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Diesel Emission Control in Review

Timothy V. Johnson
Corning Incorporated

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ABSTRACT

The paper summarizes the key developments in diesel emission control, generally for 2005. Regulatory targets for the next 10 years and projected advancements in engine technology are used to estimate future emission control needs. Recent NOx control developments on selective catalytic reduction (SCR), lean NOx traps (LNT) and lean NOx catalysts (LNC) are then summarized. Likewise, the paper covers important recent developments on diesel particulate filters (DPFs), summarizing regeneration strategies, new filter and catalyst materials, ash management, and PM measurement. Recent developments in diesel oxidation catalysts are also briefly summarized. Finally, the paper discusses examples of how it is all pulled together to meet the tightest future regulations.

INTRODUCTION

Controlling diesel engine emissions is one of the most important aspects of modern air quality management. The field is very active with upwards of perhaps 1000 papers and presentations annually on the health effects of diesel exhaust, new fuels, engine technologies, and emission control technologies. This paper will offer a review of a narrow aspect of this field, diesel exhaust emission control.

As in the past (1), the review is not intended to be all-encompassing. Rather, the objective is to summarize representative studies that show the key trends in the industry. An emphasis is placed on reports from 2005. First, the regulatory issues are addressed, followed by a quick overview of engine technologies as a means of estimating the exhaust emission control requirements. Then the author will review NOx, PM (particulate matter), and hydrocarbon control developments, and close with some examples of integrated systems.

REGULATIONS, ENGINES, AND GENERAL EMISSION CONTROL REQUIREMENTS

It is important to put emission control technologies in the perspective of regulations, which are the primary driver for advancements, and what engine technologies can reasonably deliver. The difference between the two represents the challenge to the aftertreatment technologies. Following is an attempt summarize these needs.

REGULATIONS

Before summarizing the tailpipe regulations, it is worthy to note some recent regulatory and related developments that will have future impacts.

In the US, the EPA finalized their proposal in December 2005 to tighten the National Ambient Air Quality Standards for PM (1). The proposal, which has to be met across the US by 2015, or regions could lose highway funding, calls for tightening of the PM2.5 (PM smaller than 2.5 µm) 24-hour ambient air levels to 35 µg/m^3 from 65 µg/m^3. The PM2.5 annual average standard would remain unchanged at 15 µg/m^3. Further, a new standard for particles sized between 2.5 and 10 µm would be established at 70 µg/m^3, the intent of which is to better capture anthropogenic urban emissions.

Similarly, the European Commission finalized their Clean Air for Europe (CAFE) thematic strategy in September 2005 (2). The strategy will return about €42-130 billion (0.3-1% GDP) to society in health benefits in 2020 at a cost of €7-8 billion. On PM2.5, the daily limit is 25 µg/m^3, to be phased-in from 2010-15.

The tightening of these ambient air quality standards in the US and Europe are important because significant regions are now in nonattainment to current standards. These new levels could result in further tightening of tailpipe standards and increases of diesel retrofits

Moving into current tailpipe regulations, for the purpose here, diesel tailpipe emission regulations are categorized by light-duty, on-road heavy duty, and non-road heavy duty applications.

Light-Duty Diesel

Figure 1 shows general relationships between the future light-duty diesel (LDD) regulations for the US, Europe, and Japan (no adjustment for test cycle differences). The US regulations were finalized in 1999, and the Japanese 2009 regulations were finalized in March 2005. Although the European regulations have been in discussion for a few years, the European Commission just submitted their proposal to the Parliament in December 2005. (For reference, today's Euro IV standards are at 0.025 g/km PM and 0.25 g/km NOx. Japan 2005 standards are 0.013 g/km PM and 0.14 g/km NOx)

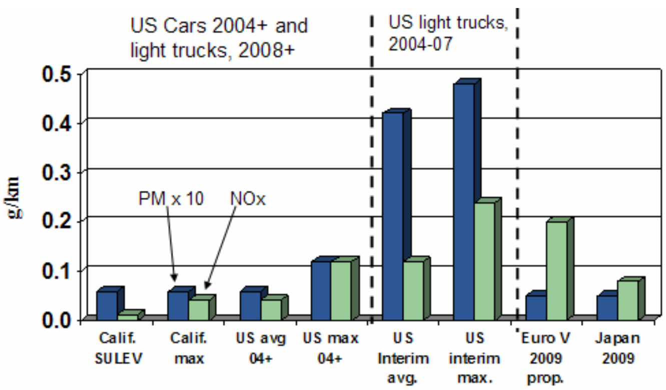

Figure 1. General comparison of light-duty diesel standards in the US, Europe, and Japan. Left bar is PM (X 10).

The US standards are based on fleet average NOx levels, while California uses a fleet average non-methane hydrocarbon (NMHC) basis. The others are maximums. Note that the maximum allowed NOx level in California is equal to the US fleet average requirement ("Bin 5"), meaning that to sell cars in all 50 states, the California maximum level (0.07 g/mi or 0.042 g/km NOx) can't be exceeded. Also noteworthy, this level of NOx is nominally 80% lower than the Euro V proposed standard. The US will require unique engine and aftertreatment technology from Europe, or even Japan.

In addition to these regulations, California finalized their CO_2 regulation, dropping greenhouse-gas CO_2 equivalent emissions 30% by 2016. This results in a similar fleet CO_2 emission as from the European commitment of 140 g/km CO_2, but four years later. Canada has adopted the California standards, and several states are considering following. Most significant, California and followers are combining the tightest tailpipe and greenhouse gas standards in the world. This presents a huge challenge to the industry, and is already sparking significant developments in powertrain technology, especially for gasoline vehicles.

Also of significance, the European Commission signaled in their proposal to Parliament the desire to add a particulate number based standard (numbers of particles per km), pending successful verification of the UN-ECE PMP (Particulate Measurement Program). The main intent is to better tie emissions standards to health impacts by regulating solid ultrafine particles.

Heavy-duty diesel

The on-road heavy-duty diesel (HDD) standards are shown in Figure 2, as are estimates of engine emissions performance.

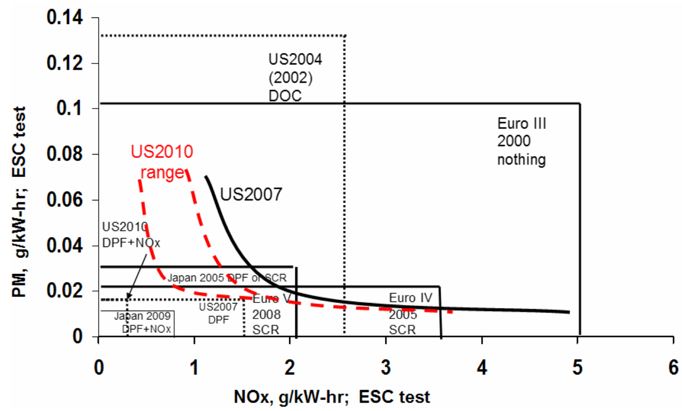

Figure 2. General comparison of on-road HDD standards in the US, Japan, and Europe. Estimated engine-out emissions for 2007 and 2010 (range) are shown. Steady-state cycle.

The European Commission is anticipating a Euro VI proposal to the Parliament in 2006 (3). It is widely anticipated that Euro VI will bring the emission levels to the US2010 and Japan 2009 ranges (<1.0 g/kW-hr NOx and <0.01 g/kW-hr PM).

Although meeting the steady-state and transient dynamometer test requirements are at the heart of regulatory compliance in Japan and Europe, and thus determines engine and aftertreatment technologies, in the US the Not-to-Exceed (NTE) part of the regulation represents the greatest challenge. In Europe and Japan, peak emissions (for example at full load) are unlimited as long as the test cycle average emissions are less than the limit values. With NTE, peak emissions are limited to 1.5X the limit values for any 30 second steady-state in-use operating point as measured with portable emission monitors, with some exceptions for operating point and ambient conditions. Thus, NTE demands high efficiency at high load, the most challenging condition.

