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Progress and Outlook on Gasoline Vehicle Aftertreatment Systems

Meeting the tightening limits for criteria pollutants and greenhouse gas emissions for China, Europe and the USA

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Driven by concerns on deteriorating ambient air quality, measures are being taken across the world to adopt and enforce tighter vehicular emission regulations to minimise tailpipe unburned hydrocarbons, nitrogen oxides (NOx) and particulate matter (PM). In regions with advanced regulations, the focus is on limiting the pollutants under real-world or in-use driving conditions. Given the intensified effort to curb global warming and limit fossil fuel use in the transportation sector, several countries have adopted targets on tailpipe carbon dioxide emissions. This confluence of stringent regulations for both criteria pollutant and greenhouse gas (GHG) emissions is leading to a rapid adoption of advanced powertrains and aftertreatment technologies. This is a review of some of these recent advances pertinent to reducing vehicular emissions and developing improved aftertreatment solutions. The scope is limited to gasoline vehicles where the adoption of gasoline direct injection (GDI) and hybrid powertrain technologies is leading to significant shifts in the aftertreatment solutions. There is significant work being done to improve diesel aftertreatment systems especially in light of real-world driving emission (RDE) regulations. These are not covered here, rather the reader is referred to a previous article in this journal's archive (1), and to a more recent review (2).

On the regulatory changes, the focus is on Europe and China, where Euro 6 regulations are being implemented along with changes to the certification test cycle and RDE targets. The combined non-methane organic gas (NMOG) + NOx target of 30 mg mile^{-1} in the USA will be the tightest standard globally by 2025, accompanied by the GHG reductions being discussed for light-duty vehicles. A variety of advanced vehicle technologies are being developed and implemented to meet these targets, and some of these are discussed along with the implications from an aftertreatment systems perspective. As gas temperatures continue to decline as the engine improvements lead to heat rejection to the exhaust, innovative approaches are focused on lowering cold-start emissions. Particulates from gasoline engines are also regulated and various control measures are under investigation. Gasoline particulate filters seem poised for wide adoption in major markets. Electrification of the powertrain is proceeding rapidly as well, and while this can deliver impressive gains in fuel economy, cool-down on aftertreatment components and emissions associated with engine restarts are being studied. On the other hand, the internal combustion (IC) engine continues to evolve, and advanced gasoline engine technologies are challenging the well-to-wheels CO_2 emissions of electric powertrains. Research continues on low-temperature combustion with the aim of avoiding soot and NOx formation altogether, and researchers are pursuing several strategies. By definition, these lead to low exhaust temperatures which pose a challenge to the aftertreatment system.

1. Light-Duty Regulations

Figure 1 gives an overview of the latest regulations for gas emissions implemented in Europe, China and the USA. The US Environmental Protection Agency (EPA) Tier 3 standards will be the toughest in the world, with reductions phased in starting 2017 through 2025 and ending with a combined limit on NMOG + NOx at 30 mg $mile^{-1}$. **Figure 1(a)** shows the concurrent targets for emissions of criteria pollutants and GHGs. **Figure 1(b)** compares this with the tightening of emissions limits happening elsewhere in Europe and China through the Euro 6 and China 6 regulations, respectively.

Euro 6 light-duty (LD) regulations introduce several important changes. A particle number limit of 6×10^{11} km^{-1} applies to GDI vehicles. Starting in September 2017, new type approvals are to be certified using the new World Harmonised Light Vehicle Test Procedure (WLTP) (3). The third RDE package has been recently published (4), which confirms the conformity factor (CF) of 1.5 for particulate number (PN). The CF is expressed as (1 + margin PN), with the margin set at 0.5, providing for measurement uncertainties associated with the portable emissions monitoring system (PEMS) equipment, and subject to annual review. The CF for NOx was confirmed at 2.1 in previous packages.

China 6 LD regulations will be implemented in two stages: China 6a starting in 2020 with RDE monitoring, and China 6b in 2023 with tighter limits and full compliance with RDE. CFs will be finalised by 2022. Major regions such as Beijing may adopt regulations earlier, but the details are yet to be clarified. There are some important differences compared to the European regulations. The regulations are fuel neutral and make no distinction between GDI and port fuel injected (PFI) vehicles. This is an important provision, as PFI vehicles have high PN emissions under cold ambient conditions, and PFI hybrids emit particulates even beyond cold-start (5) (likely due to transients associated when the engine turns on). As shown in **Figure 1**, the gas emissions in the final China 6b stage will be much tighter, by roughly a factor of 2 for carbon monoxide and hydrocarbons (HC), 40% lower for NOx and 33% lower for PM. There is also a limit of 20 mg km^{-1} on nitrous oxide (N_2O) emissions, absent in Europe (N_2O is also limited in the USA at 10 mg $mile^{-1}$). A recent study by Ricardo (6) shows that managing N_2O emissions at such low levels is not trivial, and that catalysts will have to be optimised to meet such tight limits. The durability requirement is also increased to 200,000 km for China 6b, as compared to 160,000 km in Europe, and an emission warranty and defect-reporting requirement has been implemented for the first time. China is serious about strict enforcement of these regulations, and this is reflected in the new framework for certification, conformity of production and in-use compliance testing (7). Much of the onus for these tests lies with the

