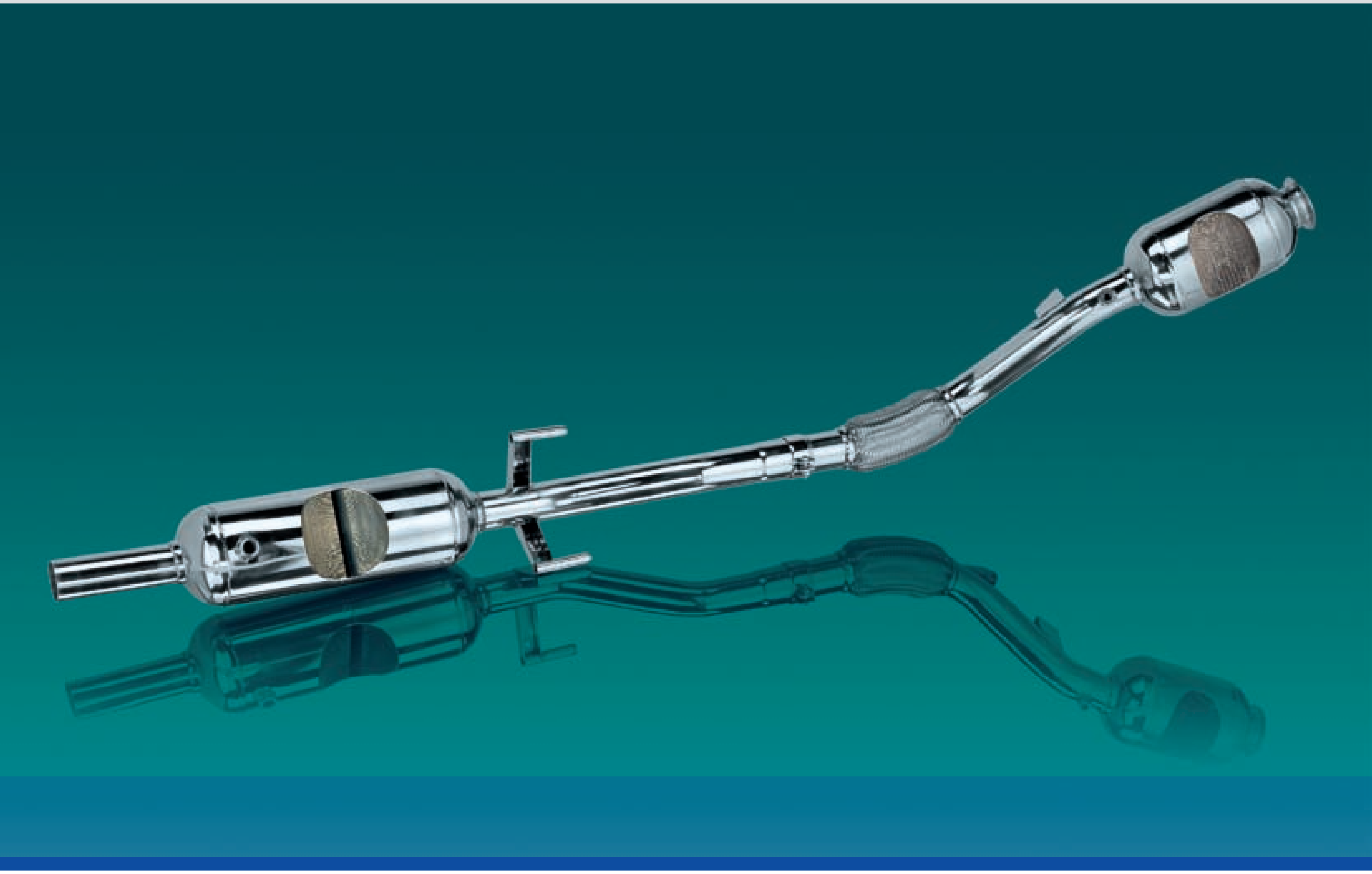

The SCRi Concept for Exhaust Gas Aftertreatment

Integrated PM Reduction in an Urea-SCR System

Particulate Matter (PM) reduction is an important step on the way to ecologically friendly diesel engines. Emitec established for exhaust gas aftertreatment the non-clogging PM-Metalit soot filter in series production in the year 2004. This filter, integrated into the Selective Catalytic Reduction (SCR) system, is the basis of the new SCRi concept that reduces particles as well as nitrogen oxides (NO_x).