Notable future trends relate to NTE. Europe and Japan are considering adapting the requirements, and if the US program is successful, the author speculates that only NTE compliance will be required in the future. Also in discussion (4) are worldwide harmonization of tests and regulations, an increased emphasis on fuel economy, and a look at number-based PM regulations, at least in Europe.

Non-Road

Figure 3 shows the US and European non-road diesel regulations (5), which are essentially harmonized. Japan is also moving in that direction. The regulations emerging in 2011-15 are about double the on-road standards regarding absolute levels, as well as four years behind. This is because the test cycles are more demanding, the applications so varied, and there are different fuel quality issues. As such, it is anticipated that similar technologies will be applied in this sector as in the on-road sector.

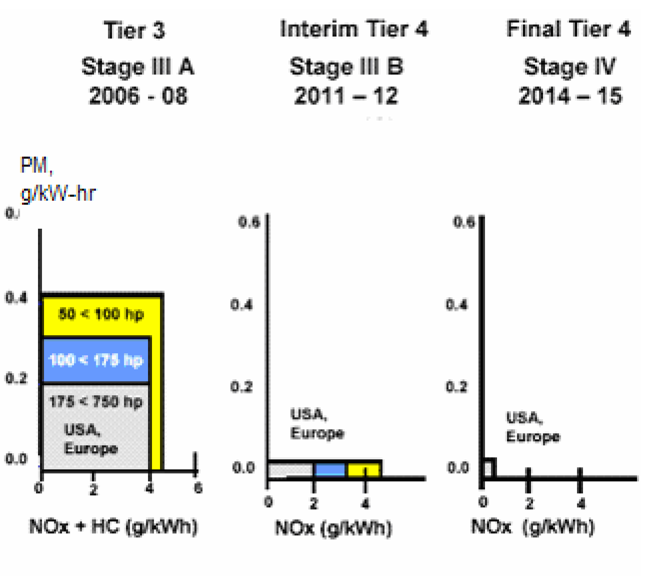

Figure 3. Future US and European non-road regulations for 50 to 750 hp engines (6).

ENGINE TECHNOLOGIES AND RESULTANT EMISSION CONTROL REQUIREMENTS

Traditional diffusion flame diesel combustion is benefiting from the continuous evolution of fuel injection equipment, air handling, exhaust gas recirculation (EGR), combustion chamber design, and sensors and control. The advancements on research engines are very impressive, even pointing towards hitting the US2010 NOx requirements without NOx aftertreatment (6, 7, 8, 9). Developing and transferring these technologies into production will impact emissions. Also, transient operation is much more difficult (10) than the steady state results shown here.

Another key trend is the advancement of cool-flame, pre-mixed "advanced combustion". There are several versions, with homogeneous charge compression ignition (HCCI) perhaps being the most common. All are typified by managing combustion via EGR, mixing, and fuel injection to keep flames out of PM and NOx formation regimes. Figure 4 illustrates the principle (11) for early and late injections at different EGR levels. Although NOx and PM are rather low, hydrocarbon and CO levels are high, often an order of magnitude higher than with traditional diesel combustion. Fuel penalties are less than 3%, and often less than 1%.

Advanced combustion is difficult to control at high load. As such, "mixed-mode" combustion is emerging, wherein advanced combustion is used at low load and traditional diesel combustion is used at higher loads. Depending on the transition point between the two modes, mixed-mode combustion can significantly reduce the need for low-temperature NOx control and for low-load regeneration of diesel particulate filters (DPF).

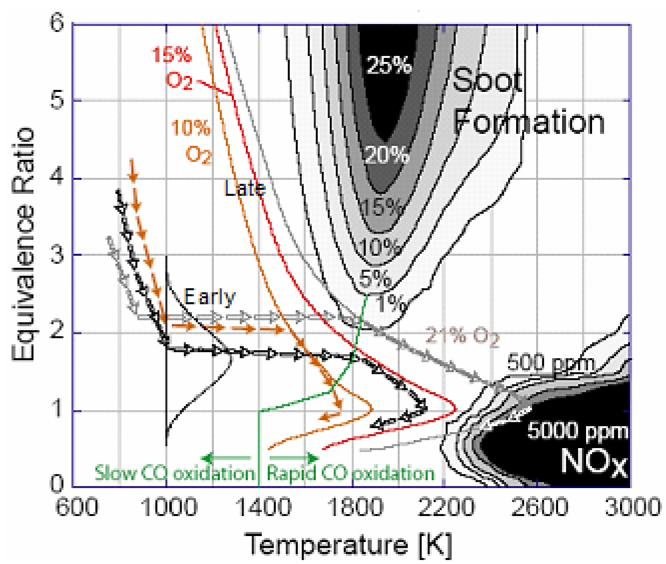

Figure 4. Principle of advanced combustion, wherein the flame temperature and equivalency ratio, , are kept away from NOx and PM formation regimes using injection timing and EGR (12).

Given that the filter technology has been commercial for five years, and NOx regulations are the most challenging to hit, engine status and aftertreatment requirements are focused on NOx.

Light-duty diesel engine status and emission control requirements

Reports on traditional diesel combustion show the potential to get down to about 0.09 to 0.14 g/km NOx on the European test cycle (12, 13). For example, a 1600 kg prototype vehicle is hitting Bin 8 (0.2 g/mi or 0.12 g/km) on the US test cycle (14). Using these engines, no NOx aftertreatment will be needed to hit the proposed Euro V requirements (even if the NOx limit is cut in half), and given reasonable advancements in technology, very little if any NOx aftertreatment will be needed to hit the Japan 2009 levels for all but the HD vehicles (>3500 kg). US Bin 5 requires nominally 70% NOx efficiency.

Given the very tight NOx requirements for LDD in the US, mixed-mode combustion developments are generally aimed at this market. The US test cycles mostly operate at less than 50% load, so most of the cycle can be run in advanced combustion mode with low NOx (9). As such, some are even hitting Bin 5 without NOx aftertreatment on research engines (14, 15). The first mixed-mode engines in the US for heavier applications will need modest NOx treatment, about 30-50% efficiency at the high load test points, but it would appear that 1600 kg vehicles can do without such by about 2009 (14).

Heavy-duty engine status

NOx emissions using traditional diesel combustion at steady-state high load operating points have been reported (16, 17, 18) in the range of 1.0 to 1.3 g/kW-hr (0.6-0.8 g/bhp-hr). Advanced combustion modes at <40 to 60% load are delivering (16, 17, 18, 19) NOx emissions in the range of 0.26 to 0.50 g/kW-hr (0.2-0.4 g/bhp-hr). A minimum threshold average emission value has been proposed by Parche (20) of around 0.6 g/kW-hr NO and 0.015-0.020 g/kW-hr PM. If the emissions performance of the research engines shown here can be transferred into production, it appears these threshold emissions will be attained.

Given that there is another year or more of engine development and engineering before freezing technologies for the Japan 2009 and US2010 regulations, it is reasonable that the above engine-out levels might be commercially realized. If so, to meet US NTE, about 50-70% NOx treatment will be needed at 500-520C. To hit Japan 2009 at 0.7 g/kW-hr NOx it appears only about 20-30% NOx control, if any, is required. However, as NOx systems can deliver fuel economy gains and are being implemented today (Euro IV and Japan 2005), HD engines in Japan 2009 will likely have such.

For non-road applications, it is difficult to predict emissions from these engines for the 2011-14 timeframe. However, it is reasonable that engine companies that make similar engines for both on-road and non-road applications will try to maximize synergies. As such, we could see Japan 2009 and US2010 engines show up in 2014 non-road applications and probably sooner to take advantages of early introduction incentives. In 2011, when generally only DPFs are expected too be needed, we will see NOx targets being met with advanced fuel injection, air handling systems, and some EGR, albeit lack of ram air limits the range of cooled EGR.

NOx CONTROL

Given the tight NOx emission regulations in the US and Japan, and the fuel economy impacts NOx aftertreatment can have, NOx control technologies will play a key role going into the future. As the state of NOx aftertreatment is more dynamic than that of PM or HC and CO control, the author elected to include it first.