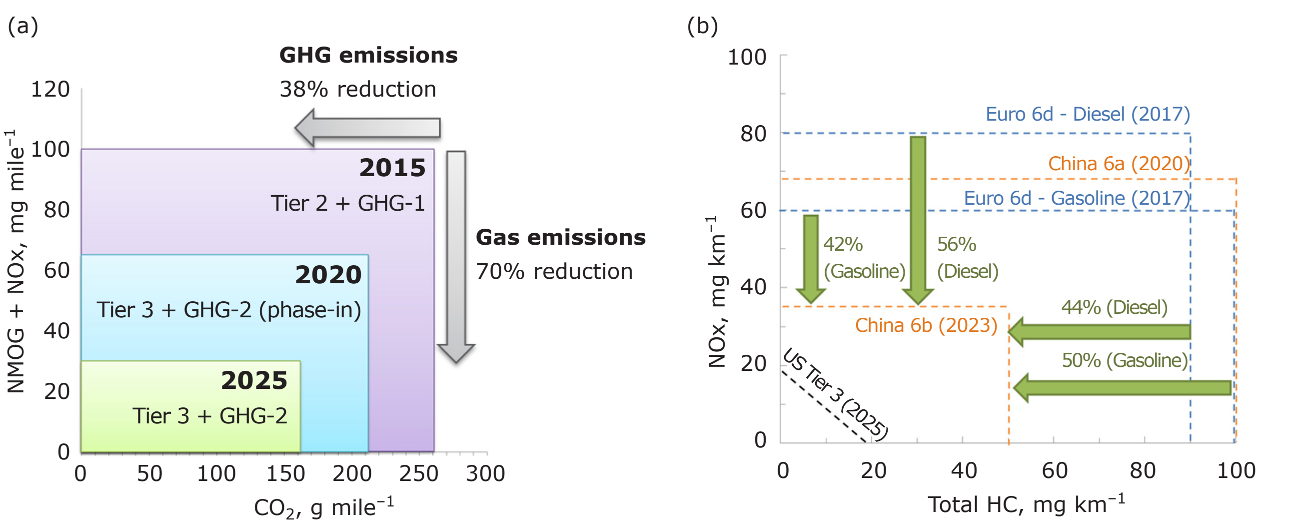

Fig. 1. Light-duty regulations for tailpipe emissions in major markets: (a) the concurrent tightening of criteria pollutant and CO_2 emission limits in the USA; (b) by 2023, the proposed limits in China will be tighter than those in Europe, roughly by a factor of two

original equipment manufacturers (OEMs), while the regulatory authorities maintain the right to check the results.

India will skip one level of regulations, the first of its kind for a major market. It will move from Bharat Stage (BS) 4 directly to BS6 for LD regulations, and BS VI for heavy duty (HD) in 2020. RDE test procedures are being developed to reflect the local driving conditions, and compliance is expected to be enforced by 2023. Other countries such as Australia and Thailand are also rapidly implementing stricter regulations with a view to Euro 6 levels in the coming years. These global changes have implications for rapid development and deployment of advanced engine technologies and engine emissions aftertreatment systems, which are covered in the following sections of this review.

In contrast to the other major regions listed above, PM regulations in the USA are mass based: starting in 2017, both EPA Tier 3 and California Air Resources Board (CARB) Low-Emission Vehicle III (LEV III) standards set limits on the allowable tailpipe particle mass at 3 mg mile^{-1} for light-duty vehicles (8). The LEV III regulations continue to tighten to 1 mg mile^{-1} starting 2025.

2. Advances in Engine Technologies

The broader technology trends are being shaped by the concurrent needs to deliver on vehicles with high fuel economy and near-zero tailpipe gas and particulate emissions. One study (9) discussed pathways to meet the CARB target of 48 g_{CO_2} km^{-1} by 2050, an 80% reduction over 2010 levels. It was shown that for the goals to be met independently through IC engines, fuel cell vehicles or pure-electric vehicles, a large shift to low-carbon fuels is needed: 60–80% of fuel will have to be derived from cellulosic ethanol, 48% of H_2 and >36% electricity generation from renewables, respectively. This points to a long-term shift, already underway, to renewable fuels and electrification of the powertrain. In the short term, meanwhile, various engine improvements are being deployed to improve the efficiency of modern vehicles.

In their joint Draft Technical Assessment Report (10), the EPA, National Highway Traffic Safety Administration (NHTSA) and CARB concluded that the GHG standards for model year 2022–2025 LD vehicles can be met using a wide range of advanced gasoline technologies. A recent analysis (11) concludes that the costs of advanced vehicle technologies are rapidly falling, such that compliance with the 2025 GHG standards could be up to 40% lower than anticipated in the joint assessment. Some of the advanced engine technologies which are already being adopted rapidly include direct injection, cooled exhaust gas recirculation (c-EGR), high compression ratio (CR) Atkinson cycle, stop-start, cylinder deactivation, advanced turbocharging and mild hybridisation.

A detailed review of all these technologies is not the focus of this paper. But we will highlight some of the recent work. c-EGR is effective in reducing the tendency to knock at high loads, therefore eliminating fuel enrichment. At part loads, both c-EGR and cylinder deactivation improve fuel efficiency through reduced pumping work. A recent study (12) looked at the incremental benefit of adding these technologies to a 2 l naturally aspirated Atkinson engine with high CR (14:1). The brake thermal efficiency (BTE) improvements over the engine map were used to assess the improvements over the Federal Test Procedure (FTP)/Highway Fuel Economy Test (HwFET) two-cycle CO_2 reductions. For a future vehicle, the improvements using c-EGR were predicted at 7.6%, while cylinder deactivation gives another 2% improvement.

Variable compression ratio (VCR) technology was commercialised recently by Nissan, achieved *via* its 'multi-link' system (13). The CR can be varied between 8:1 and 14:1. A combination of VCR, multi-port injection, direct injection and downsizing helped realise a 30% fuel economy improvement when moving from a conventional 3.5 l V6 engine to a 2.0 l engine. The technology has benefits for reduced emissions: high CR leads to improved fuel consumption and lower CO_2 emissons, while lower CR during cold start helps with higher exhaust temperatures for early catalyst light-off and lower particulates due to reduced fuel impingement on the piston.

Dedicated-EGR, where one cylinder's exhaust is fed into the intake manifold for constant 25% EGR for a four-cylinder engine, was demonstrated to deliver brake specific fuel consumption (BSFC) <200 g kWh^{-1} from 12–14 bar brake mean effective pressure (BMEP) and 1500–3500 rpm. Vehicle testing showed the potential to reach NOx + NMOG emissions of 31 mg mile^{-1}, just above the LEV III limit. Lower exhaust temperatures pose a challenge for HC emissions, and HC traps are being considered (14).