1 Introduction

Tighter global emission ceilings for NO_x and PM call for extensive reduction of emissions within the engine for passenger cars, commercial vehicles and off-road applications in the future. In addition, exhaust gas aftertreatment is required. Only with this complex and costly package of measures it is possible to remain below the low NO_x and PM limits for diesel engines in the next ten years. Under EU 4/5 NO_x reduction takes place in cars within the engine, with a diesel particulate filter (DPF) requiring active regeneration periodically being fitted as standard. For commercial vehicles the system approach predominantly selected for emissions reduction in Europe, however, is engine-internal PM reduction and NO_x reduction in the exhaust gas using the SCR system. On the other hand, two of the major European manufacturers of commercial vehicles predominantly use Exhaust Gas Recirculation (EGR) engines in order to lower NO_x emissions by engine-internal measures. Partial-flow soot filters for continuous PM reduction have proven extremely successful in this case. The SCR system reduces nitrogen oxides by a high percentage by injecting aqueous urea solution (Adblue) into the exhaust gas system on the downstream reduction catalyst. The reduction catalyst was initially introduced in commercial vehicles as a full extrudate. In place of those coated catalyst substrates are increasingly used. In systems currently being developed for increased targets of durability and robustness, including those for the off-road sector, metal substrates are being favoured. The SCRi system is the outcome of the integration of EU IV and EU V systems that have proven themselves in series applications for combined PM and NO_x reduction for compliance with US2010/EU VI and for retrofitting.

2 PM-Metalit Filter Systems in Commercial Vehicles for EU IV

The development of PM-Metalit filter systems for exhaust gas aftertreatment in EGR engines has already been described [1]. Structure, reaction pathways and reaction equations of a continuous-operation soot filter system of this type for commercial vehicles are shown in **Figure 1**. In a nitrogen monoxide (NO) oxidation catalyst V upstream of the soot filter P, the nitrogen dioxide (NO_2) proportion of the NO_2 is increased from 2 % to 3 % to 20 % to 30 %.

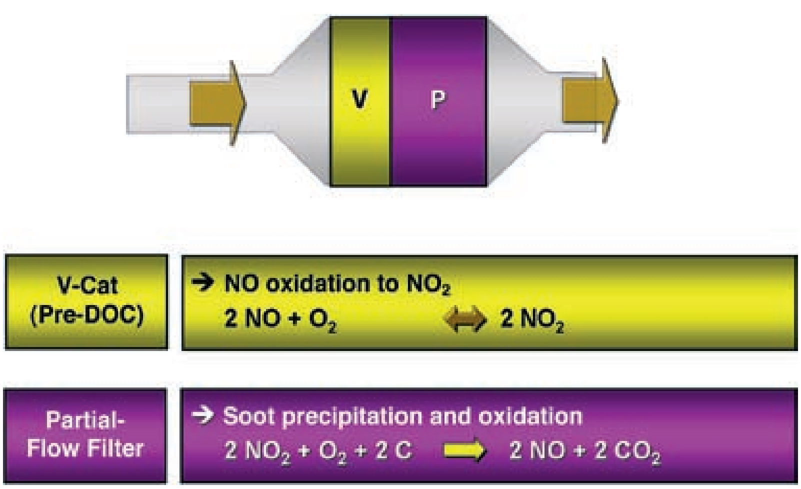

Figure 1: Functional scheme and operating principle of a soot filter system for commercial vehicles

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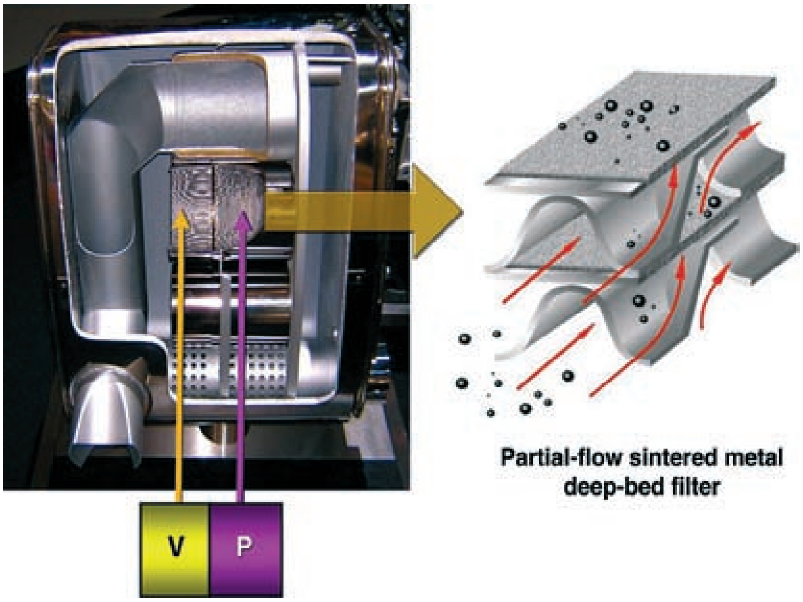

