1/1/2020 for TA of engines - 1/1/2021 for placing on the market of engines). The Commission will have to report by the end of 2020 on the potential for further measures, and by the end of 2025 on the monitoring of in-service conformity testing results, and on the use of exemption clauses. Further definition of implementation details and requirements will be adopted by the end of 2016.

Light-Duty Initiatives

Real-Driving Emissions (RDE)

Numerous reports have shown that in-use emissions from cars can be much higher than would be indicated by certification testing. For example more than half of the 14 Euro 6 diesel cars tested with SCR (selective catalytic reduction), LNT (lean NOx trap), or EGR (exhaust gas recirculation) systems had NOx emissions >6X higher than certified (2). Two cars, each with LNT or SCR systems, came in at ~25X higher than certified. In another study (3), of three such vehicles tested, the best (urea-SCR) was 3-4X higher, and the highest (EGR; and LNT+urea-SCR) were 5-7X the certified level in PEMS (portable emissions measurement system) testing. Even US Tier 3 light-duty diesel can show high in-use emissions (4), wherein two cars with either an LNT or SCR emitted 4-20X the Bin 5 allowable NOx, depending on route. Most SCR emissions were in the range of 10X. However, a third SCR vehicle had in-use emissions similar to the certification, demonstrating the feasibility of doing such. The investigators think engine calibrations, not additional hardware, can solve the problem of excessive NOx emissions.

Similarly, GDI (gasoline direct injection) PN (particle number) emissions are a concern. In one study, a Euro 6 gasoline direct injection (GDI) car had about 2X higher PN emissions on the autobahn versus on the NEDC [5]. In another study [6] two Euro 6 GDI cars had nearly 10X higher PN emissions at 130 kph (km/hr) versus the NEDC. Numerous studies have also shown most of the GDI PN comes from cold start. Khalek, et al., (7) showed 1.5 to 2.5 orders of magnitude higher hot-start solid PN emissions (10-15 second duration) from both GDI and MPI gasoline engines relative to LD diesels with DPFs. PN emissions from cars were measured coming out of a parking area on a hot day (35°C). The engines were stopped, and PN emissions were measured upon start-up. Not only were the emissions high (1000X ambient) but they were small, with PFI solid particles averaging only 6-7 nm. More work is needed, but as stop-start systems propagate, this could become an exposure issue at major intersections.

The European Commission agreed to use PEMS in regulating RDE for both diesel NOx and GDI PN. As part of their certification package, auto companies are soon required to report for monitoring purposes in-use emissions measured by two techniques - the power binning approach and the moving average window approach.

Vlachos, et al., described these two approaches (8). In general, in the power bin method data on pollutant and CO_2 emissions as well as vehicle speed and power at the wheels are calculated over intervals of 3 seconds, which are categorized into power bins. The averages of pollutant emissions for each power interval are weighed by the relative power frequency distribution from the WLTP database. Finally the weighed distance-specific and CO_2 specific emissions over all power bins are summed up to calculate a single emissions value, expressed in g/km.

In the moving average window method, second-by-second emissions data (g/km) are averaged over moving averaging windows (MAWs), the duration of which is determined from a certification cycle (i.e., CO_2 on the WLTP). Averaging windows are categorized according to their average speed into bins that represent urban (v<45 km/h), rural ($45 \le v < 85$ km/h) and motorway (v>85 km/h). The resulting distance specific emission averages can be calculated for each speed bin. Data screening is determined by the CO_2 emission in each speed bin relative to that obtain on the WLTP. Figure 1 shows some representative data using this approach. If data fall outside of the tolerance zones, which are to be determined but are key to the level of stringency of the regulation, they are eliminated.



Figure 1. Representation of the Moving Average Window (MAW) approach to determining applicability of PEMS data from in-use emissions. Solid lines represent test data. Dashed lines represent two levels of tolerance or acceptability of the data. (<u>8</u>)

An RDE regulatory proposal was recently signed by European Commission Vice President Frans Timmermans for approval by member state commissioners (9). The proposal content is uncertain at this time, but it likely calls for GDI PN and diesel NOx not-to-exceed (NTE) RDE limits by September 2017 (new engine types) and September 2018 (all engines). Further tightening is likely proposed for a few years later. The details of the test procedures and limit values will come by mid-2015.

Introduction of WLTP in Euro 6

Development of the EU-WLTP has been on-going for years. The intent is to use it to harmonize test procedures around the world using one basic test cycle that can be split into four segments (types of driving) that can be weighted to meet regional needs. There is still much discussion on the role out of the test. Indications are that it will be introduced by 1 September 2017 for new engines and September 2019 for all engines (<u>10</u>, <u>11</u>). Correlation of CO_2 emissions relative to those on the NEDC (New European Drive Cycle), and the appropriate regulatory values is in process with the objective of adoption by mid-2015. No change in Euro 6 limits for criteria pollutants are expected.

US Tier 3

The US EPA (Environmental Protection Agency) finalized the Light-Duty Tier 3 regulations in April 2014 (<u>12</u>). Although it is closely harmonized with the California LEVIII regulation, which begins phasing in this year, there are differences as shown in <u>Table 1</u> (<u>13</u>).

Table 1. Although LEVIII and US Tier 3 were developed together, there are differences. (<u>13</u>)

Similarities	Differences		
NMOG+NOx fleet average identical for 2017-2025	California Zero Emission Vehicle (ZEV) Program		
14011104 101 2017 2020	LEV III starts in MY 2015, Tier 3 starts in MY 2017		
Emission categories/bins			
essentially identical*	Full useful life standards:		
Certification fuel specifications very similar (E10) and have reciprocity	 LEV III - 150k miles; Tier 3 – 120k miles w/ optional 150k 		
	 LEV III FTP limits also apply at 50°F 		
Evaporative emission standards essentially identical*	(Tier 3 applies only after 4K miles)		
	LEV III 3 mg/mi PM standard phase-in ends in 2021; Tier 3 ends in 2022		
Similar US06 PM standard			
pnase-in: 10 mg/mi reducing to 6 mg/mi (ARB has anti- backsliding provision)	LEV III 1 mg/mi PM standard starting in MY 2025 (Tier 3 remains at 3 mg/mi from MY 2017+)		
*Some minor differences may exist relative to early phase- in/first years of implementation	LEV III fleet average based on CA+S177 state sales (Tier 3 fleet average based on 50-state sales)		
	LEV III credit life capped at 5 years (8 years for Tier 3: 2017-2022)		

Both regulations are nominally a 70-80% tightening of NMOG (non-methane organic gases) and NOx from Tier 2 or LEVII levels, and these are roughly 40% tighter than Euro 6. A key difference between LEVIII and Tier 3 are that in 2025 the LEVIII PM (particle mass) standard tightens to 1 mg/mile (subject to review this year) while Tier 3 remains at 3 mg/mile. Given today's level of GDI engine technology, the 1 mg/mile standard would likely require a GPF (gasoline particulate filter).