Water injection is gaining some interest as a potential technology to mitigate knock and fuel enrichment at high loads. Studies show the potential to achieve ~5–7% reduction in fuel consumption over test cycles, with much higher gains up to 17% observed at full load (15, 16). The amount of water needed can be high, the studies cited above have noted the best fuel efficiency obtained at water-to-fuel ratios upwards of 50%. The exhaust temperature is reduced (by ~50–100°C), such that NOx emissions are reduced, but HC emissions increase.

Much progress is also being made on achieving lean, low-temperature combustion *via* compression ignition engines. Oak Ridge National Laboratory, USA, has summarised the various levels of fuel stratification strategies being pursued (17), as shown in **Figure 2**. The overall goal is to achieve: (a) sufficient premixing of fuel and air to avoid soot formation associated with fuel rich combustion, and (b) reduced peak combustion temperatures *via* dilution with air or EGR to avoid NOx formation. While promising high fuel efficiency, the lower combustion temperatures also imply high unburned HC and CO emissions and the challenges with achieving downstream catalyst warm-up. Other

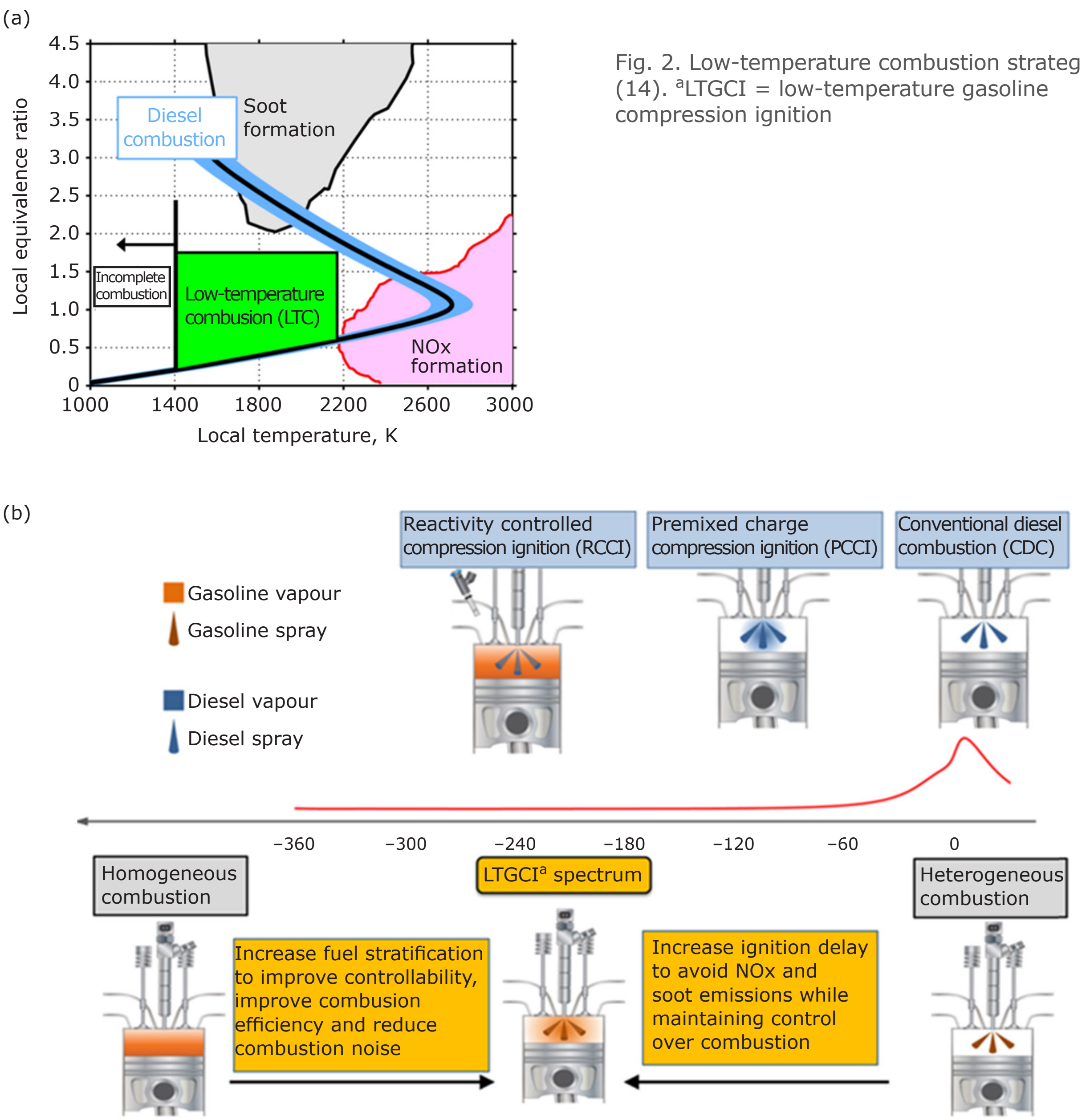

Fig. 2. Low-temperature combustion strategies (14). [a]LTGCI = low-temperature gasoline compression ignition

challenges include operability over a wide load range and combustion timing control.

An example of the progress made is the development of the gasoline direct injection compression ignition (GDCI) engine targeted to meet Tier 3 Bin 30 limits (18). BSFC of 211–214 g kWh^{-1} is demonstrated over a wide load range, utilising a high CR of 16:1 and injection pressure of 350 bar. The difficulty of achieving the very high conversion efficiencies (>99%) needed for HC have translated into a rather complex aftertreatment system, which at the latest stage includes a pre-turbo catalyst, HC trap, selective catalytic reduction (SCR) and a passive gasoline particulate filter (GPF) for off-cycle particulates. As this article was being written, Mazda (19) has announced its plans to introduce the first supercharged homogeneous charge compression ignition (HCCI) engine to market in 2019, projected to cut fuel consumption by 20–30% over its current gasoline engines. This is in line with its vision (20) for achieving 25% improvement in IC engine efficiency for well-to-wheels CO_2 equivalence with electric vehicles (EVs) in many electrical grids.