Following is an assessment of the state-of-the-art on selective catalytic reduction (SCR), lean NOx traps (LNT), and lean NOx catalysts (LNC).

SELECTIVE CATALYTIC REDUCTION

Although NOx control was not required to meet the Euro IV or Japan 2005 HDD regulations (beginning October 2005), SCR was selected by several truck manufacturers because the high NOx efficiencies in the tailpipe allow the engine to be run at higher NOx levels for better efficiency and lower PM, thus delivering competitive fuel economy and eliminating the need for a DPF. In the US, the EPA generally laid out its requirements for SCR: no operation without urea, and conveniently located urea filling stations (21). The EPA is looking for a proposal from the industry on how to accomplish this. One reasonable approach to the first requirement might be to use interlocks that are engaged if the urea:fuel ratio is out of balance right after fueling.

Hirata, et al, (22) provides a comprehensive review of a modern SCR system as developed by Nissan Diesel for Japan. Because the Japanese test cycle strongly favors low-load operation, exhaust temperatures are rather low. They chose to use a DOC in front of the system to generate NO_2, which, with equimolar quantities of NO, reacts most efficiently with ammonia at low temperatures. In adding the DOC, they were able to increase the NOx removal efficiency at 200C from 40 to up to 70%. Further, converse to Europe, a zeolite SCR catalyst was chosen over the traditional vanadia-based catalyst because of better performance and smaller size. Figure 5 shows the efficiency curves for several tested catalysts. Simulated fuel economy gains from SCR over EGR/DPF systems were estimated to be net 4% at 90 km/hr cruise, after the 5% urea consumption (assumed 50% the cost of fuel) is taken into account. The authors also elaborated on the urea and delivery system, OBD (on-board diagnostics), and on the urea infrastructure.

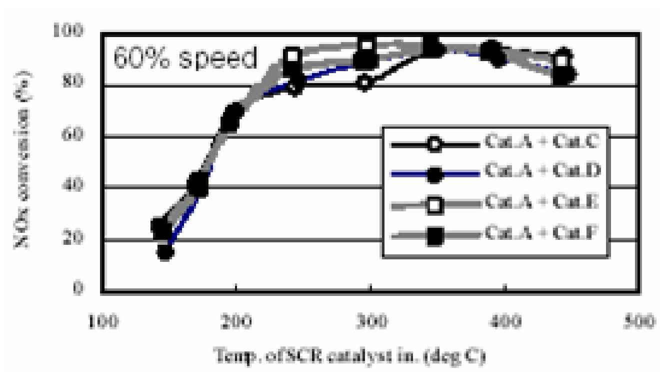

Figure 5. NOx removal efficiency curves for various SCR catalyst configurations (22). "Catalyst A" is an oxidation catalyst to promote the fast $NO_2/NO/NH_3$ reaction. "Catalyst C" is vanadia-based, and the remaining catalysts are zeolite (25% smaller than the vanadia system).

Regarding catalyst options, several reports show the advantages of zeolite for SCR applications (23, 24, 25, 26, 27, 28). For low temperature efficiency, Walker (27) shows better low temperature performance with zeolite than with vanadia, and more tolerance to non-ideal NO_2 levels. Excessive NO_2, that is NO_2>NO, is difficult to reduce using ammonia. Holcomb (28) reports more sensitivity to this effect at about 240C than at 310C. He also shows DOC length and catalyzed DPF design can be used to optimize NO_2 formation for SCR. More importantly, zeolite exhibits much better high-temperature durability. This is important when SCR systems are added directly after DPF systems, wherein DPF exit temperatures can approach 650C during regeneration. Although vanadia systems show signs of aging at temperatures less than 600C, zeolite shows impressive durability at 650C (26) or even 700C (25, 27).

SCR operation in very cold climates can cause problems, as the standard urea solution freezes at about -10C. Ammonium formate has the same ammonia content as urea but freezes at -20 to -30C, depending on formulation, and behaves similarly in SCR systems (29). An alternative is to heat the tank using engine cooling water and to use heating elements for the lines (22).

Some work is beginning to expand on earlier reports (30, 31) of possible secondary and trace emissions from SCR systems. Sluder, et al. (32), show that at 205C and with ideal NO_2/NO levels, steady state reduction of NO over zeolite catalysts doesn't begin until after 300 seconds, and for NO_2 not until after 500 seconds, independent of whether urea or ammonia is used. Reductant storage in the catalyst was hypothesized as the cause. N_2O, which is a very strong greenhouse gas, is the preferred reduction product if there is excess NO_2. Given this, it is not surprising that longer catalysts result in higher N_2O in these conditions. On the other hand, HCN emission, which occurs at similar levels (low ppm) regardless if ammonia or urea is used, was slightly higher at higher space velocities, Figure 6. Finally, small amounts of nitromethane, nitroethane, and nitropropane (<250 ppb) were detected upstream and downstream of the SCR catalyst. In the case of HCN and the nitro-HCs, the authors expect that a clean-up oxidation catalyst will virtually eliminate them.

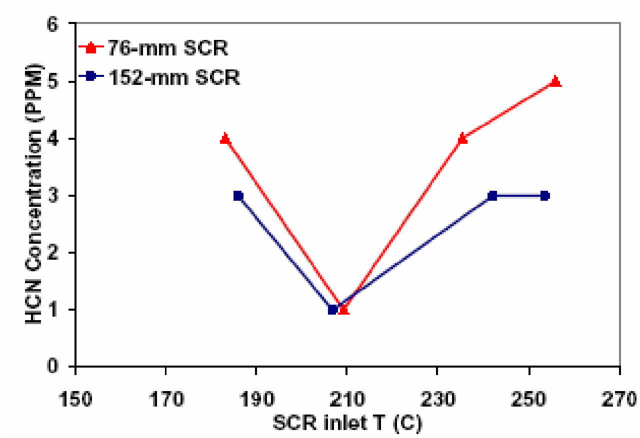

Figure 6. HCN formation from a reaction with NH_3, measured after an SCR system. HCN/NH_3 ratios are constant up to 6 ppm HCN (32).

Finally, on SCR and OBD, the European Commission finalized their requirements in September 2005 (3). It primarily calls for urea level detection and urea quality detection, combined with urea consumption correlation with engine NOx production. Alternatively, NOx sensors combined with level detection is preferred. If things are not right, torque limiting begins after the first stop.

LEAN NOx TRAPS

Lean NOx traps offer an attractive NOx solution for light-duty applications and those HD applications in which an extended urea infrastructure might be problematic. In US HD applications the challenge for LNT is acceptable high temperature efficiency to meet the NTE requirements (50-70% control at 500-520C), and do it over the useful life of the vehicle (185,000 miles for medium HDD and 435,000 miles for heavy HDD). On the light-duty side, efficiencies need to be high enough to hit Tier 2 Bin 5 (70% using traditional diesel combustion; 30-50% in the 50 to 70% load range using mixed mode combustion) over the useful life of the vehicle (120,000 miles).

HT LNT systems for lean burn gasoline systems have been developing for a number of years. Figure 7 shows the efficiency-temperature relationship for light-duty and heavy-duty applications reported by Roth (33). In light-duty applications (chassis certified) using traditional diesel combustion, low temperature NOx control is important.

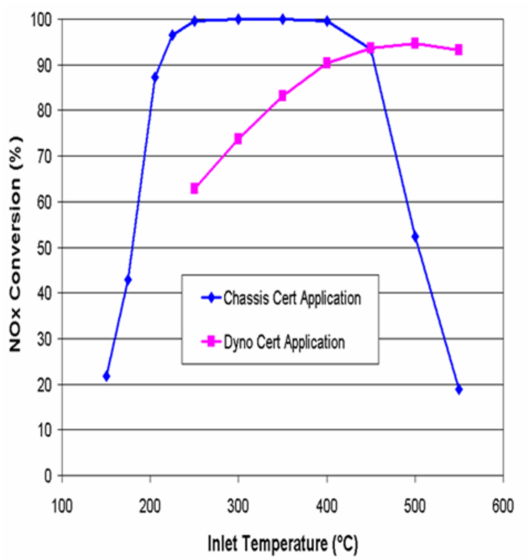

Figure 7. LNT efficiency for LDD (chassis certified) and HDD (dyno certified) applications (33). LT performance is critical for most LDD applications, whereas HT efficiency under high NOx fluxes is needed to meet HD NTE requirements. Hydrothermal aged 5 hours at 850C, 30,000/hr and 120,000/hr space velocities, respectively.