India

To address some of the most polluted air in the world, the Indian government assembled a group of experts to assess vehicle-based solutions. After a couple years of gathering data and discussing approaches, a report of findings and recommendations was published (<u>14</u>):

- Three of four major cities investigated exceed India's own PM10 (particles <10µm) ambient standards by 2-4X.
- Vehicles account for nominally 20-50% PM2.5, and 10-95% of NO₂.
- Adopt nationwide BS IV (essentially Euro IV standards) diesel and gasoline fuel (50 ppm sulfur) and emissions standards by April 2017. Adopt BS V fuel (10 ppm) and emission standards by April 2019 (new engines)-April 2020 (all). Adopt BS VI four years after BS V (1 April 2024)

China

China is by far the largest market for cars and trucks. If the rate of growth of auto sales continues at 9% per year, annual sales in 2020 could reach 40 million cars, roughly on par with that of the US, EU, and Japan combined. However, this large and continuing influx of cars will put further stress on the notorious air quality in major Chinese cities.

Although air quality is a major problem, 2014 saw very little regulatory activity in China. In late-2013, China implemented the Clean Air Action Plan, which set out city and regional air quality goals, among other top-level initiatives. Although the China IV HD regulations (similar to Euro IV) were officially in force in July 2013, very few China IV trucks have been sold, as it was preferred to buy new China III trucks (and China II trucks in some regions). In support of the MEP (Ministry of Environmental Protection), the MIIT (Ministry of Industry and Information Technology) ruled that new China III trucks are no longer allowed to be sold as of January 1, 2015. Low sulfur fuel (50 ppm) is available nationwide, and 10 ppm fuel is available in major cities. Compliance rates are still uncertain.

Also in 2013, Beijing also put forth an Air Pollution Action Plan with aggressive reductions in major pollutants. They started implementing China V requiring wall-flow DPFs (diesel particulate filters) on all new HD vehicles registered in the city. Beijing is aiming to implement China VI standards in 2016.

Perhaps the most far-reaching development of 2014 is the Special Policy Study by China Council for International Cooperation on Environment and Development (CCICED; ref. <u>15</u>). It is a farreaching document that recommends fundamental changes in how China regulates air quality. For example, recommendations are to move to a more regional approach on air quality, like setting goals and managing air quality on a regional basis. They would legally bind regions to achieve air quality standards, and group regions by air basin characteristics rather than political boundaries. Goals and assessments would be developed based on technology and scientific principles. The national government would strive to coordinate regional goals and move China into a modern infrastructure.

These recommendations, as well as much of China's air quality initiatives are on hold until a new minister of the MEP is named in the March Party Congress.

Also pertinent, China set new LD fuel consumption standards, tightening 17 to 36% depending on vehicle weight from 2015 standards (Phase III) with an average reduction of 27%, from 6.9 l/ km to 5.0 l/km on the NEDC. This is about 120 g CO_2 /km compared to 95 g/km in the EU for 2020-21. Further tightening of 15-30% is estimated for 2025 (<u>16</u>).

Other

Other on-going regulatory initiatives that will affect the vehicle emissions directions include the California HD low NOx program, and the US EPA's Phase 2 HD greenhouse (GHG) rule.

California HD Low NOx Program

Much of California is in a severe ozone non-attainment region, with parts of South Coast and San Joachim Valleys at 85 or even 95 ppb annual ozone levels compared to a 75 ppb national requirement. CARB (California Air Resources Board) showed that NOx rather than volatile organic compound (VOC) reductions are key in these regions, and 75% reductions in the inventory are not enough (17). About a third of the NOx inventory comes from HD vehicles, so CARB is sponsoring a technology demonstration program for completion in mid- to late-2016 aimed at reducing HD NOx by 90%, to 0.020 g/bhp-hr NOx (0.026 g/kW-hr) on the combined cold and hot certification cycles. CARB showed NOx emissions at 8-15 mph (miles per hour; 13-24 km/hr) were up to 7X the 2010 certification level on some trucks (18), so it is looking at expanding the NTE (not to exceed) zone on the operating map to lower loads. Further, about a quarter of the NOx inventory comes from the non-road sector, so CARB is evaluating these for further NOx reductions. As many trucks operating in California are from out-of-state, CARB is also calling on the US EPA to set a tighter national standard. In that regard, in December 2014 the EPA proposed tightening the ozone National Ambient Air Quality Standards (NAAQS) from 75 ppb

annual average to 65-70 ppb (<u>19</u>). Depending on how low the final standard is set, further national HD NOx reductions may be implemented to help states reach attainment.

HD Greenhouse Gases

In March 2015 the US EPA will be proposing the second phase of HD GHG reductions beginning in ~2021. The first phase started in 2014 and will be complete in 2017, with 6-23% CO₂ reductions from the overall vehicle and 6-9% reductions coming from the engine alone, relative to a 2010 baseline. In Phase 2, the EPA will propose regulating trailers in addition to more reductions from the engines and/or total vehicle.

The European Commission is anticipated to submit a HD CO_2 proposal soon, for implementation in the 2018-20 timeframe. It will be vehicle based using simulations to take into account configurations (20).

ENGINE TECHNOLOGIES

Driven mainly by greenhouse gas regulations in major markets throughout the world, both LD and HD engine technologies are making impressive gains in the market. But the challenges are not letting up. Looking to 2020 and beyond, LD CO_2 reductions of 16 to 30% from 2014 levels are indicated by the regulations, and further HD reductions up to 10% or more are possible.

This section will summarize the leading LD gasoline and LD and HD diesel engine technologies that will help engine and vehicle manufacturers meet these goals.

Light Duty

Gasoline direct injection (GDI) engines were introduced into the market in the 1990's, but it wasn't until 2008-09 that they started to grow in market share. By 2020 they could comprise 50-60% (<u>21</u>) of the gasoline engines, as they provide excellent performance and save about 10-15% on fuel consumption versus the multi-port injection counterparts. Most of the GDIs being built today are in the intermediate specific power ranges (60-80 kW/liter) and designed for efficiency (<u>20</u>), however some are primarily designed for performance. This is not enough to meet the emerging greenhouse gas regulations, and many LD engine technologies are being evaluated to build upon this. <u>Table 2</u> provides an overview of many of these technologies.

Another dimension to the scenario is cost. Shown in Figure 2, Kirwan (22) provided a perspective on costs for some of the technologies in Table 2. The solid lines were added to illustrate the incremental costs in \$ per % CO₂ reduction for three incremental steps.

Some of the technologies in <u>Table 2</u> will be highlighted in the next sections.

Table 2. General overview of various engine technologies showing CO2 reductions relative to the GDI engine, some key emissions issues, and rough estimate of the development status.

Engine Technology	CO ₂ Reduction	Emissions Issues	Status
GDI base, turbo, stoich; baseline	0	PN	Implemented
Stratified GDI	5-8%	PN	Implementing
Cylinder de-activation	5-8%	-	Implemented
Spark assist CI gasoline	5-10%	-	Research
Homogeneous Lean SI	5-10%	Lean NOx	Development
Reaction Controlled Compression Ignition	15%	LT HC+CO	Research
HEV (additive to others)	7-15%	-	Implemented
Downsize GDI, 18 →24 bar BMEP high CR, Miller stoic	10-15%	PN	Implementing
d- and c-EGR	10-15%	Cold start, controls	Implementing
Lean-burn GDI	10-20%	Lean NOx, PN	Implementing
Light-duty diesel	15-20%	Lean NOx	Implemented
Gasoline direct-injection compression ignition	15-25%	Lean NOx, LT HC	Advanced Engineering
Diesel 2-stroke opposed piston	25-35%	Lean NOx	Development

Gasoline Engine Technology

Homogeneous lean spark ignition engines can provide both low engine-out NOx, and lower pumping and thermal losses. Kondo, et al. (23) demonstrated a nominal 40% BTE (brake thermal efficiency) on a single-cylinder engine at the load point of 1500 RPM and 5 bar BMEP (brake mean effective pressure). The engine has a compression ratio of 13.2 and runs at an air/fuel=30. NOx emissions are 50 ppm, but still requiring lean NOx aftertreatment. The investigators contend the engine is well-suited for mass production.