As mentioned at the beginning of this section, despite the advances in engine improvements listed above, there is a growing consensus that these may be insufficient to meet CO_2 targets beyond 2025, and OEMs are increasingly adopting various levels of electrification to lower fuel consumption. Start-stop technology is becoming more prevalent, full hybrids and plug-in hybrid electric vehicles (PHEVs) can offer >30% reduction in fuel consumption, while mild hybrids using a 48 V battery look particularly attractive, providing many of the benefits of a full hybrid at a fraction of the cost. The actual benefit of hybrids is tied closely with the driving conditions and state of charge (SOC) of the battery. In a study simulating a LD vehicle with advanced engine configurations (21), it was shown that using a hybrid powertrain, the fuel economy almost doubled over the New York city cycle (compared to a naturally aspirated 1.6 l, four-cylinder baseline), while the benefit was only 12% on highway and real-world driving conditions. Pure EVs remove the use of IC engines altogether and as such can offer the highest benefits from electrification, although the well-to-wheels benefit is closely tied with the carbon footprint of the fuel used to generate the electricity.

3. Stoichiometric Gasoline Emissions Aftertreatment

New regulatory developments outlined in the first section – lower limits on tailpipe criteria pollutants, the introduction of particulate number and mass regulations for gasoline engines and challenging CO_2 or fuel economy targets – are stimulating work on new and improved aftertreatment systems for gasoline engines. The underlying challenge for catalysts is achieving early light-off despite the lowering exhaust temperatures as a consequence of efficient combustion. Improved combustion and the introduction of gasoline particulate filters is imminent to meet the particulate regulations under real-world driving conditions.

3.1 Three-Way Catalysts

The cold-start challenge is highlighted in a recent study (22) comparing the emissions across 82 LD vehicles with both PFI and GDI technology, complying with various levels of regulations (Tier 0 to super ultra-low emissions vehicle (SULEV)). Tailpipe emissions were measured on the cold-start unified cycle (UC). The ratio of cumulative cold-start emissions in the first bag divided by the distance-specific emissions after engine warm-up in the second bag was used to quantify the cold-start contributions to overall emissions. For HC, this ratio increased from 15.4 miles for Tier 1 vehicles to 101.5 miles for SULEV vehicles. Put in words, modern vehicles will have to drive over 100 miles after engine warm-up for the emissions to equate to the first few seconds after a cold-start. This points to the tremendous progress which has been made in catalyst technology – the conversion is near complete once the catalyst is hot and active – but it also points to the challenge ahead to address the cold-start emissions.

Light-off in three-way catalysts (TWCs) is not a new topic. The various factors which impact cold-start conversion have been studied extensively, both experimentally and using simulations (23). For TWCs, there are several approaches being explored to achieve lower cold-start emissions. One is increasing the precious metal content, but that adds to the cost and also produces limited improvements: the additional catalyst will still be ineffective at very low temperatures. Another is optimising the location of the platinum group metal (pgm). According to one analysis (24), the pgm costs could vary by a factor of two (US\$35–US\$70) depending on the catalyst location for achieving non-methane hydrocarbon (NMHC) + NOx emissions of about 42 mg $mile^{-1}$. Conversely, a US\$40 pgm loading might deliver NMHC + NOx emissions between 105 mg $mile^{-1}$ and 30 mg $mile^{-1}$, depending on the catalyst design. Continuing along the pgm route, another approach is the catalyst design: choice of precious metals, their concentrations and the choice of support. One such study (25) aims at lowering the light-off temperature to achieve 90% conversion of HC, CO and NOx on aged catalyst at 150°C (T_{90}). Ongoing work has considered several design levers:

(a) catalyst preparation: the results suggested post-impregnation may provide better access to pgm than catalysed slurry, under the conditions tested;
(b) palladium level: the best performance was found at 2% concentration on alumina (Al_2O_3) support, while higher loadings helped on zirconia (ZrO_2) and titania (TiO_2) supports;
(c) the support itself: Al_2O_3 has much higher surface area than ZrO_2 and TiO_2. Fresh ZrO_2 has lower fresh T_{90}, but Al_2O_3 has better aged performance;
(d) impact of ageing: the performance actually improved with ageing (lower T_{90}) for Al_2O_3 due to better Pd dispersion, while it deteriorated on the other supports.

Much progress has been made *via* optimising these variables, so that T_{90} for HC was lowered from 345°C on a commercial catalyst to 253°C on a combination of 0.5% rhodium/TiO_2/Al_2O_3, under aged conditions (26). While this is impressive, there is clearly more work to be done to reach the ambitious goal of T_{90} at 150°C. Another study (27) focused on improving the oxygen storage capacity (OSC) and thermal durability using an advanced alumina and OSC material. An improvement in OSC of 15–30% was achieved over a wide range of temperature and mass flow rates, translating into conversion improvements for CO up to 20% and NOx up to 10% over wide air:fuel ratios.

A very different approach being considered is the reduction or elimination of pgm to reduce costs. New catalysts are being developed, aimed primarily at the underfloor location where the performance requirements are not as stringent as at the close-coupled position, and the lower temperatures pose milder conditions for the catalyst ageing. A new barium/zirconium/ceria (Ba/Zn/CeO_2) catalyst formulation was shown (28) as a viable alternative to Al_2O_3. The replacement of Al_2O_3 from the first layer of an underfloor Pd/Al_2O_3/OSC catalyst with Ba/Zn/CeO_2 led to a 10% reduction in NMOG + NOx emissions over the Los Angeles Route Four (LA4) test cycle using a 2011 model year Honda Civic (partial zero emissions vehicle (PZEV), 1.8 l L4). The Zn reduces the oxygen binding energy, increasing the O-availability and CO oxidation at low temperatures, while the barium reduces CO_2 adsorption which limits the oxygen availability. This catalyst was commercialised and applied to the 2016 model year Civic. Another example (29) is the use of iron-based catalysts as a way to reduce pgm. The combination of Fe/CeO_2 was shown to work: the CeO_2 reduces the binding energy of oxygen to Fe, and helps the catalyst respond to oscillating redox conditions. Normally Fe would react with alumina to form aluminates which leads to loss of aged performance, so a perovskite oxide ($LaFeO_3$) structure was adopted to inhibit structural changes. At only 10% of the Rh loading compared to a conventional catalyst, the aged performance of this catalyst was found to be similar on the Extra Urban Driving Cycle (EUDC). The reduction of pgm is going to be especially critical for meeting stringent upcoming regulations in cost sensitive markets, as is the case with BS6 in India. The viability of using advanced spinel oxides with low pgm loadings has been demonstrated (30) in both underfloor and close-coupled TWCs.