Hinz, et al. (34) also report on a HT LNT system for HD applications in which a bypass system is used during NOx regeneration. The system achieves at least 75% efficiency at all full load points, at a fuel penalty of only 2 to 3% at all but the low-load points. The swept volume ratio (SVR; ratio of LNT volume to the engine displacement) is 1.4.

Small amounts of hydrogen in the exhaust stream enhance the low temperature performance of LNTs. Fuel reformers are emerging to accomplish this task (35, 36). If a few percents hydrogen are present in the exhaust during rich regeneration, the LT efficiency markedly improves, even down to 150C. One can envision such a system for HD applications in which NTE is addressed by placing the LNT further back from the engine (35).

LNT durability problems surface when desulfating the LNT under rich and HT conditions. Reports of several years ago showed upwards of 50% loss of NOx storage capacity. Later results (37) are showing 10 to 20% capacity loss, with minimal decline after about 20 desulfations (generally 100,000 km). HT LNT formulations require HT desulfation, generally up to 700C, which would be expected to result in more deterioration. Wu (36) shows that temperature might be better controlled using exotherms in the LNT from combustion of reformer gas, thus minimizing deterioration.

As more is learned about sulfur loading and desulfation, deterioration issues are being addressed. In this regard, Takahashi, et al. (38) show that both CO and hydrogen in the regeneration gas are important, and most of the sulfur is present in the inlet regions. Sulfur at the exit is very difficult to remove.

Concerning cost, platinum group metal (PGM) loadings are coming down, saving on cost. In a very thorough and well-designed experimental plan, Theis, et al. (39) explain the positive and negative impacts and trade-offs of high PGM loadings in a gasoline application. Figure 8 is one example. At low temperature, higher PGM loading aids NO_2 formation, thus improving efficiency. However, at high temperatures, the PGM causes faster nitrate decomposition, thus dropping efficiency. This works in the direction of NTE and mixed mode requirements. In the case of mixed mode combustion, wherein little NOx is formed at LT, much reduced PGM loadings would appear possible.

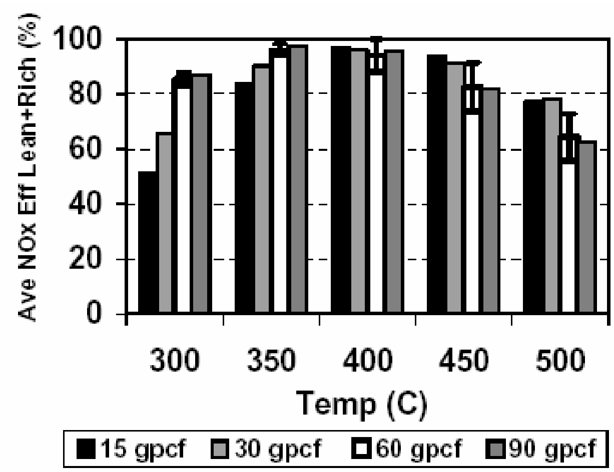

Figure 8. At lower temperatures, high PGM loadings are preferred, but at higher temperatures, lower loading are better (39).

Finally, Lambert (26) put an SCR catalyst behind the LNT to make use of ammonia that comes out of the LNT during rich periods to take up NOx regeneration spikes. Figure 9 shows the results. As the rate controlling step in LNTs is the reduction of NOx coming off the LNT during rich spikes under many conditions, it follows that the SCR combination can result in significant PGM savings.

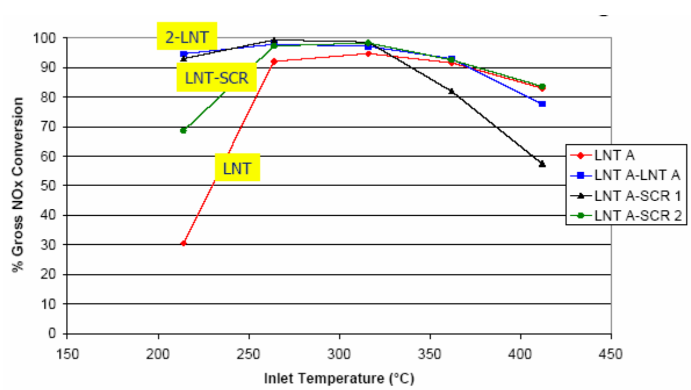

Figure 9. When an SCR is added to an LNT, system efficiencies go up (26).

LEAN NOx CATALYSTS

Lean NOx catalysts are attractive because they use fuel as a reductant, and potentially cheap catalyst (silver, zeolite, others). The problem is that the reductant has to be used continually, unlike with an LNT, and performance has been only in the 10-30% range with relatively high fuel penalties (6%).

Advancements are forthcoming. The KNOWNOX consortium in Europe reported (24) that small amounts of hydrogen (<2%) can markedly increase silver-based LNC reduction efficiency over much of the temperature range (200-400C). Further, a US Department of Energy consortium has developed a rapid LNC formulation screening process involving computational chemistry, synthesis, and rapid testing (40). More than 4500 compositions have been evaluated. Figure 10 shows an example of a new formulation that is getting close to SCR HT efficiencies.

One can envision combining the two discoveries: Hydrogen additions to improve LT performance, and the new LNC formulations for HT performance.

On-Board Diagnostics (OBD)

NOx OBD is centered on NOx sensors. A recent report (41) summarizes the performance and durability after 6000 hours of testing as part of the US DOE APBF (US Dept. of Energy Advanced Petroleum-Based Fuels) program on HD SCR. Figure 11 summarizes the results. At low NOx levels, as would be experienced after a NOx control system, accuracy and durability are impressive, but if placed ahead of the NOx control system, durability issues arose, albeit they appear predictable.

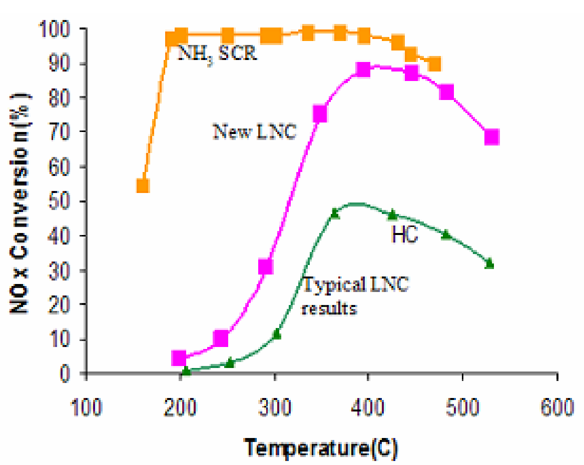

Figure 10. New LNC formulations discovered using a rapid screening process show improved HT performance compared to typical formulations (40).

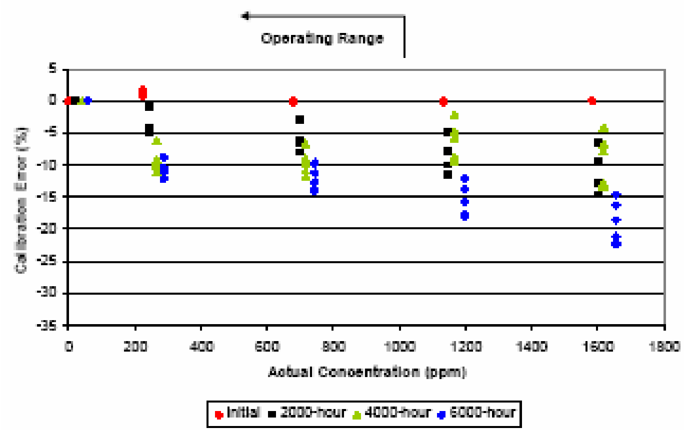

Figure 11. Sensor calibration error as a function of NOx concentration and exposure time to exhaust for one type of NOx sensor. Performance is consistent and durable at low NOx levels, as would be observed for an OBD sensor after the NOx control system (41).