Emerging RDE and more realistic test cycle development for CO₂ reductions will push more emphasis on higher power levels. Fraidl (<u>21</u>) showed that by adding the Miller cycle (extended expansion stroke), cooled EGR (driven by a small supercharger), and higher compression ratio to a 1.6 liter turbo-charged GDI engine, high-load fuel consumption (2000 RPM 14 bar BMEP) is cut by 15%, down to ~200 g/kW-hr. Even at 4 bar BMEP the fuel savings is 10%. The low-fuel consumption "sweet spot" on the engine map is quite large, and the region of minimum fuel

consumption is lower than 220 g/kW-hr. A 1250 kg demonstration vehicle was built using the engine technology along with a cooled exhaust manifold, reduced friction, and enhanced warm-up strategy, It achieved 3.8 l/km fuel consumption on the NEDC and the 2020 EU CO₂ values without hybridization.



Figure 2. CO_2 reductions and cost for a variety of LD engine technologies, relative to a 4-cylinder dual overhead cam multi-port injection engine with dual independent cam phasing. (22)

A dedicated-EGR engine, wherein one cylinder's exhaust is fully fed into the intake manifold for constant 25% EGR for a four cylinder engine, was demonstrated on a vehicle (24). Besides the configuration change, other additions include pistons for CR=11.7, an additional port fuel injector to allow separate fuel control on the dedicated cylinder, and a low temperature coolant loop for the additional supercharger and aftercooler. The fuel consumption was cut 10% relative to the base GDI vehicle, and torque and peak power increased 25%, and was higher for the 2.0 liter engine than for a comparable 2.4 liter baseline GDI engine. Because the engine runs at stoichiometry, a three-way catalyst achieved emissions just shy of LEVIII. The boosting system was not well matched, more sophisticated controls are needed for a production version, this base engine is not as dilution-tolerant as others, so further optimization is possible.

An interesting detail on cooled EGR was presented by Tsuchida (25). The knock suppression of cooled EGR is improved about 4 crank angle degrees if the EGR is taken after the catalyst, with much-reduced NOx levels. The explanation is related to the oxidation effect of NOx.

The gasoline-based engine technology with the lowest potential CO_2 emission is the GDCI (gasoline direct-injection compression ignition) engine, mainly due to dilution and use of the diesel cycle. Sellnau (<u>26</u>) reported the first results on a multi-cylinder engine, and has one installed on a vehicle, although it is too early to have results on it. In an interesting comparison to eight advanced gasoline and diesel engines in the same size and power class, fuel consumption of the GDCI engine at the eight load points was compared to the "composite" engine with the best fuel consumption at any load point. Averaged over all points, the GDCI had 2% lower fuel consumption than the composite engine. The best fuel consumption of the GDCI engine was only 2% higher than the best diesel engine fuel consumption. Cost is expected to be competitive. In a schematic

depiction of <u>Figure 2</u>, the GDCI had CO_2 reductions much above the line at the intermediate cost levels. NOx emissions will require aftertreatment, but PM and PN are quite low.

LD Diesel EngineTechnology

Although gasoline engines are closing the CO_2 gap relative to today's diesel engines, diesel engines are also improving. Gerhardt (<u>27</u>) showed that relative to the best in class diesels of today, a further 5% CO_2 reduction can be realized in a compact vehicle with engine and turbocharger friction reduction, increased injection pressure (2200 bar to 2500 bar) and by either downsizing from 1.6 to 1.2 liters (3-cylinder) or decreasing peak cylinder pressure. Another 2% reduction can be obtained with advanced common rail injection with hydraulic flow control. With other vehicle improvements, 80 g CO_2 / km is thought achievable without hybridization.

In a new project, an exhaust-driven 48 volt turbine can complement a stop-start and drive electric auxiliaries so an advanced diesel engine might drop a compact car fuel CO_2 emission to ~70 g/km (<u>28</u>). The engine architecture is shown in Figure 3. The system features a 48V electrical architecture, a 12.5 kW belt-integrated starter generator, and an advanced lead-acid battery. Project completion is scheduled for early-2016.

The 2-stroke opposed piston diesel engine is now being investigated for light-duty applications (29). Compared to a traditional 4-cylinder diesel engine with similar power and torque, the concept has six pistons in three cylinders and a displacement of 2.25 vs. 2.8 liters. Estimated from single-cylinder engine tests, the fuel consumption is 20 and 25% lower on the LA-4 and US highway fuel economy cycles. The sweet spot on the engine map extents much into the low-load regime (210 g/ kW-hr fuel consumption at 1500 RPM and 3 bar BMEP). Idling exhaust enthalpy is high and post turbine temperatures are about 340°C for fast catalyst light-off, and NOx emissions are on the order of half those of a traditional US diesel. Most interestingly, vibrational moments are about two orders of magnitude less than those for a V6 engine.



Figure 3. Architecture of a 1.5 liter diesel engine that shows promise to drop a compact-car CO2 emissions to 70 g/km without hybridization. (27)

HD Engines

HD engine technology has been through a remarkable ten years. Since Euro III regulations (ended 2005 in Europe), total engine costs have more than doubled (<u>30</u>), but BSFC (brake-specific fuel consumption; g/kW-hr) dropped 10% (<u>31</u>), and NOx and PM emissions dropped by 90-94%.

It is interesting to compare a variety of modern engines within the same market and application. MackAldener (<u>30</u>) reported on a competitive breakdown analysis of seven Euro VI engines in the 400 HP (310 kW) class. All estimated engine costs, reported on a relative basis and only including advanced components - air handling, fuel injection, EGR, etc. - are within 25% of the average cost. The cost of the EGR engines is about 15% higher than those of the three non- or low-EGR engines. Average engine-out NOx emissions of low- or no-EGR engines is 9.5 g/kW-hr vs. 4.3 g/kW-hr for EGR engines, but the exhaust aftertreatment costs were similar despite that the cycle-average NOx removal efficiency ranged from 90 to 95% for the two groups. The aftertreatment costs were all within 25% of the average, and comprised ~45% of the total engine cost.

Despite significant recent gains, HD engine technology is still rapidly advancing. The US Department of Energy is funding the multi-year \$280 million SuperTruck program to demonstrate technologies that decrease freight-specific fuel consumption by 33% on a truck using a realistic road drive cycle, relative to a 2009 truck. The engine goals are to demonstrate an engine with >50% BTE under steady-state road-load conditions, and to do a scoping study on achieving 55% BTE. Figure 4 shows the status and general approaches of the four engine program leaders as of mid-2013 (<u>32</u>). All are starting from a 42% BTE baseline, and rely heavily on combustion improvements and waste heat recovery (WHR).