Figure 2: Soot filter catalyst system in the main silencer of a EU IV commercial vehicle from MAN [4]

Continuous soot oxidation takes place in the filter due to the NO_2 generated and the oxygen present in the exhaust gas, consuming the generated NO_x again. As far as the use of particulate filters in commercial vehicles is concerned, the focus is especially on service intervals and fuel consumption. Ideally, a filter should operate over its entire service life (1 million km) without any need for maintenance and avoid any additional fuel consumption for active regeneration and increasing the purging action of the engine. Compared with passenger cars, a commercial vehicle engine has, in the majority of operating conditions, a stoichiometric NO_x/soot ratio which is favourable to continuous regeneration. In order to fulfil the 'maintenance free' requirement, a commercial vehicle filter system should as far as possible not be optimized for the filtration of engine oil ashes (approximately 90 % gypsum) but should instead focus on the elimination of soot, in particular soot particles smaller than 100 nm [1]. The Emitec PM-Metalit soot filter meets these requirements and uses the available NO_2 concentrations better by a factor of 2 to 4 than a traditional wall flow filter [2]. Due to the partial-flow deep bed filter effect, it reduces, depending on its length, the particulate mass and numbers up to more than 80 % [3]. At MAN Nutzfahrzeuge this filter system was being fitted at the end of 2004 as standard in the EU IV vehicles. In this system, oxidation catalysts V were integrated upstream of the soot filter P in the silencer with EU III dimensions, **Figure 2**. This eliminated the need for additional space to be made, for example, at the expense of the size of the fuel tank and the low weight made it possible to maintain the potential payload. To date, more than 120,000 MAN EU IV vehicles have been sold and achieved mileages of more than 800,000 km per truck. Studies carried out on individual vehicles with high kilometer readings confirm the results obtained in the development and test phases. The PM catalytic converter developed by MAN Nutzfahrzeuge fulfills the requirements in terms of continuous particulate reduction and low exhaust back pressure and represents a maintenance-free filter component that is reliable in operation. PM-Metalit soot filters are also used by a number of manufacturers of HGVs and buses in order to comply with EU IV, V or with the Enhanced Environmentally Friendly Vehicle (EEV) standard.

3 SCR Catalyst Systems in Commercial Vehicles

3.1 The MAN Adblue System for Advance Compliance with EU V [4]

Among the SCR catalytic converter systems for reducing NO_x in European commercial vehicles, the MAN Adblue System for compliance with EU V stands out due to the fact that it is compact. **Figure 3** illustrates the structural principal and the chemical processes in this three-stage system. The urea decomposition catalytic converter (H-cat), arranged in a first stage silencing chamber close-coupled to the engine, is a multi-function reactor operating in the partial flow which combines evaporation of water, catalytically assisted thermolysis of the urea into isocyanic acid (HNCO) and ammonia (NH_3) and cata-