NOx SUMMARY

For LDD, traditional diesel combustion will meet foreseeable European and Japanese needs without NOx aftertreatment. In the US, upwards of 70% NOx control is needed. Both SCR and LNTs apparently can meet this requirement. SCR has significant infrastructure issues (45,000+ diesel filling stations), and LNTs will require significant control. LNT PGM costs are coming down. Perhaps the combination of LNT+SCR can do it more economically and easily. However, as we move into mixed mode combustion (2009+), LNT costs will come down significantly simply because the NOx burden is lower, but more importantly, with reduced low-load NOx this challenge is lessened PGM savings can be realized.

For HD, the issues are different. Fuel economy and current approaches (and urea infrastructure) in Japan and Europe will keep SCR in play for the coming years. However in the US, although urea infrastructure is more challenging, NTE HT requirements place a huge demand on LNTs. SCR shows better HT efficiencies and is developing a track record. LNTs can hit the required HT efficiency, but desulfation issues with the HT formulations also raise durability questions not yet addressed in the literature. For the medium and light heavy duty vehicles, despite needing to hit NTE, fuel economy considerations are not as significant as in the heavy sector, and SCR infrastructure for these urban applications is more significant. LNT is a more plausible option here than for long haul applications.

If SCR is chosen for US2010 applications, we need to expect that in following years, hydrocarbon based NOx solutions like LNT and LNC will advance faster than SCR optimization. The rapid developments on LNC are especially interesting for less fuel-consumption sensitive applications. It is conceivable that HC-based deNOx could be more attractive than SCR in later years.

PM CONTROL

Diesel particulate filters have been commercially applied on passenger cars in Europe for more than 6 years, but are just beginning to go commercial for HD applications, with Japan 2005 and US2007 regulations coming into force. Despite that the field is generally in an optimization and cost reduction stage, developments and understanding is still advancing rapidly. Although the LD and HD applications have their differences, the similarities dominate. As such, this section consolidates LD and HD developments.

REGENERATION

Zink (42) provided a comprehensive review of active DPF regeneration approaches and systems. The numerous examples for LDD applications (43, 44, 45, 46, 47) show common characteristics, summarized in Figure 12:

- Estimation of DPF soot loading using engine and back pressure models, and fuel consumption.
- Preheat the catalyst to ensure that injected hydrocarbons can ignite and heat up the filter.
- Increase of exhaust hydrocarbon levels via in-cylinder or supplemental fuel injection, for burning on a catalyst.
- Control and monitoring of the regeneration as a function of operating point and conditions.
- Recalculation of pertinent models to account for ash build-up.

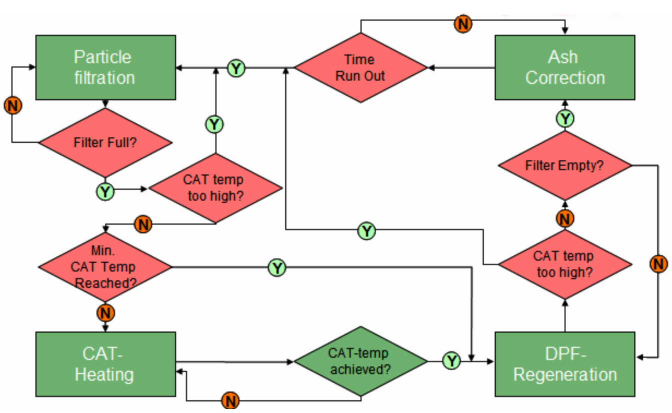

Figure 12. Example of LDD DPF regeneration plan. (45)

Each aspect of the plan is specific to the system and manufacturer, and can be rather detailed and complex. Key control parameters are fuel injection timing and quantity, air flow using throttling and EGR, and filter type and catalyst. The approaches are also pertinent to heavy duty and non-road applications.

Manns, et al. (48) do a very thorough review of LDD system layout and implications to calibration. They conclude that close coupled catalyzed filters, with or without a DOC are preferred due to better thermal management, less oil dilution by fuel, and better hydrocarbon conversion. They also discuss heat losses, thermal gradients in the filter, and post- and late-injection strategies.

For HDD applications, Kodama, et al. (49) described the system Mitsubishi FUSO will be using. One aspect of HDD filter regeneration that can be especially difficult is achieving temperatures high enough to burn fuel in the exhaust system under low load and/or cold ambient conditions. Key aspects of their approach is to use both intake and exhaust throttling to achieve DOC temperatures of 250C to enable fuel burning. They can accomplish this at speeds as low at 14 km/hr and in temperatures as low at -10C. Also noteworthy of the Mitsubishi approach is that instead of limiting regeneration duration by time, they chose to use total oxygen delivered to the filter, Figure 13. They save 20% on regeneration fuel consumption at 44 km/hr, but this savings diminishes as the speed goes down.

Other solutions that aid the low load problem involve exhaust system insulation, close placement to the engine, and improvements in catalyst formulation. In the latter case, Holcomb (28) was able to drop the DOC ignition temperature from 240C down to 215C by converting from a platinum to a platinum/palladium catalyst formulation.

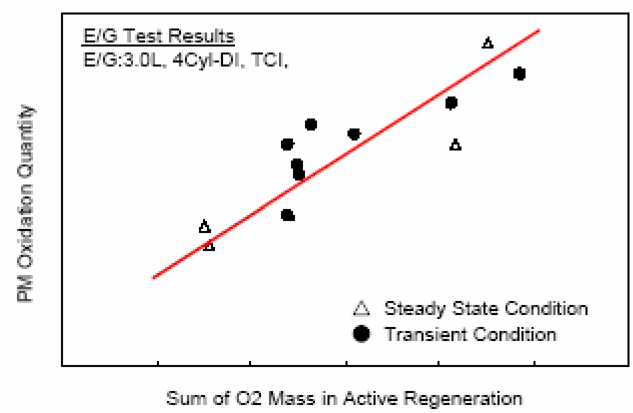

Figure 13. Total oxygen mass delivered to the filter during regeneration is used instead of time to determine when to stop DPF regeneration (49).

Mitsubishi is using SiC filters in their HDD application. Craig, et al. (50) describe the various regeneration properties of cordierite. As shown in Figure 14, peak filter temperatures and gradients, and completeness of regeneration are a function of DPF inlet temperature. Flow rate, soot loading, catalysts, and filter thermal mass also have significant impacts. The authors suggest that to increase soot loadings and improve regeneration efficiency, initial inlet temperatures of 550C might be used, which are then increased as soot loading decreases.

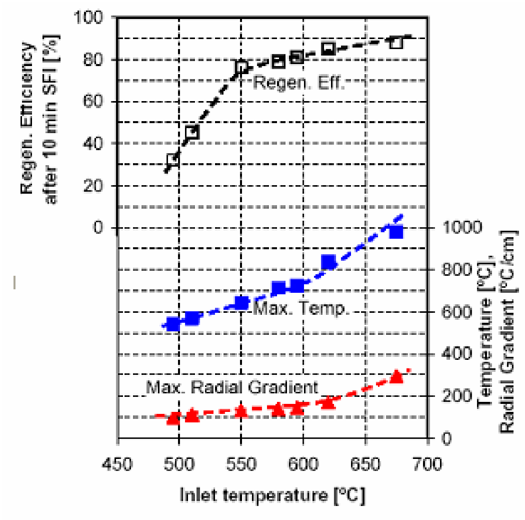

Figure 14. DPF inlet temperatures of >550C are needed to efficiently regenerate catalyzed cordierite filters with a 4 g/l soot loading. To further enhance regeneration, the inlet temperature can be increased later in the process (50).