Cummins, being the first participant to start the program, updated their final steps to attain 51% BTE (<u>33</u>). Closed-cycle gains comprised piston bowl and fuel injector optimization, increased compression ratio, and calibration. Open-cycle gains were EGR loop and turbocharger efficiency gains, valve and port optimization, and downspeeding. Combined, they deliver 3.5% BTE gains. Friction and parasitic reductions account for 1.5% BTE gain, and include fluid pump improvements, lower viscosity oil, downspeeding, and shaft seal, piston, and crankshaft friction reduction. For achieving 55% BTE, more improvements in bowl design (0.5% BTE) and injectors (1.3%), reduced heat transfer (0.9%) parasitic load reduction (0.5%), and optimized WHR turbine (0.6%) are mentioned on the conventional diesel combustion path. A dual-fuel low-temperature combustion (LTC) approach is also being investigated. More details on the two approaches to 55% BTE were offered by Koeberlein (<u>34</u>).

Singh (<u>35</u>) described Daimler's measures to attain 50% BTE. They downsize the engine from 14.8 to 10.7 liters, reduce EGR usage, reduce aftertreatment back pressure, and put much emphasis on model-based control of the engine. Unlike Cummins, who puts WHR turbine energy to the crankshaft, Daimler uses it to generate electricity, but is looking at a lower 2.3% BTE improvement. Similar to Cummins, Daimler increases compression ratio, optimizes the piston bowl and injectors, uses low viscosity oil and drops friction

and parasitics using similar methods. They are just starting to scope out technologies for 55% BTE, but also are looking at further traditional engine improvements and a dual fuel LTC approach.



Figure 4. Plan and status (mid-2013) of four companies on meeting the 50% BTE goal of the DOE SuperTruck program. (32)

Volvo (<u>36</u>), like Daimler, is downsizing the engine while maintaining power, but going from 13 down to 11 liters. They are adding 2-2.5% BTE from better combustion, 3% from better air handling, and 2% from WHR. They are spending much effort on advanced combustion simulation techniques (like Probability Density Function) to explore 55% BTE approaches. Partial pre-mix combustion (PPC) is a leading approach. They are targeting an additional 2% BTE from combustion improvements, 1% from aftertreatment optimization, 2% from reduced pumping losses, and 1% from improved WHR. A two-fuel strategy, as with the others, is currently not being looked at.

As in light-duty, HD test cycle can have a big impact on efficiency results. Roberts (<u>37</u>) showed this and the relative contributions of engine and vehicle contributions, <u>Figure 5</u>. In particular, friction, downspeeding (also ref. <u>32</u>), and turbocharger effects are sensitive. While engine methods add to efficiency up to a few percentages per example (here, 0.3 to 4%), some illustrative vehicle-based measures, which are much more sensitive to drive cycle, contribute 4-14%.



Figure 5. Some representative engine and vehicle improvements and how drive cycle affects efficiencies. (37)

Roberts set goals to use advanced low-temperature combustion (LTC) methods to achieve 52-53% BTE (47% achieved today), while traditional combustion methods might achieve 54% (achieved 49%).

Many of the approaches with the two combustion methods are similar, but LTC has higher pumping loss due to high EGR rates, while traditional combustion has higher heat losses. LTC combustion can benefit from new piston and fuel system design to deliver faster dilute combustion. Conventional combustion can benefit from reduced piston crown heat loss (also ref 34).

Finally, first results on a multi-cylinder medium duty 2-stroke opposed piston engine were reported (<u>38</u>). Importantly, results extrapolated from single-cylinder work were substantiated. The 3-cylinder 4.9 liter engine delivers 275 HP (212 kW) and 1100 Nm torque. Fuel consumption was measured before much optimization at an average of 202 g/kW-hr in 12-mode steady state testing. The peak BTE is 48% BTE, but the sweet spot (45% BTE) extends to all but the lower third of the engine map. All emissions appear manageable using traditional methods: 3.2 g/kW-hr NOx, 0.06 g/kW-hr PM averaged over the 12-mode test. Most interestingly, the engine enables better extrapolation into HD results. With no WHR or other advanced technologies, 51.5% peak BTE (160-170 g/kW-hr fuel consumption) is thought achievable with production-ready technologies.

Natural Gas Engines

Spark-ignition (SI) stoichiometric natural gas engines have tailpipe CO_2 emissions that are about 20% lower than diesel engines, and 25% lower than MPI gasoline engines. As the HD GHG emissions tighten this could be a key driver for CNG (compressed natural gas) SI engines. However now, natural gas trucks are of interest due to the reduced fuel price, about \$1.30/gallon lower in the US (<u>39</u>). Mainly due to the high cost of CNG tanks, only applications that consume a lot of fuel are of interest, and because of a limited refueling infrastructure (1% of public natural gas filling stations vs. diesel in the US), applications are largely limited to day fleets. Transit buses, regional trucks, and refuse haulers are the majority of CNG applications.

This is large enough of a market to drive some engine technologies. Stanton (<u>40</u>) showed that, when considering methane tailpipe emissions, the latest SI CNG engine has similar GHG emissions ($CO_2 + CH_4$) to near-term diesel engines (2017 US GHG limits), but 13% less than those for gasoline engines. However, by implementing reductions unique to natural gas engines, GHG emissions can be reduced perhaps 17-20%. Key improvements include closing the crankcase ventilation (5%), combustion improvements (6%), turbo charger optimization (3%), and methane oxidation catalyst improvements (2%).

LEAN NOX CONTROL

Lean NOx emissions control has been used for a little more than a decade, when lean NOx traps (LNT) were first applied in limited LD diesel applications, and also went mainstream in HD applications with SCR (selective catalytic reduction) in early introduction to Euro IV standards. Lean NOx control offers both NOx reductions to meet tailpipe regulations, but also is key to fuel consumption savings by allowing for more-efficient engine calibrations which can result in higher engine-out NOx. HD deNOx (SCR) was chosen in Euro IV

applications for this reason, but EGR was the preferred route in the US to meet an even tighter NOx regulation mainly due to lack of a urea infrastructure.

Rapid improvements are still occurring in both SCR and LNT applications. This section will cover catalyst and system-level developments.

SCR

SCR development is the most active area right now in all of exhaust emissions control, and the work is providing significant benefits. From the first commercial applications about a decade ago, SCR specific emissions (percent NOx slip normalized to catalyst size) is down 90%, reflecting improvements in catalysts, system design, and control.

There are three general types of SCR catalysts that are in use today - vanadia, Fe-zeolite, and Cu-zeolite. Ummel and Price (<u>41</u>) described the key attributes of each, particularly pertaining to sulfur contamination. In Non-Road Transient Cycle (NRTC) testing with system-appropriate DOC precious metal loadings, vanadia catalysts achieved 92% efficiency, compared to 95% and 94% for the Fe- and Cu-zeolites respectively. However, upon sulfur exposure (20 ppm SO₂ until 1-3 g/liter is accumulated), the vanadia catalyst system deteriorated by 5% to 87% NOx efficiency, but the zeolite systems were more affected, dropping 9% to 86 and 85% respectively. Both the vanadia and Fe-zeolite systems were significantly desulfated by operating over an NRTC peaking at 300°C, and fully recovered after a ramped 8-mode cycle peaking at 450°C. The Cu-zeolite system required 600°C for 15 minutes to fully recover.