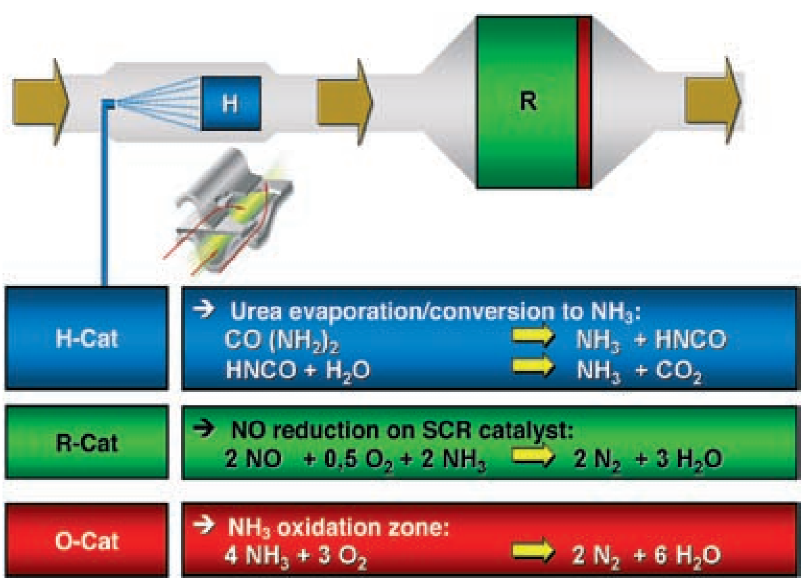

Figure 3: Functional scheme and reaction pathways: SCR catalyst system

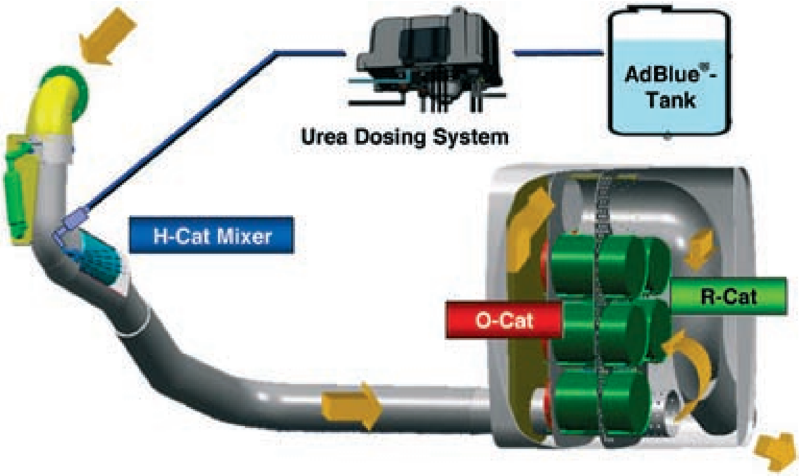

Figure 4: MAN Adblue system as preferred EU V solution

lytic hydrolysis of HNCO into NH_3 and carbon dioxide (CO_2) with homogenization of the reduction agent. This is achieved by means of titanium dioxide (TiO_2)-coated, 'turbulent' catalyst substrates with staged cell thickness [5]. The R-cats reducing NO_x (97 % NO, 3 % NO_2) consist of extruded homogenous catalysts of the $V_2O_5/WO_3/TiO_2$ type (vanadium oxide/tungsten oxide/titanium dioxide). A further characteristic of the system is a zone at the end of the R-cat, approximately 10 mm wide and impregnated with platinum, which provides protection against ammonia slip during dynamic processes. **Figure 4** shows the design of the system in relation to the vehicle. The interconnect piping between the first stage silencing chamber and the main silencer ensures, due to the length and by means of two 90° elbows, that the NH_3 is homogeneously mixed with the exhaust gas prior to entering the plane-parallel channels of the R-cat. In this way the R-cat volume requirement has been significantly reduced so far that it has been possible to accommodate it within the silencer volume used already in the EU III and IV vehicles. The EU V vehicles with the SCR system have proven their viability as well. In particular, unlike with some other Adblue technology applications introduced in the market, none of the corrosion phenomena were observed.

3.2 Further Development of SCR Systems

An objective of further development is downsizing the catalysts for exhaust gas aftertreatment. Optimum NO_2 formation as well as a reduction in NO_x together with reduced volume dimensions can be obtained by means of the turbulence-generating longitudinal structures (LS) of the Metalits. This makes building of significantly smaller exhaust gas aftertreatment units possible. The increase in volume-specific reduction activity due to the transition from smooth channel extrudate catalysts (300 cpsi) to 'turbulent' catalysts with LS/PE 300/600 substrates has been demonstrated in a particularly spectacular way with SCR catalysts: despite a 35 % reduction in length, a similar NO_x conversion rate is achieved, with reduced back pressure and ammonia slip as well [4,6,7,8,13]. Another option for increasing the activity of SCR catalysts in low NO_x-engines is offered by the use of metal-exchanged zeolite catalysts. The specific surface of these catalysts can be extended into the range of 500 m^2/g. In this way two activity-enhancing effects can be combined:

1) acceleration of the chemical kinetics by increasing the number of active centers
2) reduction in the layer thickness in order to accelerate pore diffusion.

This permits the use of high cell density substrates (for example 800 cpsi). Acceleration of pore diffusion can also be achieved by using mesoporous zeolites. The extent to which Cu zeolites are used depends on the potential for enhancing their currently poor product selectivity (avoidance of nitrous oxide formation) [9,14].