The choice of substrate has played a critical role in facilitating cold-start performance of TWCs. Reducing the thermal mass of the substrate is an effective method to enable early heat-up of the catalyst. This has been achieved through increased porosity of the substrates from the traditional 27–35% up to 55%, while meeting the strength requirements for canning and designing the right microstructure to maintain on-wall coating to provide good access for the reactants to catalyst sites (31). The substrates were shown to reduce light-off time and cold-start emissions by up to 24%, and have been commercialised (32). A different approach taken recently (33) is the use of substrates with varying cell design aimed at gas flow redistribution. The idea is to use higher cell density in the centre which increases resistance to flow, redistributes it towards the outer channels and improves overall gas catalyst contact. Vehicle testing on the US06 Supplemental Federal Test Procedure (SFTP) using the new substrate design in the underfloor position led to similar or better NOx conversion despite 20% lower pgm. However, as compared to the close-coupled position, the pgm content and the extent of flow non-uniformity are both smaller in the underfloor position. Other approaches, such as better packaging, are often used to address this issue in the close-coupled position.

Other than addressing cold-start and pgm content, there are some new system layout choices emerging to address the tightening limits of both criteria pollutants and fuel economy. One example (34) is the use of a 'fuel-cut NOx trap (FCNT)' replacing the underfloor TWC and placed downstream of the muffler. Fuel-cut events help improve fuel economy, but lead to excessive NOx emissions due to lean conditions. NOx emissions were found to increase by a factor of three due to such fuel-cut events on the FTP-75 test cycle. The NOx trap addresses this issue. Moving the trap behind the muffler helped reduce the temperature by 60–70°C and facilitate an optimal temperature window for the lean NOx trap (LNT). The FCNT helped upgrade an ultra-low emissions vehicle (ULEV) vehicle to SULEV standards, while retaining the fuel economy benefits of fuel-cuts.

The biggest change for TWCs from a systems-level perspective is the introduction of catalysed GPFs. As will be discussed in more detail in the subsequent section, GPFs are being developed for catalysed applications in both the close-coupled and underfloor position, where they combine the functionality of filtration and TWC. This is similar in many respects to the combination of SCR deNOx functionality on diesel particulate filters (DPFs) in diesel aftertreatment. Several studies have now shown that it is possible to maintain high reaction performance with catalysed GPFs replacing the traditional TWC coated flow-through substrates. One study (35) showed that an aftertreatment system was able to meet the SULEV30 emission targets of 30 mg NMOG + NOx *via* the use of a close-coupled high-porosity substrate mentioned in the previous paragraph and an underfloor GPF catalysed with Rh. While most of the NOx conversion occurred over the close-coupled TWC, the GPF helped to increase conversion by 18–30% over the US FTP-75 test cycle. These have also been tested in conjunction with an underfloor catalysed GPF, where a 10–12% reduction in NMHC and NOx over the FTP cycle was found over standard substrates. The system tested had two close-coupled catalysts and most of the conversion was found to occur over the first close-coupled catalyst. The best system tested achieved 25 mg mile^{-1} of NMOG + NOx, below the US Tier 3 Bin 30 limits, with the use of high-porosity substrates in both close-coupled positions.

In another study (36), the emissions of a GDI vehicle were measured over the FTP-75 test cycle with and without a catalysed GPF in the underfloor position. Through the use of the coated GPF, the tailpipe emissions of CO, total hydrocarbon (THC) and NOx were reduced further by 86%, 38% and 34% respectively. The catalysed GPF also helped reduce NOx preferentially under the aggressive driving conditions of the US06 test cycle, leading to 88% reduction in NOx over baseline.

The durability of coated GPFs was demonstrated for a 1.4 l GDI engine in China (37). After 160,000 km testing, the engine-out emissions were unchanged. The TWC performance was robust, with only a slight (~15°C) increase in light-off temperature, and also maintaining filtration efficiency (~85%) over its lifetime. A study evaluating the impact of RDE on emissions from two Euro 6b certified turbocharged GDI vehicles highlights the challenge ahead (38): one of the vehicles tested showed a NOx emissions increase under real-world driving conditions by a factor of two or three times compared to WLTP values. The NOx emissions exceeded the limit of 60 mg km^{-1} even with a fresh exhaust system. Replacing the underfloor TWC with a coated GPF helped to reduce both CO and NOx emissions.

3.2 Gasoline Particulate Filter

Boosted, downsized GDI engines offer improved fuel economy over the traditional PFI engines. The charge cooling associated with direct injection of fuel in the cylinder reduces the risk of engine knock and enables operation at higher compression ratios. Also, coupled with electronic controls, the amount and timing of fuel injected into the chamber can be more precisely controlled. However, similarly to diesel, insufficient time for mixture preparation and fuel impingement on the relatively cooler combustion chamber surfaces leads to pockets of fuel-rich combustion and an increased propensity for particulate formation. Europe and China have PN regulations to limit the fine particulates emitted, and there is accordingly considerable focus on engines, fuels and aftertreatment systems (for example the GPF) to control particulates. As mentioned earlier, particulate matter regulations in the USA are mass based, and while studies show that it may be possible to meet the 3 mg mile^{-1} standard using engine methods, the 1 mg mile^{-1} limit may require the use of GPFs. Several studies have shown a direct correlation between solid particulate number and mass emissions. Broadly, the studies (for example (39–41)) report a correlation of

2×10^{12} particles mg^{-1}, based on which the PN limit of 6×10^{11} km^{-1} translates to ~0.5 mg $mile^{-1}$.