Vanadia SCR catalysts are preferred in markets with high-sulfur fuel. However, under continuous low temperature operation (T<400°C), significant catalyst deterioration can occur. Xi, et al., (<u>42</u>) quantified the formation of ammonia sulfate species on vanadia SCR catalysts. They and others (review in 43) found that if SCR catalysts are exposed to cold start conditions without going up to ammonia sulfate decomposition temperatures (~350-400°C) the ammonia sulfates can build up and block catalyst activity.

A key issue today with vanadia catalysts is vanadia stability. Liu, et al., (<u>44</u>) did a study of vanadia and tungsten release from a commercial SCR catalyst using a bench and engine-based method. They found vanadia emissions on the engine method were always higher, as much as 3X higher at 500°C, but only 1.5X higher at 700°C, Figure 6. The engine-based approach clearly provides a more complete measurement of metal emissions, especially at lower temperatures where the dominant release mechanism may not be the thermal/chemical route. This is most likely because the engine-based approach can measure particle-phase and particle-bound vapor-phase metal emissions which are adsorbed onto particulate matter. The trends measured on the bench reactor are similar and the results within the same order of magnitude, so the authors contend the lab

method is satisfactory for qualitative assessment, but the cost is much lower. They found new catalysts are much more durable, with no emissions at 500°C and two orders of magnitude lower emissions at 600°C.



Figure 6. Release of vanadia from a commercial honeycomb catalyst measured in a lab reactor and on an engine. Engine results are higher likely due to particulate effects. (44)

In that regard, Spengler, et al. (<u>45</u>) showed that vanadia SCR catalyst durability after 100 hours exposure at 650°C can be improved significantly. By stabilizing the titania support, and then immobilizing the vanadia catalyst on the titania, they increased the NOx efficiency for a relatively stable catalyst from 30% at 300°C catalyst to 95%.

SCR catalysts perform better if there is stored ammonia. However, ammonia slip from SCR catalysts during cold to hot transitions is a critical control issue. Work by Kamasamudram, et al., (<u>46</u>) may help reduce this problem. Strong acid sites and good NH₃ oxidation activity, which is related to the nature of the copper species, will help much in controlling transient NH₃ slip. Upon heating, ammonia is transferred from weak acid sites to strong acid sites. If the oxidation function overlaps with this adsorption dynamic, the ammonia will be oxidized in situ rather than be slipped.

Certainly substrate design and geometry can impact SCR performance. Strots, et al. (<u>47</u>) found 600-csi (cell per square inch) SCR substrates improve SCR efficiency by about 10% at 300°C. However, the differences due to wall thickness or cell shape (square or hexagonal) were only $\pm 2\%$ at 60,000.hr space velocity, and half of that at 100,000/hr.

As engines become more efficient, exhaust temperatures will drop and shift overall SCR performance to be more dependent on reaction-rate control and enhanced by high-catalyst loadings. New substrate designs with higher porosity can enable these higher catalyst loadings without significant back pressure penalty. Pless, et al., showed (<u>48</u>) NOx conversion at 220°C improved from 74 to 85% for highly loaded SCR catalysts on high-porosity substrate versus standard catalyst loading on conventional substrate at equivalent csi, <u>Figure 7</u>. In addition, NOx conversion is improved by increasing cell density from 400 to 750 csi. Overall, a volume reduction of 40-50% is achieved by coating SCR on high cell density, high porosity substrate, while maintaining similar NOx reduction efficiency, but with a 20-30% increase in high load back pressure.





HD SCR systems need to function well for a million kilometers, requiring durable systems. Partridge, et al. (<u>49</u>) is investigating the aging of Cu zeolites. The NOx conversion efficiency and NH₃ storage characteristics of degreened (4 hours at 700°C) versus aged catalysts (50 hours at 800°C) don't change much, as most of the deterioration occurs in first 25% of the catalyst. However, the aging results in increased NH₃ and NO oxidation (<u>50</u>). Partridge, et al. distinguished between simple NH3 oxidation and parasitic NH₃ oxidation (in the presence of NOx), in which the latter was not impacted much at 400°C. This shows a different reaction pathway for NH₃ oxidation with or without NOx present. However, at 450°C and higher temperatures, Yezerets (<u>50</u>) showed loss of NOx conversion efficiency with aging due to HT NH₃ oxidation.

Chemical poisoning can also affect SCR catalyst performance. Shwan, et al., (<u>51</u>) concluded Fe-BEA zeolites deteriorate from phosphorous poisoning due to metaphosphates replacing hydroxyl groups on the active isolated iron species. Deactivation by potassium is due to ion exchange and loss of Fe active sites due to cluster formation (<u>52</u>).

Strots, et al., (47) evaluated different SCR systems. They reported the best NOx reductions on the weighted WHTC (cold and hot start World Harmonized HD Test Cycle) are achieved with a catalyst that has improved LT efficiency, followed by dosing with ammonium nitrate for NO₂ generation to help LT reactions (53) and putting SCR catalyst on a DPF (SCR filters). It didn't help much being able to dose ammonia at 140°C versus 180°C (baseline), probably due to the lack of catalyst activity at this temperature and the temperature distribution on the WHTC. SCR filters were also demonstrated to have good LT activity. In other work, SCR filters showed no deterioration in NOx performance over 4000 hours of high-load testing in the higher RPM ranges (54).

SCR system control is becoming more important as system deNOx efficiencies increase. This usually involves NH_3 storage and overdosing, and control of ammonia slip especially in transient conditions. Model-based control is the leading emerging approach for doing this. Iivonen and Wabnig (55) describe a system controller that incorporates observer and controller models. The result is reduced

calibration time, reduced emissions at any given ammonia slip level, use of off-line calibration, and reduced hardware costs. Chavannavar (56) provided some insight to their approach wherein NOx conversion and NH₃ slip over a given transient cycle can be tuned by the "Slip Factor", which is used in SCR Offset Controller.

Finally, a comprehensive review book was published covering all important aspects of SCR catalysts and systems (<u>57</u>).

Lean NOx Trap and Related Systems

The lean NOx trap is the deNOx method of choice for smaller light-duty diesels in Europe to meet Euro 6 requirements. It is used as a stand-alone system, delivering nominally 70-80% NOx removal, or it can be combined with a passive SCR, in which the NH_3 is generated in situ in the LNT during rich cycles, or can complement a urea-SCR by enhancing LT deNOx performance.

Umeno, et al., (<u>58</u>) described an improved LNT with higher sulfur tolerance. The main NOx adsorbing material, barium oxide (baria) is supported on one basic material, and strontium oxide, which acts as a scavenger for the sulfur to protect the baria, is coated in the whole catalyst with high dispersion. In bench testing the NOx removal efficiency of the new catalyst is 2X that of a standard baria LNT at a 3 g/liter sulfur loading. The sulfur is also released at a higher rate and at lower temperatures.