4 The SCRi Concept

4.1 Application in Commercial Vehicles for EU VI and US2010

4.1.1 Combining Systems for PM and NO_x Reduction

The emission limits of the future EU VI regulation, which are announced to be defined in 2009, are expected to be quite similar to the US2010 values. The current proposal from the European Commission confirms this assumption. Similar thres-

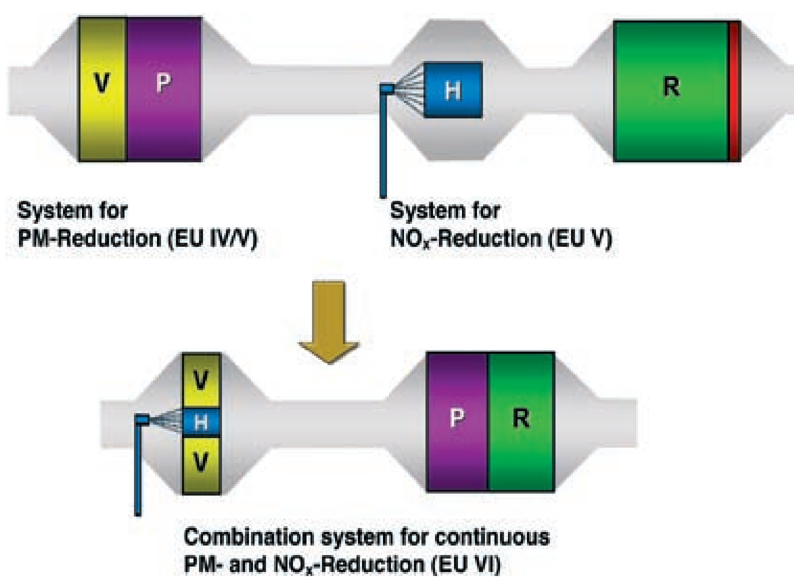

Figure 5: Combination of EU IV and V systems that have proven themselves in serial production

holds will also be in force for the off-road sector, by 2014 at the latest. The technologies available for emissions control in this context include extensive engine-internal NO_x- and PM reduction and the use of combined exhaust gas aftertreatment for PM and NO_x. One of the technologies being considered for combined exhaust gas aftertreatment is the low NO_x/high PM-EGR engine with active-regeneration soot filter, as introduced in North America to meet the US2007 emissions regulation, combined with a downstream SCR system. In addition to the considerable space requirements and weight, this combination also brings a number of operational disadvantages, such as higher fuel consumption for the (frequent) regeneration of the filter, the high thermal load on the downstream SCR catalyst during regeneration and the challenges for control of Adblue dosing under the different loading conditions of the periodic-operation filter and especially during regeneration events. An alternative approach therefore suggests itself: a series arrangement of the EU IV and V systems that have proved themselves in serial application for PM and NO_x reduction. This kind of combination is shown in the upper part of **Figure 5**. Since it does not seem possible to realize the space requirements even for this kind of system in a commercial vehicle without reducing the fuel tank volume, integration of the PM filter into the SCR module has been developed, as shown in the bottom part of Figure 5. This offers a number of advantages and improves the functionality of the SCR system. With the help of engine test bench investigations [4,5] it has been shown that the continuous soot oxidation by means of NO_2 (measured using Photoacoustic Soot Sensor, PASS) and the simultaneous production and presence of ammonia from the injected urea (thermolysis/hydrolysis, measured using Fourier transform spectroscopy, FTIR) do not negatively impact each other. The position of the PM-Metalit for soot reduction upstream of the NO_x reduction catalyst is ideal. Model gas tests were conducted to obtain more detailed information about the reaction of soot with NO_2 in the presence of ammonia. Soot was collected with a PM-Metalit on the engine test bench downstream of the oxidation catalyst. The soot-loaded PM-Metalit was installed in a model gas test bench at temperatures of 200 °C, 250 °C and 300 °C at a space velocity of 35,000 h^{-1}, where soot reacted with NO_2 and, as a comparison, with a mixture of NO_2 and NH_3. The results are shown in **Figure 6**. The presence of ammonia results in a soot oxidation rate that is almost 30 % higher at 200 °C, identical at 250 °C and approximately 20 % lower at 300 °C. The acceleration of the soot oxidation at 200 °C by ammonia is attributed to the intermediary formation of ammonium nitrate. Ammonium nitrate partially dissociates into ammonia and nitric acid at 200 °C. Nitric acid produces higher soot oxidation rates than NO_2. Surprisingly, during these experiments, an SCR effect in the 20 % range was observed on the soot-coated PM-Metalit at 250 °C and 300 °C [10], which explains the decline in the soot oxidation rate at 300 °C through a reduction in the NO_2 concentration. On the engine test bench it has since been possible to confirm the SCR activity of soot deposited on various models of particulate filters in the presence of NO_2 with conversion rates of 20 % to 30 % [11].