Other items of note on recent DPF regeneration reports include:

- By moving the filter system from an underfloor to a close coupled position, the amount of regeneration fuel goes down because the system is inherently hotter. As such, the optimum soot loading, which balances fuel consumption due to increased pressure drop with regeneration frequency, drops from 10-11 g/liter down to 6-8 g/liter for SiC filters (51).
- Regeneration fundamentals using oxygen and NO_2 are investigated for catalyzed filters. NO_2 formation and utilization for passive regeneration is a complex relationship between NOx flux and temperature (for NO_2 formation and for oxidizing soot). Maximum passive regeneration occurs at medium to high speed and medium load conditions (52).
- It takes 65 to 70% of the total regeneration fuel to burn the last 25% of soot (53).
- Description of a supplementary fuel vaporizer that improves fuel distribution in the exhaust (54).

FILTER MATERIALS AND CATALYSTS

A new filter material besides SiC is being used in a high volume series production application (55). In an earlier report, Ogunwumi, et al. (56) describe the fundamental aspects of the aluminum titanate based composition, and later Heibel, et al. (57) describe engine and vehicle testing. In the latter paper, back pressure with soot and ash, regeneration properties as a function of soot loading, long term durability testing, and ash loading properties are described. Back pressure is lower due to tight pore size distribution and cell geometry, specifically, using an asymmetric cell structure (inlet cells larger than exit) and minor changes in cell density and wall thickness. Figure 15 shows some back pressure comparisons for fresh and loaded filters. Even though the filter has low thermal conductivity, the high heat capacity and physical properties enable an unsegmented design with maximum soot loadings of about 8 g/liter. This compares with cordierite in the 4 g/liter range and SiC in the 10-15 g/liter range.

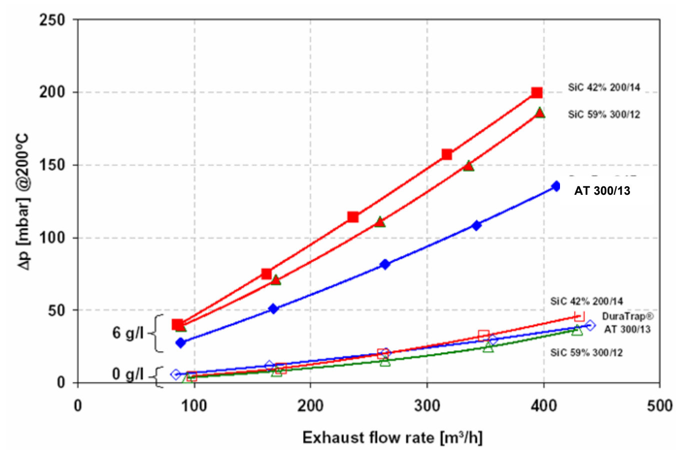

Figure 15. A new aluminum titanate filter that is now in series production has comparable unloaded back pressure as an SiC standard, but is 25% lower at 6 g/liter soot loading (57).

On other alternative filter materials, Mao, et al. (58) gave an update on a mullite filter with a novel whisker microstructure. The high porosity (60%) 25 μm pore structure allows low back pressure at high catalyst loadings (15% lower than SiC despite 3X washcoat loadings – 150 g/l). There was no deterioration in the 95% filtration efficiency after 100 regenerations at 8 g/liter soot loadings. On other designs, although metal “open” filters, wherein gas can either pass through filter media or go through unfiltered, are in both a LD and HD series applications they may have a niche in some retrofit applications (59). Presti, et al. (60) show 40% filtration efficiency for an unloaded open filter in a LDD application, but this drops to 25% efficiency as the filter loads up. Konstandopolous, (53) did a comprehensive paper on DPF design and performance. They show that depth filters, with an average pore size of about 40 µm, result in lower pressure drop and better soot penetration into the wall. This results in lower regeneration temperatures due to more intimate contact between soot and catalyst.

Improving catalyst behavior on DPFs will be important as engines move into mixed mode combustion. It is expected that oxidizing the high concentration of HCs from these engines at low load on the DPF will also continually burn soot (6). Ido, et al. (61) show that much catalyst within the wall might be under-utilized. Despite using DPF substrates with widely different pore size distributions, after washcoating the distributions all broadened. This will result in gas preferentially flowing through the large pores. Catalyst in the small pores thus is under-utilized. This phenomenon might contribute to the observation that system CO removal is higher if the PGM is distributed more on the DOC than a catalyzed DPF (62). On the other hand, catalyzed filters can give better regeneration behavior than a comparable system with a DOC+ catalyzed DPF (63). The filters, with an improved coating, burned more soot at 450 and 600C than if the HC oxidation process occurs primarily on a DOC. Instead of mainly residing on the wall, the improved coated penetrated into the wall.

ASH MANAGEMENT

MECA (Manufacturers of Emission Controls Association) published a paper on the state of ash management (64). The reader is directed there for a much more comprehensive review of the topic than can be covered here. The report covers sources and composition of ash, filter designs, filter maintenance intervals, and cleaning procedures.

Soeger, et al., (65) show that, although high filter back pressure is one reason to clean filters of ash, performance can be adversely affected. Figure 16 shows that the balance point temperature (temperature at which soot accumulation and oxidation are balanced) decreases from 369C for a filter aged 75,000 km in a long haul application, to 349C after ash cleaning. Similar results are available for NO_2 generation and HC oxidation.

Plumley (66) shows at 25 and 50% load, lube oil consumption rates are similar, as they are for higher loads at lower engine speeds. However, as shown in Figure 17, at 50 and 75% load consumption markedly increases. Given that low load applications, like refuse trucks and transit buses, require more frequent ash cleaning (around 48,000 to 90,000 km) than high load applications, like fuel transporters (600,000 km), these results indicate that perhaps hours of operation rather than mileage might be a better index of ash cleaning interval. Plumley also shows that soot is about 10% ash.

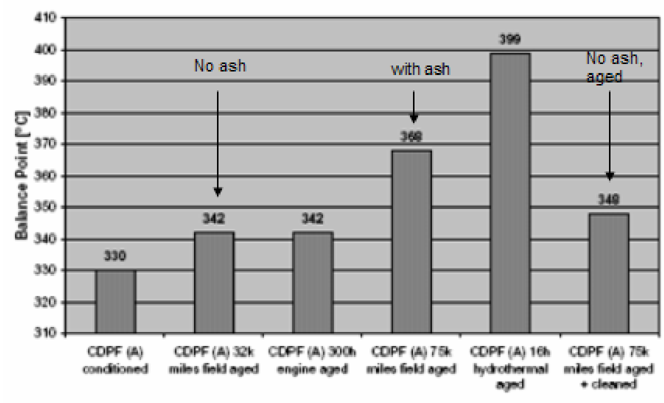

Figure 16. Ash can affect balance point temperature (65).

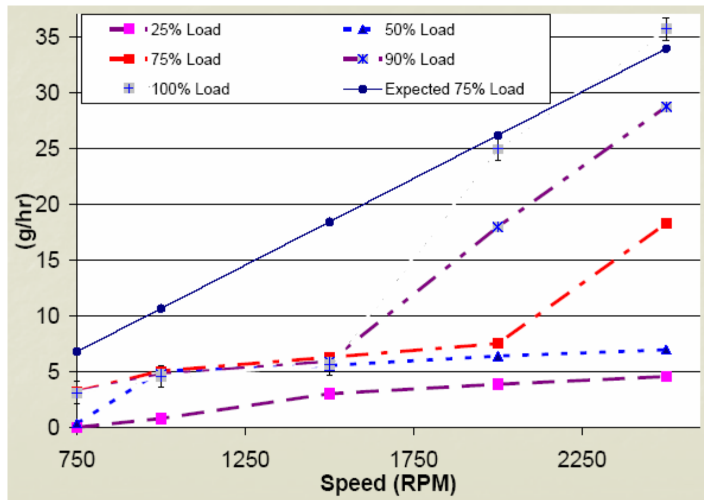

Figure 17. Lube oil consumption rates at 50, 75, and 90% load are similar up to about 1500 RPM, after which the consumption at 90% load increases (66). 2002 Cummins ISB, 5.9 liter engine.