Harle, et al., (59) showed LNT improvements by modifying the ceria component with basicity adjustment. The ceria shows high lean NOx removal efficiency (~60-70%) at 120°C for NO compared to baria (45%). This is important because little NO₂ is formed at that temperature. The NOx is released in lean conditions first at 220°C and more at 335-350°C depending on formulation. This feature makes the LNT an attractive candidate for use ahead of an SCR catalyst to enable a wider range of NOx control. Theis (60) further characterized the performance of LNTs that release NOx at lower temperatures. Storing NOx as a nitrate is preferred, as it releases the NOx at a somewhat higher temperature, and is more sulfur tolerant. Theis also shows NOx is released at higher temperature in the presence of hydrocarbon (C_2H_4). Walker (<u>61</u>) showed how advancements in the understanding of these materials can translate into improved system performance. Second generation adsorber material holds more than 2X the NOx at 130°C versus the first generation, and releases it at 60C° higher temperature (240°C). A downstream SCR (on DPF) captures most of the released NOx.

LNTs can be managed in different ways to get better high-load efficiencies. Earlier, Yasui, et al., (<u>62</u>) reduced high-load NOx flux to an SCR catalyst by running stoichiometric up-transients, resulting in the DOC reducing most of the NOx. They now extended this work to LNTs (<u>63</u>). They run either rich and/or stoichiometric during the transients depending on conditions, and use the three-way catalyst functionality of the LNT to reduce NOx during these periods. System cycle-averaged efficiency increases from 65% to 80%, and the fuel penalty drops from 5% to 4.5%. Basaiji, et al. (<u>64</u>) improved the performance of the DiAir system wherein hydrocarbons are dosed

into the LNT at a variable lean level on a 1-2.5 Hz frequency (<u>65</u>). Catalyst improvements increase efficiency 2.3X, while better hydrocarbon control increases the utility of the hydrocarbon.

LNTs are replacing DOCs and are being used with SCR to improve system performance. Grubert, et al. (<u>66</u>) added SCR catalyst to the DPF (SCR filter) in an LNT system and improved NOx removal efficiency by 8-20%, depending on test cycle. The results are similar to those reported by Holderbaum (<u>67</u>), who shows a 20% NOx reduction efficiency improvement on the Artemis cycle by adding an SCR filter in an LNT system. Krutzsch (<u>68</u>) showed 15-20% improvements for an earlier and commercialized system using a separate flow-through SCR catalyst. When a DOC is replaced with an LNT in a urea-SCR system, the NOx emission is cut more than 50% on the NEDC due to better system low-temperature performance (<u>69</u>). Conversely, keeping the deNOx efficiency the same, adding an LNT to a urea SCR system increases the fuel penalty by 0.4 to 0.6%, but drops urea consumption by 40-50% (<u>67</u>).

Interestingly, Euro 6 OBD (on-board diagnostics) for these LNT+SCR systems can be straightforward as each deNOx component can remove enough NOx to keep the tailpipe level below the OBD threshold (<u>70</u>). The threshold is crossed only if both components malfunction.

PARTICULATE CONTROL

Particulates from internal combustion engines are perhaps the most toxic component of their emissions. Further, the ultrafine fraction (less than 100-120 nm in diameter) are beginning to emerge as the most toxic portion of the particulate. Mayer (71) analyzed some epidemiology studies in which cardiovascular mortality was correlated to both particulate mass (PM; weighted towards the larger particles) and particulate number (PN; weighted to the ultrafine particles), and converted the PN concentrations to PM allowing a side-by-side comparison on the health impacts. In this comparison, the toxicity of the PN fraction is 8X that of the whole PM2.5 (PM less than 2.5 µm) population. In other words, upwards of 85-90% of the toxicity of the whole PM2.5 is attributable to the ultrafine subfraction ("PM0.12"). Horn (72) showed that PN levels on highways can be more than 50X background levels, and the variation from weekdays to weekends can be 5X, indicating high exposure to PN from vehicles.

Stein (20) showed that PM and PN can be unrelated in diesel applications, wherein PM-based solutions will not necessarily drop PN emissions. Figure 8 shows the PN and PM levels for different technologies. Solutions that drop PM using non-DPF solutions have little correlation to PN reductions - a 5X range of PM levels has no impact on PN, albeit the range of PN is an order of magnitude. DPF solutions have much lower PM levels (50% of the best non-DPF solution) but also much lower PN emissions - one to three orders of magnitude lower. The wide range of PN is due to the state of loading of the filter, with high loadings (soot or ash) correlating with low PN.



Figure 8. Relationship between PN and PM for various diesel emissions systems. Dropping PM without DPFs (top set of points) will not necessarily drop PN. (20)

This section will cover filters used for PN reductions, both for diesel and gasoline applications.

Diesel Particulate Filters (DPF)

Diesel particulate filters (DPF) were first commercially utilized in significant numbers in the light-duty sector in 1999, and in the heavy-duty truck sector in 2005. They have been through several generations of improvement, and there are only smaller incremental improvements now. A noteworthy exception is the combination of SCR catalyst applied to a DPF.

When SCR systems were added to heavy-duty trucks in the US in 2010, the engines were recalibrated for perhaps 2.5-3.5X higher NOx than for EGR+DPF systems. This change also dropped engine-out PM. The increased NOx/soot ratios enabled almost all of the PM burn on the DPF to be through NO2 oxidation, or the commonly-called passive regeneration route. Li, et al., (73) studied the oxidation of soot by NO₂ in the filter. They observed that soot within the porosity oxidized first, quickly dropping back pressure. In the next regime, soot was mainly burned at the soot/wall interface. The source of this NO₂ is the oxidation of NO on the catalyst residing in the filter wall near the soot interface, and the back-diffusion of this NO₂ to the soot. On average, the NO is recycled five times in the beginning of the soot burn and three times after an hour at 400°C. The recycling and soot burn rates decrease due to the increasing gap between the soot and catalyst sites. Aging of filter can lead to highly variable NO₂ production rates, but pre-calcining of the washcoat at 700-800°C stabilizes the NO₂ production. The Pt:Pd ratio also impacts NO₂ generation stability.

Ash accumulation and behavior in a DPF is of significant importance, as it governs the life of the filter. Sappok, et al. (74) reported on their latest work in this regard. The amount of soot accumulated in the filter prior to regeneration plays a key role in influencing the extent of ash migration from the channel walls to the back of the filter. A thicker soot cakes both has less adhesion due to proportionately more gaps between the soot and the filter; and they also have higher drag forces from the gas acting on the soot. Large (500 μ m to 800 μ m) portions of the soot cake, containing ash particles, detach from the filter surface and move down the filter channels. Contrary to other reports, quantitative and conceptual models indicate the amount of

soot accumulated may be more important than whether there is active or passive regeneration. In cases where little soot accumulates in the filter prior to regeneration it can be expected that more ash will accumulate on the channel walls relative to cases where a thicker soot cake is built-up prior to regeneration.

Back pressure sensors are used to help monitor the state of soot loading of the DPF. Kim, et al., (75) reported some anomalous behavior in this regard. After long periods of soot build-up followed by cooling (like overnight), there is a sudden Δp increase across the filter upon driving again, but not if there is an idling period. Upon cooling, water condenses in the soot, and if there is a gap between the soot and the filter due to in situ NO₂ regeneration (above), rapid heating can cause the wet soot to expand and break off, increasing the back pressure of the filter. Idling after soak allows the water to slowly leave, reducing the extent of the problem.