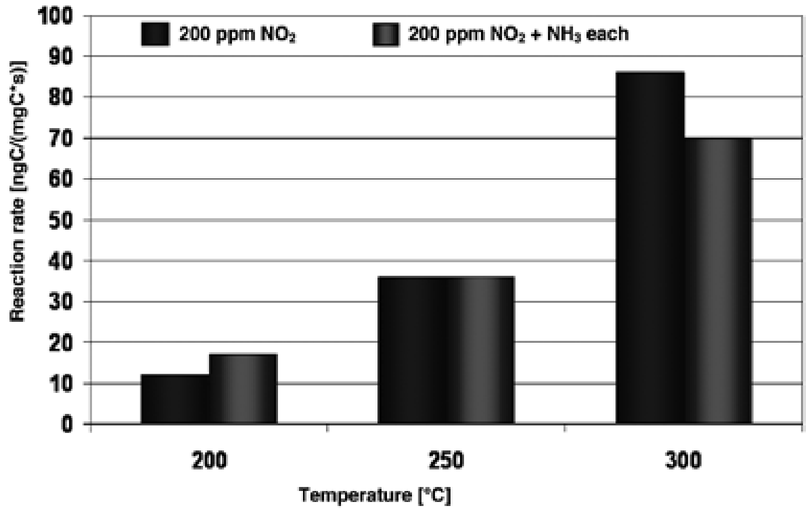

Figure 6: Soot oxidation with NO_2 in exhaust gas containing NH_3

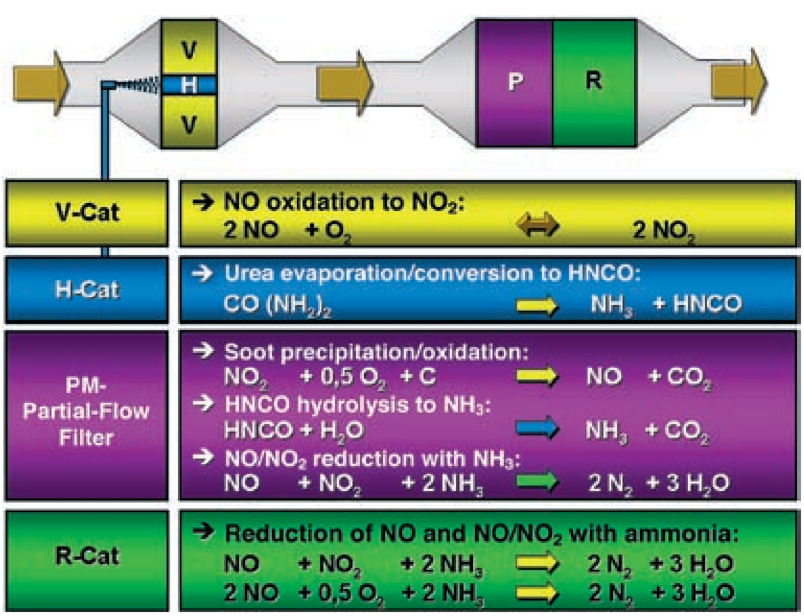

Figure 7: Functional scheme and reaction pathways: SCRi system

Figure 7 shows the reaction paths and equations for the different stages of the SCRi system. The first stage silencing chamber closed-coupled to the engine primarily houses a pre-oxidation catalyst (V-cat) for NO_2 production and a parallel urea-decomposition catalyst (H-cat), which is very small by comparison. This catalyst gasifies the Adblue spray through water evaporation and catalytic thermolysis of the urea into NH_3 and HNCO. NO_2 is the key substance for the continuous soot oxidation and for the NO/NO_2 SCR reaction. Both reactions are known to run best when a sufficient high NO_2 fraction has been produced from the engine NO by an oxidation catalyst (ideally around 50 %). This enables the deposited soot to be reduced to the extent necessary, whilst at the same time allowing the NO/NO_2 SCR reaction, which is particularly necessary in cold operating conditions, to take place. With a cost-effective, thin hydrolysis coating of the PM-Metalit, it is possible to achieve conversion of the urea thermolysis product HNCO into ammonia. The PM-Metalit with its many flow direction changes and porous fleece layers between the individual channels is also ideal for blending the multi-component flow of HNCO, ammonia and engine exhaust gas. This ensures better utilization of the reduction catalyst, as the frontal section is otherwise required for urea decomposition, which consequently means that it is bigger than would actually be necessary. The soot oxidation enabled by NO_2 in the presence of ammonia also explains a soot-reduction effect previously observed during NO_x reduction in a VHRO system (with “Vor” pre oxidation-, Hydrolysis, SCR- and post oxidation catalyst). The observed soot deposition and soot oxidation must have occurred on the BQ 150 full-flow H-cat with turbulent substrate (predecessor of the LS substrate) [12].

4.1.2 Engine Test Bench Results [4,13]

The exhaust gas is delivered by a two stage turbo-charged low NO_x-engine (~2 g NO_x/kWh in European Steady Cycle, ESC, and European Transient Cycle, ETC), with exhaust gas temperatures predominantly in the 200 °C to 400 °C range. The design of the SCRi catalyst system is similar to a vehicle design shown in **Figure 8**. In the main silencer there are five PM-

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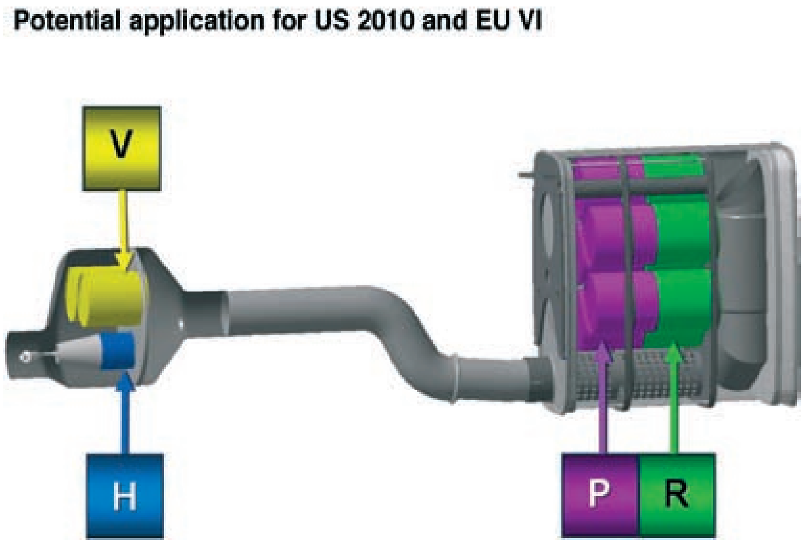