Finally, researchers investigating ash effects have proposed a puzzle. Bardasz, et al. (67) did a mass balance on the lube oil consumed and the ash collected on a high-efficiency DPF, and found that only 46% of the phosphorous, 37% of the calcium, 31% of the magnesium, and 5% of the boron from the lube oil are captured on the filter. Others have found similar results to varying degrees, and no component is collected at >50%. One might jump to the conclusion that this ash goes through the filter, but no one has shown such, and these filters generally have high efficiency (80+%) in the size range of the fundamental ash particle (10-30 nm), and 99%+ efficiency for agglomerates 2-3X this size.

PM MEASUREMENT AND FILTER PERFORMANCE

Measuring the PM emissions when DPFs are used is quite challenging. Emissions are relatively undetectable compared to those if filters are not used. As shown in Figure 18, Khalek, et al., (68) shows that qualified analytical filters can result in a 3.5X range of PM emissions, only depending on filter type. The error is not on the low weight filters letting PM through, but rather those filters that collect more PM may have flaws that allow non-PM aerosols to accumulate. Similar results on dilution ratio variability and other parameters also are shown to impact the measurement (69, 70).

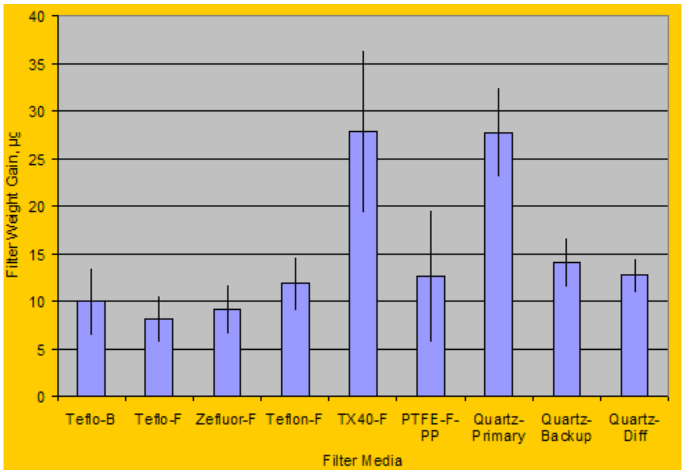

Figure 18. The choice of analytical filter can impact PM emissions results for systems using DPFs. Here two approved filters have 3.5X more emissions on them than other approved types (68).

To alleviate some of these problems, the European Union is investigating a number-based protocol instead of a mass-based one. Figure 19 shows preliminary results (71). Although there is more variability in the number-based measurement for filtered diesel, the method is able to differentiate multi-port gasoline (MPI) from direct injection gasoline (GDI) much better than the mass-based method.

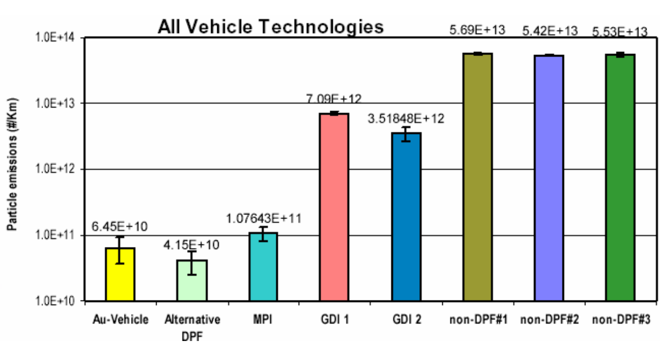

Figure 19. Preliminary results from the European PMP test program to develop a number-based PM measurement method (71).

Regarding performance, an earlier report on Paris taxis (72) shows ultrafine numbers increasing 10X after 80,000 km (but still removing 99% of them). A more recent AECC (Association of Emission Control by Catalysts) shows (70) little additional deterioration after 160,000 km of aging, and these emissions are still only 10% that of similar Euro IV gasoline vehicles. In fact, this and other studies (57) show PM mass efficiency improvement with age due to ash build-up.

Finally, DPF systems are not without issue. A key problem that is gaining attention is the generation of NO_2 from filter systems that use the compound to oxidize soot. Not included are systems that primarily use a fuel borne catalyst and/or heat from excess fuel to oxidize all the collected soot (no NO_2 regeneration). Mayer (73) shows that tailpipe NO_2 concentrations increase 2.5 to 6X at 25 to 50% load across the filter systems, depending on vehicle and system. However, one has to consider that NOx flux is low under these conditions. Even so, at full load NO_2 from modern filter systems can go up 20 to 60%. California is considering an NO_2 limit, even though modeling shows no significant increase in ground level ozone in conservative scenarios, and personal exposure is well below occupational health limits. Better system engineering, and some new system designs (74, 75) are beginning to address the problem.

SUMMARY – PM CONTROL

Although we may see LD and HD engines meet the tight US and Japanese standards without NOx treatment, it is doubtful we will see the regulations being met without filters. Given the growing evidence on the adverse health effects of fine particles, it is reasonable that diesel particulate filters will propagate through all countries, just like we are seeing with the catalytic converter. As we learn more about regeneration control, and as the materials (substrate and catalyst) improve, costs will continue to decline and performance will get better.

In that regard, filter regeneration strategies are reviewed, with new methods described for controlling regeneration duration, efficiency, and safety. The performance of new filter materials are summarized, as well as some new catalysts and coating methods. A summary of the ash management is provided, while presenting a question on ash deposition and the mass balance. Challenges in measuring PM at the very low emission levels are summarized, as well as that of tackling the NO2 emission issue. More results on performance and durability were summarized.

HYDROCARBON AND CO CONTROL

Although diesel oxidation catalysts (DOC) are a relatively mature technology, having been placed on 10's of millions LD diesels in Europe, and on HD applications in series production in the late 1990's, two recent developments are very noteworthy. The first is the developments of an oxidation catalyst that can operate with high-sulfur fuel (2000 ppm) without significant SO_2 formation (76). Figure 20 shows that with the new catalyst sulfur-based particulate is the same as that coming out of the engine, compared to a

3X increase with other typical formulations. Such a development has interesting applications in retrofit applications in developing countries with high sulfur fuel and older diesel engines.

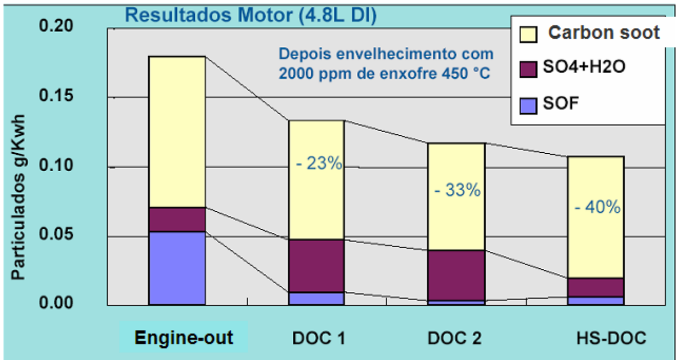

Figure 20. A new high sulfur DOC (HS-DOC) does ot form sulfates in 2000 ppm sulfur fuel, despite high temperatures (76).

The second development (77) is a low temperature DOC that has application in both retrofitting of older vehicles with cold exhaust, Figure 21, and possibly for the emerging mixed mode engines, wherein highly efficient oxidation catalysts will be needed to treat cool exhaust with high HC and CO levels. For the case of older engines at idle, 80-90% of the soluble organic fraction (SOF) is removed even though the temperatures are only 105-163C.