When SCR catalyst is incorporated into the DPF, the passive regeneration behavior is compromised due to the loss of NO₂. Hohl (<u>76</u>) looked at various non-road transient test cycles and quantified the temperature and NOx:soot ratios needed for passive regeneration. Engine management and catalyst formulations can shift from the non-passive to the passive regeneration regimes. For example, increasing temperature from 225 to 325°C and the NOx:soot ratio from 200 to 250 respectively, enables passive regeneration; or increasing NOx:soot ratio from 100 to 150 at ~310° with changes in the DOC enables passive regeneration.

Finally, PM sensors are being developed for both DPF regeneration control and for OBD purposes. Sappok (77) is developing a radio frequency (RF) sensor, the signal of which correlates well to soot and ash loading on the filter. This might result in more optimized regeneration strategies.

Samaras and Geivanidis, et al., (<u>78</u>) reported on consortium work on the feasibility of three types of PM sensors for OBD purposes. Monitoring to the EU OBD threshold limit for heavy-duty vehicles of 25 mg/kWh is feasible with the existing sensors. There were indications that durability was still an issue to be solved, since all sensors showed at least one failure during testing, but durability improved since the testing. A small scale durability testing with newer samples of the sensors revealed no failures. An important factor for sensor integration, which defines sensor performance, is the OBD algorithm. Depending on the OBD algorithm, a detection model may need a travel distance for a valid diagnosis that is higher than the OBD type-approval procedure.

Gasoline Particulates and Filters

Driven by gasoline PN regulations in Europe, gasoline particulate filters (GPF) are emerging as a viable solution. Activity on them is also increasing in China. Contrary to diesel applications, in which engine-based solutions cannot easily meet PN regulations, GPFs are but one approach, and use of them will depend on cost, fuel consumption considerations, and green marketing. Storey, et al. (79) investigated the composition of hydrocarbons associated with the PM from a GDI engine for various fuels. They operated the engine rich ($\lambda \sim 0.9$), as generally the PM is generated during rich operations. A new method for soot HC speciation was developed that uses a direct, thermal desorption/pyrolysis inlet for the gas chromatograph (GC). Results show high levels of polycyclic aromatic hydrocarbons (PAH) in the PM, including downstream of the catalyst, and the aldehydes were dominated by the alcohol blending. In follow-up private communications (80), it was observed the gasoline (E0) PM has a much wider range of hydrocarbon species than both diesel and gasoline-alcohol blends. Fuel injection design and the air-fuel ratio also influence the amount and diversity of hydrocarbon species that are present. It was postulated that the hydrocarbons are associated with the soot before entering the three-way catalyst. The results are important because of implications to health effects.

Lee, et al. (<u>81</u>) also investigated the nature of GDI soot. The three-way catalyst (TWC) was found to be an important component that reduces PM emissions (volatile organics and soot). Because GDI soot contains a higher proportion of ash than diesel soot, the ash plays a larger role in the oxidation reactivity of GDI soot than for diesel soot. Crystalline structures of GDI soot are slightly less ordered than those of diesel soot, except for the idling condition, and do not change significantly with engine operating conditions. Soot chemistry (hydrocarbons, weakly bonded carbon, ash) is a major component for the enhanced oxidation of GDI soot. Figure 9 shows the relationships. In consideration of the effects of those chemical components, a kinetic model of GDI soot oxidation has been developed, resulting in a good agreement with experimental data.



Figure 9. GDI soot oxidation rate increases with conversion or time, and shows various regimes of oxidation. WBC: Weakly bonded carbon. SOF: Soluble organic fraction. (<u>81</u>)

Morgan, et al., (82) showed that TWC formulations can also enhance soot burn on a GPF, dropping burn temperatures 100-200C° relative to uncoated filters (675°C). Fuel cut-offs on decelerations were shown to result in significant burning of soot due to both high temperatures and the presence of more oxygen. Investigators are striving to consolidate all of the TWC onto the GPF, creating a "four-way catalyst". Kern, et al. (<u>83</u>) report that a closecoupled all-in-one GPF has higher emissions than a traditional TWC with an uncoated GPF, even though PGM loadings are the same. Others have shown that this may be due to poisoning affects, wherein a traditional TWC may be more tolerant of such.

Black carbon is emerging as a potent greenhouse gas, even though it is short-lived, as it might be 2000X more potent than CO_2 . The UN International Panel on Climate Change reported black carbon is the second highest contributor to anthropogenic climate change (<u>84</u>). Chan, et al., (<u>85</u>) found that GDI vehicles (model years 2011 and 2012) have black carbon emissions on the order of 1.8 mg/km as measured on the US06 and Phase 2 and 3 of the US FTP-75 test cycles. GPFs reduce these emissions by 80% or the equivalent of 2.9 g CO_2 (eq)/km. This is about 2.4% of the average allowable CO_2 emissions in the US in 2020. Using vehicle and engine measures for these CO_2 reductions in that timeframe might cost \$200 (see the Figure 2 discussion). Cold start and cold ambient conditions result in significantly higher black carbon emissions.

OXIDATION CATALYSTS

Diesel oxidation catalysts (DOC) serve several functions: oxidizing hydrocarbons and CO, supplying an oxidation exotherm to enable active regeneration of the DPF, and oxidation of NO to NO_2 for passive regeneration of the DPF. They are the oldest of diesel emission control technologies, but are still improving. Methane oxidation from natural gas engines is also a key function of these oxidation catalysts.

Ahari, et al., (<u>86</u>) looked at DOC aging phenomenon. Lean and hot environments create unfavorable oxidation states on the precious metal clusters, decreasing their effectiveness. Aggressive HC dosing during DPF regeneration floods the catalyst with an elevated level of reductant which in turn reduces the precious metal oxidation states to enable a higher level of activity. If periodic rich excursions are not incorporated into the DOC application, the DOC cannot recover to full functionality.

Ummel and Price (<u>41</u>) reported on new DOC formulations for a highly passive-DPF/copper zeolite SCR or DOC+Cu zeolite (non-DPF) system that balances sulfur tolerance, easy desulfation, and high thermal durability. NO₂ generation decreases with sulfur exposure for all Pt-containing catalysts. High Pt formulations are less sensitive, but some Pd is needed for active regeneration or Cu zeolite desulfation durability. The 6:1 Pt:Pd formulation was found to be the best compromise of sulfur behavior and durability for these applications.

Satoshi (<u>87</u>) developed DOCs for Euro IV applications in developing countries that have good performance and sulfur tolerance; or reduced precious metal loadings for Euro V at 10 ppm fuel sulfur levels. With 50 ppm sulfur, all formulations that were evaluated oxidize CO and hydrocarbons similarly, but a Pt:Pd=4:1 formulation performs better at 350 ppm sulfur. The catalyst loading of the base Pt:Pd=2:1 for Euro IV (50 ppm sulfur) can be cut in half and still maintain acceptable performance for Euro V applications at 10 ppm sulfur.