Figure 8: Commercial vehicle model of an SCRi system

Metalits P arranged in parallel (diameter: 150 mm, length: 145 mm) followed by five SCR catalysts R ($V_2O_5/WO_3/TiO_2$ coating on Emitec LS/PE 300/600 substrates, diameter and length both 150 mm). The NO_x reduction achieved in the ESC and ETC was a little over 80 %, meaning that values of around 0.4 g/kWh NO_x at < 10ppm NH_3 slip were achieved with no special optimization of the Adblue dosing algorithm and with no engine thermal management yet. The soot emission, measured using PASS, was reduced to less than 10 mg/kWh with the PM-Metalit.

4.2 Application in Retrofitting

Since 2006, in the European Union stringent ambient air quality requirements for fine particulate matter apply. Many cities exceed the maximum allowed days of violating the ultra-fine PM level (max. 5µg/m³ daily average for PM 10) at present and are required to implement action plans to reduce the ambient ultra-fine PM. Besides other restrictions defining environmental zones limiting the entry to “clean” vehicles meeting the recent emission standards and to vehicles retrofitted with diesel particulate filters will be enforced beginning of next year. In 2010, in addition to the requirement on ultra-fine particles a stringent limit on NO_2 in the ambient air will be introduced in the EU. Similar to exceeding the ultra-fine PM level, many cities and regions will than be in violation of the NO_2 requirement as well, and this will consequently call for retrofitting vehicles with combined NO_x and PM reduction technology. Retrofitting is the only way to reduce the immission related to vehicles short-term addressing the emissions of the existing vehicle fleets that will be in use for many more years. The continuous SCRi principle is particularly suitable for retrofitting. The compactness and continuous operation of SCRi systems significantly reduces the work involved in retrofitting combination systems. A retrofit-system according to the SCRi-principle is shown in **Figure 9**. This retrofit system, here shown for a light-duty delivery van as exhibited for example at the IAA 2007-convention in Frankfurt, is becoming commercially available at the end of 2008. In the first stage NO_2 is generated and optionally the PM emission can be reduced to some extend if applying a coated PM-Metalit. Stage 2 features a PM-Metalit with a thin TiO_2 coating, which performs the evaporation and catalytic thermolysis of Adblue, and the required PM reduction as well. Stage 3 is the SCR catalyst with a platinum zone coating at the rear end (zone-coated ammonia slip catalyst).