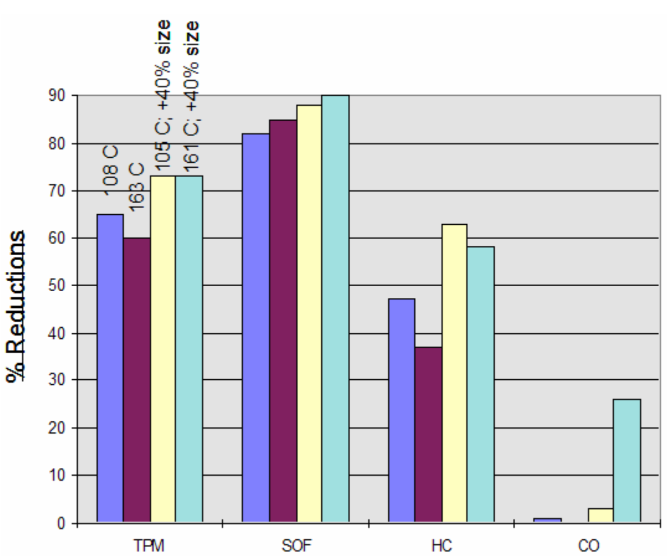

Figure 21. Results on a new LT DOC on an older engine. 80-90% SOF reductions given 60-70% total PM reductions under idle conditions (105-161C). Reference 77.

INTEGRATED NOX AND PM CONTROL

The first applications of integrated NOx and PM control will likely be in US Tier 2 Bin 5 LD applications. At the 2006 Detroit Auto Show, DaimlerChrysler announced an E-class that will use a DOC followed by an LNT, DPF, and an SCR system (78) to meet the Bin 5 standards. VW officials have made a less formal announcement of a Bin 5 Jetta for 2006 introduction (79), although the system was not described. On a related matter, the US EPA showed a Ford minivan prototype that uses high EGR, boost, and advanced fuel injection and a DPF without NOx aftertreatment to obtain Bin 5 emission levels (80). Ford and International Truck and Engine Company are in a cooperative research and development agreement with the EPA on this engine.

Neely, et al. (81), provide the first insights into an integrated system using mixed mode control in a LDD application, Figure 22. As LT NOx is not a significant problem, the LNT is placed after the DPF. Both low-pressure (after DPF) and traditional HP EGR are used for engine control. Rich PCCI (premixed charge compression ignition) and rich LTC (low temperature combustion) are used to regenerate the LNT under low load conditions. Steady state simulations show Bin 3 potential for the system.

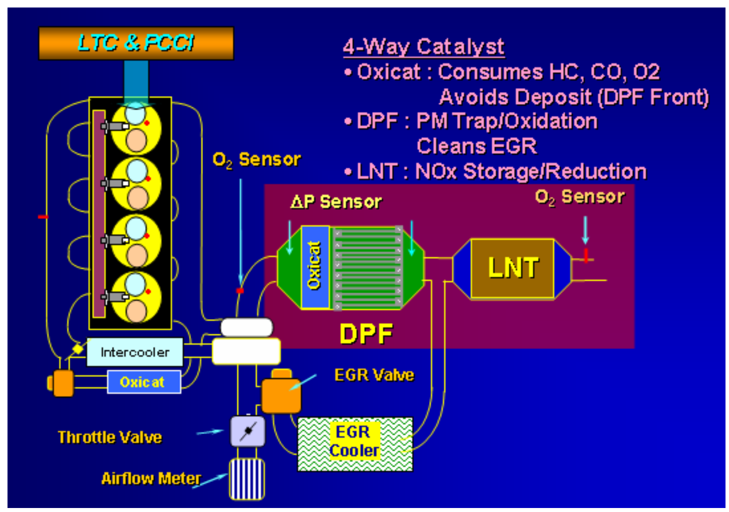

Figure 22. Example of an integrated emission control system for a mixed mode LDD engine. Converse to typical LDD architecture with traditional diesel combustion, the LNT is behind the DPF as LT NOx control is no longer a major issue (81).

Looking towards US2010, a DPF+SCR retrofit system on a 12 liter Caterpillar engine retrofit with an EGR system (82) hit the required NOx levels at 0.18 g/bhp-hr and PM levels at 5 mg/bhp-hr, with no deterioration after 6000 hours of testing. The more challenging NTE performance was missed at one steady-state load point.

Finally, although the tight US and European non-road regulations are five years off (2011), Baumgard, et al., (83) showed the first insights into how a US Tier 4 2011 system might look, Figure 23. They used a high pressure common rail system and cooled EGR to take a Tier 2 John Deere 6068H 6.8 liter engine to Tier 4 interim NOx levels. A DPF got them to the PM levels.

The new configuration results in a 3 to 5% fuel penalty versus Tier 2.

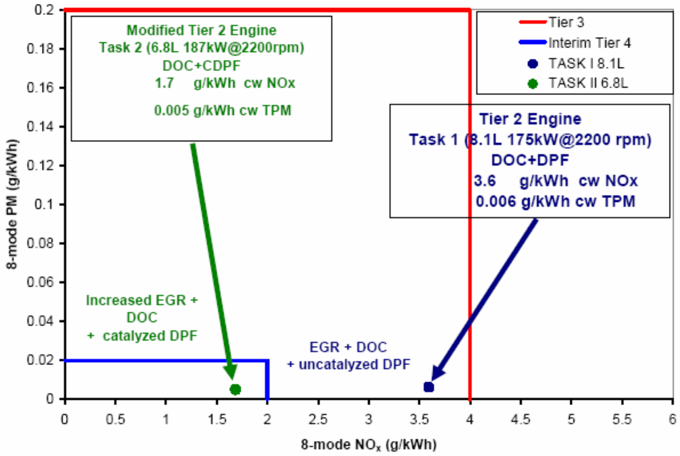

Figure 23. A interim Tier 4 (2011) non-road system, using high-pressure common rail fuel injection and cooled EGR to hit the NOx levels and a DPF to hit the PM levels. Fuel penalty is 3-5%. John Deere 6068H Tier 2 engine (83).

CONCLUSIONS

The regulatory framework for LDD, HDD, and non-road applications through 2014 are described, as well as emerging engine technologies. This helps define the future aftertreatment requirements. In general, for LDD using incremental advances in traditional diesel combustion, although DPF will be needed, nominally US Tier 2 Bin 8 (0.12 g/km) can be achieved today with no NOx aftertreatment, as well as Euro V, and Japan 2009 will likely be hit in the future. US Bin 5 will need 70% NOx control and DPFs. As mixed mode combustion develops, Bin 5 passenger cars will likely require no NOx aftertreatment in 2009, followed by the same status for heavier LD vehicles in 2011. For HDD The Japan 2009 regulations show good chances of being hit with advanced combustion, at least as indicated by today's research engines, but NOx control will likely be extended from Japan 2005 to offer better fuel economy. For US 2007, 50% NOx control at 500-520C is indicated by today's research engines to hit the tough NTE requirements.

For NOx control, LNTs will meet the LDD requirements, and SCR leads the HD field. The middle sized applications could go either way. SCR is moving towards zeolite catalysts, and reports are surfacing on secondary emissions. For LNTs, HT formulations to address HD NTE are being reported, and PGM loadings are dropping.

Filters are continuing the movement towards cost reduction and performance improvement. DPF regeneration strategies are summarized and quite sophisticated. New filter materials are emerging, and for the first time in six years, a wall-flow filter of a new material (aluminum titanate) is in series production. Substrate materials are improving, as are catalysts, and reports are offering insights for improvements. Ash management is emerging as a key concerns, and PM measurement in clean exhaust has its challenges.

Regarding HC and CO control, two recent developments are significant, especially for retrofit applications in developing countries: a high sulfur DOC is reported that does not form sulfates, and a LT DOC that works well at 105C is available.

On integrated systems, the US Tier 2 Bin 5 application will be the first high production application for integrating both PM and NOx control. We may see two vehicles meeting this requirement by the end of 2006. The first mixed mode system for LD application is also described. US2010 appears within reach, and the first paper on hitting the interim US Tier 4 non-road regulations for 2011 is summarized.

CONTACT

JohnsonTV@Corning.com

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