A detailed review of fuels' impact on particulates is beyond the scope of this article, but there is sufficient evidence that components with higher resistance to volatilisation – such as aromatics – increase particulates, while other components such as ethanol – which adds oxygen – decrease particulates (36). The increasing particulates with aromatics is especially important and is being studied in China (42) given the variability in fuel quality. A PM Index (PMI) model has been proposed (43), which quantifies the relationship between fuel composition and particulate emissions. The model accounts for the double bond equivalents, vapour pressure and weight fraction of each component in the fuel, and several studies (44) have shown a strong correlation between the PMI and measured particulate emissions.

Many studies have confirmed that particulate emissions are highest during cold starts, due to lower fuel volatility, fuel impingement on colder surfaces and less time for evaporation. Cold ambient temperature has a similar effect, and one study (45) has shown that even PFI and hybrid vehicles exceeded the EU regulated PN limit of 6×10^{11} km^{-1} on the New European Driving Cycle (NEDC), when tested under sub-zero ambient temperatures. The average PN during the initial 180 s was almost identical for both GDI and PFI vehicles. In the EU, cold-start emissions are included in the RDE analysis following the latest (third) RDE package, and the PN limits apply to both GDI and PFI vehicles in China, so these findings are important.

In light of the particulate regulations discussed above, it appears that the aftertreatment systems of most modern GDI vehicles and also some PFI vehicles will include a GPF. The potential system layouts are shown in **Figure 3** (46), and include bare (uncoated) or TWC-coated GPFs. In the former, the main functionality is filtration only, while the latter provides the added functionality of gaseous emissions conversion. The filters can be added in the close-coupled or in the underfloor position.

Several studies have now shown that the backpressure with the addition of a GPF can be maintained relative to the base OEM system, thereby having little or no impact on fuel economy. There are several design levers which can be turned to avoid incurring a CO_2 penalty due to addition of GPF. The choice of larger filter diameter, optimised washcoat loading, cell design of both the filter and upstream TWC, are all factors which can help keep tailpipe CO_2 at levels close to the original system. In a previously cited study (32), fuel economy with a bare GPF was found to be equivalent to the OEM system with a traditional underfloor TWC, while particulate emissions reduced to 0.27–0.4 mg $mile^{-1}$ over the FTP-75 test cycle, comfortably meeting the 1 mg $mile^{-1}$ proposed standard. Other than proper design of the GPF, the pressure drop reduction was also assisted by replacing the 900/2 close-coupled catalyst with a 600/3 cell design. Moreover, the GPFs are designed for low pressure drop taking the soot and ash accumulation over the vehicle lifetime. In an analysis on high-mileage GPFs (47), no significant increase in fuel consumption was seen after 130,000 miles of vehicle ageing, despite the approximately two-fold increase in pressure drop due to accumulated ash. Meanwhile, the nature and role of ash is getting increasing attention. Generally, the ash amounts are much lower than accumulated by filters in diesels.

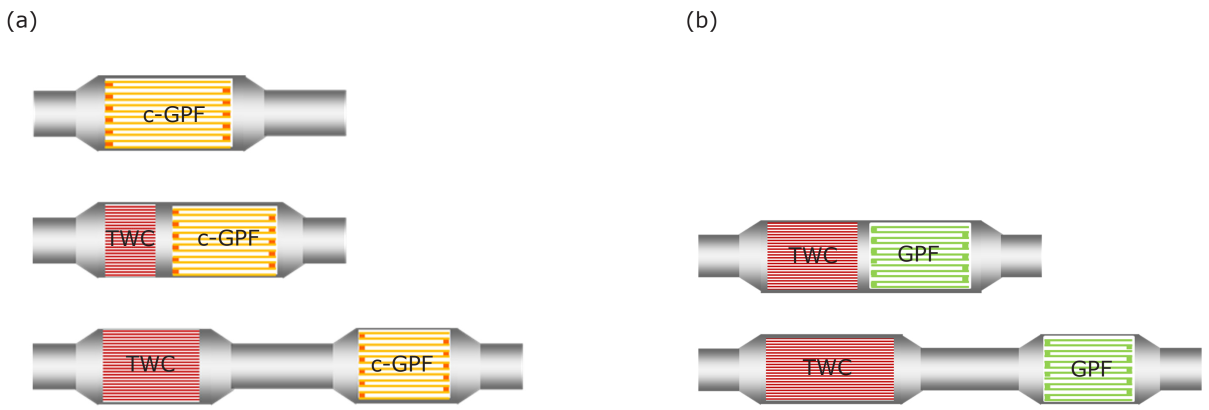

Fig. 3. Light-duty gasoline system architecture with GPF. Various options shown, broadly classified into: (a) TWC-coated c-GPF and (b) uncoated GPF systems for filtration only (46)

Lambert *et al*. (47, 48) reported 0.95–1.62 g of ash in the first 3000 km under real and random driving conditions and 58–61 g after 150,000 miles. This translates to ~0.3–0.5 mg km^{-1} in the low mileage conditions and ~0.25 mg km^{-1} over the lifetime. Even a small amount of ash accumulation on the channel walls helps increase filtration efficiency for GPFs, although with a pressure drop penalty, as already known from DPF experience. In the study mentioned above, an increase in filtration efficiency from ~60% to 80%, that is an increase of ~20%, associated with a pressure drop penalty of 20–22%. Studies are also converging on the source and ultimate location of the ash within the filter. The study found 50% of ash derived from engine oil consumption, the rest related to possible engine wear and some washcoat loss from upstream TWC. An ash accumulation study (49) on an underfloor GPF after 150,000 miles on a 3.5 l GDI vehicle also showed that only 50% of ash was collected from engine oil sources. After accumulating 150,000 miles on vehicles, ash was distributed ~60% on channel walls and 40% as plugs.

