

21st Century Emissions Technology

REVIEW OF AN INSTITUTION OF MECHANICAL ENGINEERS CONFERENCE ON EMISSION CONTROL

The Combustion Engines Group of the Institution of Mechanical Engineers (IMEchE) held a conference on '21st Century Emissions Technology' at their London headquarters on the 4th and 5th December 2000. Nearly 100 participants from the U.S.A., Japan and most European countries gathered to hear 23 excellent talks and to discuss engine and aftertreatment technologies.

The major theme of the conference was the need for ongoing research in emissions reduction strategies and technologies for both on-highway transport and off-road mobile machinery. The conference was thus a forum for the leading experts from motor manufacturers, oil companies, vehicle testing houses and universities to discuss the reduction of particulate matter and nitrogen oxides, NO_x, in order to meet the forthcoming stringent European and U.S. federal emissions legislation. Vehicle emissions can form a large part of the measured pollutant levels in ambient air, thus, this review examines the role that catalyst systems, based on platinum group metals (PGMs) play in ensuring the continual global lowering of vehicle emissions.

Emissions Control Technology for 2000–2010

Robert Searles (AECC, Belgium) addressed the challenges to and opportunities for catalyst technology in the 21st century. He highlighted the need for cleaner fuels, with lowered sulfur levels, which enhance and make possible the use of more sophisticated catalyst systems, such as NO_x traps and particulate filters. He mentioned the launch of the new Peugeot 607 diesel passenger car, which is an industry first for a European diesel passenger car to have a particulate filter system. There was speculation about whether a strategy of using non-continuous regeneration with a fuel-borne catalyst, and an appropriate conventional platinum-based catalytic converter would be a realistic way forward for all classes of passenger cars. He reported growing interest in the selective catalytic reduction

(SCR) of NO_x for use in the aftertreatment of heavy-duty diesel emissions. Searles also summarised work on hydrocarbon-absorbers, electrically heated catalysts, and on active and passive NO_x.

The technologies currently available for passenger cars and larger vehicles were reviewed by Timothy Johnson (Corning, U.S.A.). He discussed developments in various catalyst supports (made of cordierite) and trends, such as:

- advanced integration of catalyst and engine management
- lower ammonia emissions from gasoline vehicles
- better cold start response
- hybrid (or layered) catalysts and
- PGM flexibility.

Johnson noted that flexibility in using the PGMs is an important new concept for major automobile manufacturers. This will help to ease cost volatility by substitution of one metal for an equally active one, where possible. This might lead to a need for certification of multiple catalyst systems at the initial design phase and production testing of vehicles, and would ultimately result in cost savings for most parties.

The costs of various SCR systems were assessed by James Warren (The Open University, U.K.) for Euro 5 heavy-duty diesel trucks. It was shown that most long-haul transportation trucks can 'pay back' the complete cost of the catalyst system within one to two years if fuel savings of 3 to 5 per cent are achieved, by tuning the engine for higher, raw (precatalyst) NO_x emissions, and then choosing an appropriate catalyst and control system.

A possible way of reducing NO_x via engine method alone was presented by Patrick Flynn (Cummins Engines, U.S.A.). He described work on exhaust gas recycling (EGR) which clearly indicates that combustion chemistry is the limiting factor in determining the minimum NO_x produced in both gasoline and diesel engines (heavy duty, cylinders > 1 litre). The minimum levels of NO_x achievable are listed in the Table, along with the proposed European and U.S. federal limits. His

Source/Standard	NO _x , g (kW h) ⁻¹
Lowest possible by engine design (SI)	~ 0.56
Lowest for diesel	~ 1.12
Europe 2008 (proposed EU 5)	2.0
U.S.A. 2007 (proposed)	0.15

talk clearly demonstrated that there is a vital need for all types of aftertreatment, especially to comply with the very low U.S. limits. He concluded that even with the best possible engines using EGR, aftertreatment will still be required to meet the 0.15 g (kW h)⁻¹ NO_x limit proposed for the U.S. in 2007.

Another route to lowered NO_x levels is offered by the non-thermal plasma (NTP) technique. Henmann Breitbach (Delphi, Luxembourg) showed that a potential NTP system, coupled with a zeolite-based catalyst, can reduce emissions from passenger cars. Currently, when running the NTP system on a car, ~ 300 W (~ 1 kW at peak condition) is required to generate good dielectric behaviour. This results in a fuel penalty of ~ 3 per cent. However, with the future 42 V electrical systems, coupled with integrated starter motors, NTP in cars will be much easier to integrate and to supply with power.

Diesel Particulate

Diesel particulate and its measurement – down to aerodynamic particle diameters of ~ 7 nm – remains of strong interest to engine manufacturers and, indeed, to the public at large. The use of CRTTM (Continuously Regenerating Trap) systems, containing a platinum-based catalyst and particulate filter, is well established for particulate reductions.

The characterisation of particles from heavy-duty diesel (HDD) and light-duty diesel (passenger cars) was examined by Barbara Wedekind (Ricardo Consulting Engineers, U.K.). She also discussed the effects of a CRTTM on a Euro 2 HDD, as well as the effects of diesel particulate filters on a selection of passenger cars. Work on the Peugeot 607, VW Golf and Peugeot 406, all fitted with a variety

of oxidation catalysts and filters, showed that vehicles fitted with these devices were capable of achieving very low particulate mass emissions. Under all conditions tested, the CRTTM consistently reduces particulate mass. However, further work on the characterisation and measurement of particulate is required both to establish the origin of any particulate observed after filtration, and to find a standard set of measurement conditions on which all test houses can agree, for possible future legislation and correlation.