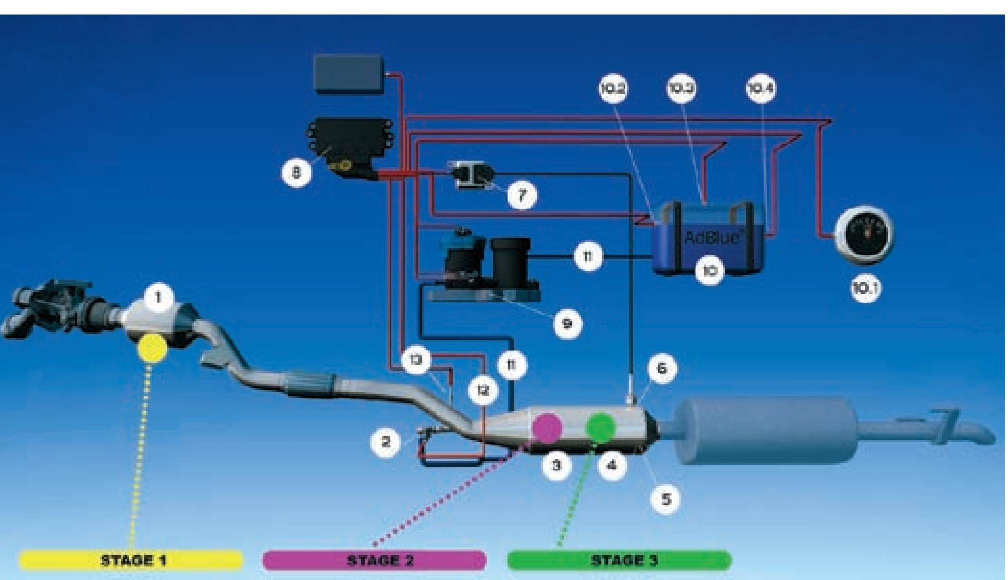

Figure 9: Exhaust gas aftertreatment from Twintec for retrofitting using the SCRi principle:
1 Catalyzed PM metalit filter or oxidation catalyst, 2 Injection of Adblue, 3 PM Metalit coated with TiO_2, 4 SCR catalyst, 5 NH_3 slip catalyst (optional), 6 Smart NO_x sensor, 7 NO_x control unit, 8 Adblue-injection control unit, 9 Adblue pump, 10 Adblue tank, 10.1 Level indicator, 10.2 Temperature measurement, 10.3 Tank heating, 10.4 Level measurement, 11 Adblue line (heated), 12 Injection control cable, 13 Temperature sensor

This system, complemented for example by advancing the injection timing of the engine, has the potential to allow trucks originally meeting only the EU III emission standard to be retrofitted for the EU V or EEV-level.

5 Summary

For the US 2010 and EU VI emission ceilings, a combination of particulate filter and SCR will be necessary. Besides the familiar SCRT concept, a new, particularly compact system deserves a mention – the SCRi system, which combines a PM-Metalit with an SCR substrate fitted immediately downstream. The special feature of this system is that the urea is injected upstream of the PM-Metalit (partial-flow deep-bed filter). This means that the compact design can be realized with no loss of pressure or heat between the two components, providing good conditions for fitting the system in the main silencer. Another advantage of this arrangement is the mixing function of the PM-Metalit, which ensures optimum utilization of the reduction catalyst. The continuous regeneration means that the higher thermal load observed on the reduction catalyst during thermal regeneration in a DPF (which also involves higher fuel consumption) does not occur. The SCRi system is being tested for OEM serial application and is also intended for retrofitting in commercial vehicles and passenger cars as a combined solution for simultaneous nitrogen oxide and particulate reduction.

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