Accumulation of soot leads to an increase in pressure drop, so the periodic regeneration of filters *via* soot oxidation is important. The possibility of active regeneration is an area still being explored. Under conditions of high engine out soot and low temperatures (for instance underfloor GPF location), active strategies may be required. However, in other cases, passive regeneration could be sufficient, given high exhaust temperatures typical of gasoline engines coupled with the low soot accumulation rates (compared to diesel). One study (50) has shown that soot oxidation can occur passively through fuel-cut events, characterised by high O_2 concentration and low flow rate, and that an equilibrium is reached such that incoming soot from the engine is balanced by the soot that is oxidised.

Overall, GPFs are now a robust technology and are expected to be prevalent in upcoming gasoline vehicle aftertreatment systems. Other than their benefits in capturing particulates, GPFs also help reduce polyaromatic hydrocarbons (PAH), several of which are known carcinogens. In an analysis (51) of the exhaust plumes of seven different GDI vehicles under idle, cruise and accelerating conditions, aromatic compounds using the benzene, toluene, ethylbenzene and xylene (BTEX) proxy were found in the 62–96th percentile (higher in winter) of all vehicles on the road. Similarly, a 2014 GDI pick-up was found to have fourteen times more total PM-bound PAH emissions than a similarly powered PFI truck, and approximately four times more gas-phase PAH emissions (52). Through a study on three Euro 5 GDI vehicles on the WLTC (53), it is surmised that PAH are likely associated with soot, as uncoated GPFs placed after the TWC were found to reduce PAH by up to 90% for the larger species; and 98% for benzo[α]pyrene, a five-ring known carcinogen. Smaller two- and three-ring PAH are reduced 40–60% with the uncoated GPF. Looking ahead, Europe would like to continue to revise the conformity factors downwards as measurement accuracy improves, and include particles below 23 nm in the PN measurements considering health impacts of fine particles (54). Studies show the latter looks feasible, and that GPFs are effective in capturing these sub-23 nm particles, given that filtration improves with Brownian motion of smaller particle sizes. In an investigation of particulate emissions from five GDI vehicles (55), high PN emissions (~6×10^{12} km^{-1}) were recorded when sub-23 nm particles were included, and GPFs were found to be effective in near total capture of these small particles. Another study (56) on a 1.4 l Euro 6b GDI engine under real-world driving conditions found an increase in engine out PN of >50% when particles >7 nm and <23 nm were included, while tailpipe emissions after a catalysed GPF increased by only ~20%. All tailpipe emissions with the use of GPF comfortably met the limit of 6×10^{11} km^{-1} even with the inclusion of particles below 23 nm.

4. Lean-Burn Gasoline NOx Control

Lean-burn gasoline engine technology has the potential to improve fuel economy by 5–15% compared with a stoichiometric baseline. TWCs are ineffective to convert the high NOx generated under lean (λ>1) conditions and as such lean gasoline requires additional deNOx aftertreatment. Similarly to diesel, NOx could be treated using active SCR, but the additional cost and urea consumption would negate some of the fuel efficiency improvements. Passive SCR is one solution, wherein an upstream TWC generates ammonia under periodic short rich conditions, which is utilised by a downstream SCR catalyst to reduce the previously stored NOx. Studies focus on catalyst formulations and rich-lean modulation conditions for generating sufficient NH_3 for deNOx, without incurring a fuel penalty due to excessive NH_3 slip. One study (57) found an NH_3:NOx ratio of 1.13 at the TWC outlet optimum for achieving 99.5% NOx conversion, while also delivering 11.5% reduction in fuel consumption. Reduced cycle time was found to be beneficial to reduce NH_3 oxidation during lean periods. The oxidation history of the catalyst was also shown to strongly affect the NOx reduction performance (58). Rh was identified as the key catalyst for NO reduction and also responsible for the hysteresis between the oxidised and reduced states in the catalyst. Future challenges for LNTs include HC and CO slip and sulfur tolerance (59).

A novel pgm-free underfloor NOx catalyst was developed (60) using a copper/zeolite upper layer and a nickel/CeO_2 bottom layer. The former helps with SCR-based conversion of NOx while the latter helps with reduction of NO using exhaust CO. The NH_3 required for SCR conversion is generated *via* the upstream TWC, which is a trilayer formulation of Pd/Rh/Pd. On a 2.4 l naturally aspirated PZEV engine, the new underfloor catalyst converted 85% NOx on the LA4 test and 40% on the US06 test.

Despite the efficient combustion of lean gasoline engines, GPFs will likely be needed to meet the particulate regulations. Particulate emissions remain high in both the lean stratified and lean homogenous combustion modes (61). The modulated rich periods required to make ammonia for passive NOx control make the particulate emissions even worse. GPFs are very effective, capturing particulates with >95% filtration efficiency even with the highly transient nature of this combustion strategy.

As major regions continue to reduce the allowable tailpipe CO_2 emissions, lean-burn gasoline engines may provide one approach to meet these future regulations. Ricardo has analysed potential pathways for achieving further reductions in tailpipe NOx and CO_2 from gasoline vehicles, beyond those mandated in the latest Euro 6 and Tier 3 regulations (6). With a view to achieving improvement in fuel economy, lean stratified combustion was considered, and several aftertreatment system choices were simulated. The study concludes that for a C-segment vehicle, a twin LNT system and a GPF is the most cost-effective way of keeping tailpipe NOx at 60 mg km^{-1} (40 mg km^{-1} + CF = 1.5). Exhaust temperatures are predicted to decrease with efficient combustion, thus challenging the catalysts to convert CO and HC effectively. Also, the challenge of meeting the N_2O regulations in the USA and China is highlighted.

5. Hybrid Vehicles

Powertrain electrification is proceeding rapidly due to the improved fuel economy it offers. Accordingly, there is also more attention being given to the real-world driving emissions from various levels of hybridisation of the powertrain. While hybrids emit less pollutants due to the reduced fuel usage, there are also some unique challenges which need to be addressed for meeting the stringent gas and particulate emission standards. One key issue is the lower exhaust temperatures and catalyst cool-down which occurs when the engine is off and the energy demand is being met by the battery. Much has been said earlier about the cold-start problem, and this is in some ways exacerbated in the case of hybrids, where temperatures can remain near or below light-off even during cruising conditions. The actual decrease in temperature and catalyst effectiveness depends on the driving conditions and battery SOC. One study analysed fuel consumption, CO and NOx emissions as a function of the SOC and engine-off duration, using the Toyota Prius full hybrid vehicle (62). For engine-off periods longer than 30 s, the CO emissions index (EI, emissions divided by fuel consumption) increased up to 63%, while NOx EI increased up to 73%, likely due to catalyst cooling after engine-off and the resulting loss in its efficiency.