The post-catalyst exhaust gases from gasoline-powered vehicles (aiming at SULEV (super ultra low emission vehicle) emissions) are so clean as to be almost immeasurable with some of today's devices. Indeed, in some instances the exhaust gas has been cleaner than the intake ambient air. This illustrates the need for more advanced quantitative methods for measuring evaporative hydrocarbons, particulates and other pollutants.

Future Trends in Engines

Compressed natural gas (CNG) and controlled auto-ignition (CAI) were among other engine/fuel powertrains described. Both promise potentially lower NO_x emissions. While gaseous fuels are currently relatively inexpensive, the potential emission of methane from such engines will need to be addressed catalytically. CAI gives NO_x values as low as 10 ppm, but system control is extremely difficult, and CAI has not yet been fully demonstrated in a running vehicle.

Paul Kapus (AVL, Austria) reviewed work on the 'G90' vehicle. The 'G90' is a modified GM/Vauxhall/Opel Corsa with a typical fuel consumption of 5.7 litres/100 km (as purchased). After modifications to body shape (for superior aerodynamic drag) and transmission, and the addition of variable valve actuation, along with partial down-sizing, the fuel consumption was reduced to 3.9 litres/100 km and CO₂ emissions were lowered from 137 to 90 g km⁻¹. This shows that the way forward to enable engines to meet CO₂ limits will be a smart combination of technologies.

The views of Toyota on hybrid powertrains, were presented by Tokuta Inoue (Toyota and Genesis Research Institute, Japan). He described

the Prius, Toyota's hybrid electric vehicle (HEV). Currently, the Prius (second generation coupled to a gasoline internal combustion engine) contains a new rectangular battery module (21 kW) coupled to a 33 kW gasoline engine. This delivers 3.4 litres/100 km under the Japanese driving test. When compared to a pure electric vehicle, this HEV has a longer driving range, good fuel economy and good drivability without the need for recharging. The year 2001 Prius meets the Japanese and U.S. SULEV emissions, as well as the Euro 4 regulations. The Prius is the first Toyota vehicle, and perhaps one of the first-ever production vehicles, to use a catalyst support of 900 cells inch⁻², wall thickness 0.5 mm, carrying a palladium and rhodium catalyst. The increase in highly active geometric surface area has been optimised together with an ~ 19 per cent lower total catalyst volume than prior systems. It contains a hydrocarbon absorbing component to assist hydrocarbon capture at low temperatures with subsequent oxidation after light-off is complete.

Future Vehicles and Technologies

It is expected that future vehicles will include more models with direct injection (gasoline and CNG) with advanced variable valve timing. Powertrains will be based more on combinations of technologies, such as turbo-charged CNG, CAI with direct injection, and CNG with direct injection. The market will become segmented with some sectors finding unique solutions (HDD vehi-

cles will remain for trucks/transport of goods). In other areas multiple solutions are possible, with fuel cells and hydrogen-based fuels expected to eventually gain a large part of the market share. Hybridisation and hybrid vehicles will begin to play a prominent part in powertrains, with electric/fuel combinations being prominent. Clearly, the variety of powertrain combinations will have an effect on the catalyst systems they use. Future developments will need to take all component into consideration (filters, catalysts, monitors, sensors, fuels, etc.) and to maximise any synergistic effects to achieve the lowest overall emissions, and the lowest possible CO₂ levels for their class. Vehicle designers and component suppliers will probably work together closely to achieve cost-effective solutions. In general, consumers will increasingly demand comfort, safety, cost reductions, good driving performance, enhanced durability, reliability and low emissions. However, the catalytic converter will clearly remain the main pollution-control device for the next generation of vehicles, although, 15 years from now, it may not be recognisable in its present form.

The papers presented at this conference will be published in Spring 2001 in the IMechE Seminar publication series. Information can be found at: <http://www.imeche.org.uk>.

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Palladium Cross-Coupling Reaction for Brain Tracers

Brain imaging via positron emission tomography (PET) using the short-lived ¹¹C positron emitter (half-life ~ 20 minutes) requires a fast chemical reaction to incorporate the small amounts of ¹¹C into a carrier (tracer) which delivers it to the tissue under examination.

Researchers in Japan and Sweden have now developed a new rapid, efficient Stille methylation of arylstannanes which produces a structure system that can deliver sufficient quantities of ¹¹C to prostaglandin receptors (IP₂) in the brain (M. Suzuki, H. Doi, K. Kato, M. Björkman, B. Långström, Y. Watanabe and R. Noyori, *Tetrahedron*, 2000, 56, (42),

8263–8273). The Stille methylation involves a palladium-promoted cross-coupling reaction of methyl iodide and tributyltin derivatives of tolylisocarbacyclins (TICs); the TIC binds to the IP₂ receptors. In one methylation, which is a novel stepwise operation, an initial methylpalladium complex is produced and is then mixed with other materials for the cross-coupling. The Pd promoter is a Pd(0) complex generated from Pd₂(dba)₃ and P(*o*-CH₃C₆H₄)₃ in the presence of CuCl and K₂CO₃. The synthesis is highly reproducible, and with ¹¹C incorporated, results in a [¹¹C]-labelled PET tracer with radioactivity of several GBq, which can be injected intravenously.