Methane is quite stable, so it oxidizes at temperatures 50 to 100C° hotter than C2 and C3 aliphatic hydrocarbons. Improvements in light-off can be realized with catalyst formulation improvement, but deactivation of methane catalysts is a major issue. Kim, et al. (88) showed the deactivation of Pd-based catalysts is due to strongly adsorbed oxygen species from water and reaction intermediates (HCHO). They use metal oxide modifiers to improve durability, and show that going richer (λ ~1.2) at 400-450°C can partially recover performance.

Finally, new low-temperature combustion engines show promise for fuel consumption savings, but their exhaust temperatures are low and hydrocarbon and CO emissions can be high. Parks, et al., (<u>89</u>), show CO poisoning is a key issue with precious metal oxidation catalysts. Hydrocarbon light-off can increase 50 to 70C° due to this effect. A co-precipitated Cu-Co-Ce oxide shows good CO oxidation and is more tolerant to propylene inhibition. A Pd-ZrO₂ based catalyst shows good performance, with CO light-off (T50, 50% conversion temperature) at about 180°C and HC light-off at 200°C.

GASOLINE CATALYSTS

The new LEVIII and US EPA Tier 3 light-duty regulations are resulting in more innovation in three-way catalyst developments, but Ball and Moser (<u>90</u>) showed that current vehicles certified to PZEV (partial zero emission vehicle) standard essentially meet the new SULEV20 level (super ultra-low emission vehicle 20 mg/mile NMHC+NOx). However, for other vehicles an average of 15% more Pd and 33% more Rh by 2025 will be needed.

Cold start emissions reduction is a key issue to address further. Chang, et al., (<u>91</u>) improved the low temperature performance of three-way catalysts by developing a new $Al_2O_3/CeO_2/ZrO_2$ mixed oxide catalyst washcoat. Compared to conventional CeO_2/ZrO_2 mixed oxides with similar compositions, the new material exhibits higher oxygen storage capacity, especially at low temperatures. The improved thermal stability of the new material further stabilizes and improves the precious metal dispersion on the support, giving reduced light-off temperature. For more cold start HC emission reductions, HC storage components were improved to increase the HC trapping capacity and HC release temperature.

Catalyst durability requirements are increasing, so understanding the aging and testing for it are important. Fathali (92) showed that for fresh and 40-hour aged samples, fuel-cut after acceleration has the highest contribution towards deactivation of the catalyst system. Also, the retardation fuel-cut is detrimental to the catalyst system but not to the same extent as an acceleration fuel-cut. During the aging procedure, exotherms were observed at the start of fuel-cut and the intensity of these exotherms increase with the length of aging time. The increasing exotherms are explained by the decomposition of HC into C and H₂, and their subsequent oxidation at lean conditions. Also, fuel-cut-off temperature measurements demonstrate that the magnitude of those exotherms is related to the total number as opposed to the total length of the fuel-cut.

Regarding TWC precious metal management and interactions, Goto, et al., (<u>93</u>) found that at Pd:Rh~1 is the optimum ratio. The performance of the catalyst is related to the amount of free Rh⁰, which depends on the Pd/Rh ratio. If the ratio is higher the Pd and Rh alloy.

Lean burn gasoline shows much promise for significant fuel consumption reductions, but emissions are an issue. Parks, et al., (94) updated their project lean-burn gasoline NOx work using TWCs, LNTs and SCR by moving it from the bench to an engine. Bench performance of 99%+ NOx removal was duplicated. Operating over a TWC at λ =0.96 delivers a good balance between NH₃ generation and fuel consumption over a wide range of conditions. Adding NOx storage material to the TWC increases lean time and decreases rich time. There is a delay in NH₃ production, however. Transient test modeling shows a 10% fuel consumption improvement using lean modes over the base GDI engine. Stoichiometric operation provides ~25% more NOx than the lean modes, which helps to reduce the rich/lean time ratio.

SUMMARY/CONCLUSIONS

This paper provides a high-level overview of the key regulatory and technical developments on engine efficiency and emissions from 2014. Following is a summary.

Regulations

The main regulatory developments of 2014 include an EU proposal on Non-Road Mobile Machinery (Stage V) for 2019-20 that introduces a PN (particle number) standard. EU light-duty initiatives on real-driving emissions are moving forward, with two methods of data evaluation being required for monitoring purposes as part of the certificate of compliance. Full implementation could be 2017-18 with a focus on LD diesel NOx and gasoline direct-injection PN. India's roadmap on fuel quality and emissions is proposing starting nationwide Bharat IV (Euro IV) in April 2017, followed by Bharat V in 2020, and Bharat VI in 2024. California is investigating 80-90% HD and NR NOx reductions for 2020, and the US EPA in March 2015 will be proposing the next round of HD greenhouse gas (GHG) standards for ~2021.

Engine Technologies

Some promising LD engine technologies are highlighted that can deliver up to 35% CO₂ reductions relative to a turbocharged GDI baseline. Homogeneous lean, spark-ignition engines show potential to be commercialized, and deliver 5-10% CO₂ reductions. Further downsizing the GDI engine and using the Miller Cycle (increased expansion stroke) can drop fuel consumption by 10-15%, as can using more cooled-EGR. Gasoline compression ignition is moving into multi-cylinder testing and may get 15-25% reductions. LD diesel technologies, including better control, and moving to a 2-stroke opposed piston design are delivering 15-35% reductions.

In the HD sector, technologies are being demonstrated that can drop fuel consumption by another 10% from the best engines of today. Much of these improvements are coming from combustion improvements, pumping loss reductions, waste heat recovery and friction reduction. Pathways to another 10% reduction (to 55% brake thermal efficiency, BTE) are being explored.

NOx Control

NOx control is clearly focused on selective catalytic reduction (SCR). Catalysts are being characterized further for sulfur degradation and tolerance. Low-temperature performance is enhanced with high catalyst loadings, enabled by high-porosity substrates. High temperature performance is improved using higher cell density catalysts. More information has been reported on reducing ammonia slip from SCR catalysts, and on understanding and improving durability.

Lean NOx traps (LNT) are also improving regarding sulfur tolerance and passive release of the NOx using only temperature increases. LNTs are being combined with SCR in the LD sector to reduce urea consumption and improve system low-temperature performance.

Particulate Control

The regeneration of diesel particulate filters using NO₂ was further quantified and the implications are summarized. The soot oxidation mainly occurs between the soot layer and the filter material, significantly decreasing the adhesion of the soot, causing it to flake off under some conditions. This impacts back pressure and ash distribution. The NO₂ regeneration is also further quantified for filters with SCR catalyst.

Gasoline particles are further described, and can contain a wide assortment of hydrocarbon species. Ash in the particles can aid in the soot oxidation, as well as three-way catalyst (TWC) incorporated into the filter.

Gaseous Pollutant Control

Diesel oxidation catalysts (DOC) are being optimized for use with higher sulfur fuels. It was shown that periodic rich excursions are needed to improve both DOC and methane catalyst performance by reducing oxidation poisoning.

TWCs are being designed with better low-temperature oxygen storage capacity. More is learned about the effect of fuel cut-offs on catalyst deterioration. Lean burn gasoline NOx reduction systems using LNTs and passive SCR are showing 99%+ NOx reductions and estimated fuel consumption reductions of ~10% after accounting for rich excursions to generate NH₃.

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