As with gaseous emissions, there is evidence that particulate emissions for hybrids can be equivalent or even higher than vehicles without any electrification. In a study in Japan (63), particulate emissions were measured on the JC08 test cycle from several gasoline and diesel vehicles, including one GDI and one PFI hybrid. As has been described earlier, most of the particulate emissions for GDI and PFI engines were found to occur during 100–200 seconds after a cold start, with almost no emissions after the engine warmed up. However, in the case of hybrids, emissions were found to occur even after engine warm-up and through the entire test cycle. Overall, emissions from hybrid PFI vehicles were found to exceed those from diesels fitted with DPFs. The issue here is the emission of particulates associated with transient engine turn-on events which occur throughout the drive cycle. Another study used a GDI engine to simulate hybrid electric vehicle (HEV) operation by stopping and starting the engine during NEDC testing (64). Although the engine only operated 28% of the time during the cycle, the PN emissions were 4.5 times higher than when the engine was run in conventional mode.

The above problem is especially acute with PHEVs, where 'high-powered cold-starts' have been shown to greatly increase the particulate emissions (65). This is the scenario when the engine turns on for the first time when the vehicle is already moving at a high speed or under high load, and results in cold-start emissions which are larger than they would be under normal cold-starts. A recent study focused on this issue and measured real-world emissions from a 1.5 l GDI PHEV certified to Euro 6b standards, and driven with various levels of charge on the battery (66). As expected, the PN emissions in urban conditions were totally eliminated when the battery was fully charged as the battery alone propelled the vehicle. Under charging mode though (empty battery), the demand on the IC engine was high and PN emissions were the highest. For the total RDE drive cycle though, for all cases with various levels of charge including a 100% charged battery, the PN emissions were lowest for the reference GDI conventional vehicle with a GPF. For the fully charged battery, there were no cold-start emissions in the urban part, but the emissions were much higher when the engine turned on later along with a high power demand. Hybrids and their power management strategies are evolving and clearly there is room to improve on some of the above shortcomings. In the meantime robust aftertreatment solutions are seen to be necessary to cover the operation of hybrids under widely varying real-world conditions. This would likely involve some of the advanced emission control solutions mentioned through the paper: cold start strategies, advanced substrates and catalysts which promote early heat-up and emissions conversion, improved heat retention to avoid rapid cool-down when engine is turned off, and the use of filters to capture particulates during regular engine operation and transients associated with the switching from battery to engine.

6. Summary

In response to mandates of lower fuel consumption worldwide, the IC engine continues to evolve and deliver impressive gains in fuel efficiency. Along with powertrain electrification, advanced gasoline vehicle technologies are being adopted rapidly, at lower cost points, and are seen as important tools to lower GHG emissions. New engine and vehicle technologies also present new challenges to the aftertreatment systems: efficient combustion and hybridisation lead to reduced exhaust temperatures, producing further demands on the catalyst to work more efficiently at lower temperatures. And regulations continue to tighten globally, such that new vehicles will be required to reduce criteria pollutants to near-zero levels, and under real-world driving conditions. Finally, gasoline vehicles are receiving attention for their particulate emissions: there is a growing body of knowledge that gasoline vehicles can produce more particulates than diesels equipped with DPFs (67). In response to these challenges, progress continues on all fronts: engine improvements, fuels and aftertreatment systems. Advanced catalysts, substrates, improved packaging and gasoline particulate filters are being developed and implemented, and are well poised to mitigate vehicular pollution.

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Abbreviations

BMEP	brake mean effective pressure
BS	Bharat Stage
BSFC	brake specific fuel consumption
BTE	brake thermal efficiency
BTEX	benzene, toluene, ethylbenzene and xylene
CARB	California Air Resources Board
c-EGR	cooled exhaust gas recirculation
CF	conformity factor
CO	carbon monoxide
CO_2	carbon dioxide
CR	compression ratio
DPF	diesel particulate filter
EI	emissions index

EPA	Environmental Protection Agency
EUDC	Extra Urban Driving Cycle
EV	electric vehicle
FCNT	fuel-cut NOx trap
FTP	Federal Test Procedure
GDCI	gasoline direct injection compression ignition
GDI	gasoline direct injection
GHG	greenhouse gas
GPF	gasoline particulate filter
HC	hydrocarbons
HCCI	homogeneous charge compression ignition
HD	heavy duty
HwFET	Highway Fuel Economy Test
IC	internal combustion
LA4	Los Angeles Route Four
LD	light-duty
LEV	Low-Emission Vehicle
LNT	lean NOx trap
LTGCI	low-temperature gasoline compression ignition
NA	naturally aspirated
NEDC	New European Driving Cycle
NHTSA	National Highway Traffic Safety Administration
NMHC	non-methane hydrocarbon
NMOG	non-methane organic gas
NOx	nitrogen oxides
OEM	original equipment manufacturer
OSC	oxygen storage capacity
PAH	polyaromatic hydrocarbons
PEMS	portable emissions monitoring system
PFI	port fuel injected
pgm	platinum group metal
PHEV	plug-in hybrid electric vehicle
PM	particulate matter
PMI	particulate matter index
PN	particulate number
PZEV	partial zero emissions vehicle
RDE	real-world driving emission
SCR	selective catalytic reduction
SFTP	Supplemental Federal Test Procedure
SOC	state of charge
SULEV	super ultra-low emissions vehicle
THC	total hydrocarbon
TWC	three-way catalysts
UC	unified cycle
ULEV	ultra-low emissions vehicle
VCR	variable compression ratio
WLTP	World Harmonised Light Vehicle Test Procedure

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