

Controlling the Exhaust Emissions from Alternative Fuel Vehicles

NEW OPPORTUNITY FOR PLATINUM METALS CATALYSTS

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To meet the increasingly exacting emission control standards that are emerging, alternative fuel vehicles will be needed. It appears that these will require optimised and dedicated systems utilising innovative control strategies to achieve the necessary standards. In this paper, the major emission control issues for engines fuelled with alcohol, natural gas, propane, or diesel fuels are identified. Control strategies employing dedicated emission control catalysts are discussed and performance results from laboratory, engine and vehicle tests are highlighted. Comparisons are made with gasoline fuelled engines, where significant experience has been accumulated over the past decade with the use of closed loop fuel control plus a catalytic converter to suppress the three main polluting emissions. The use of catalytic converters containing flow through catalysts is emphasised.

The development of vehicles using clean alternative fuels, such as methanol, ethanol, natural gas, propane, reformulated gasoline and clean diesel, has been hastened by tighter emission standards, such as the United States Federal Clean Air Act Amendments of 1990. As a result of legislative initiatives the regulatory agencies, the automotive industry and the manufacturers of emission control components are assessing the emission characteristics of these vehicles. At the same time, efforts are being made to develop effective and durable emission control systems suitable for vehicle fleets and prototype engines being developed for future automotive markets. Part of this development involves the catalytic converter technology already installed on over 150 million gasoline-powered vehicles to reduce hydrocarbons, carbon monoxide and oxides of nitrogen (1). This same technology offers considerable potential for responding to the emission control challenges associated with alternative fuelled vehicles. This paper discusses the emissions from such vehicles and how they differ from gaso-

line fuelled vehicles; it highlights the significant control issues and summarises recent developments in emission control catalysts.

Fuel Properties

The combustion properties of several fuels are summarised in Table I. The stoichiometric air:fuel ratio varies with the energy content of the fuel. The air:fuel ratio required for optimum combustion characteristics and thermal efficiency will affect both the engine operational parameters and the resulting exhaust gas composition, which determines the after-treatment strategy. This is highlighted below for natural gas and methanol fuelled vehicles.

For natural gas vehicles, the high stoichiometric air:fuel ratio implies that at an air flow rate comparable to that through gasoline engines a relatively low concentration of fuel will flow through the combustion chamber, thereby limiting the power generated by the engine. For lean burn engines where the air:fuel ratio is even higher, typically around 1.5 times the stoichiometric value, turbochargers are usually used to

Table I
Combustion Properties for Several Fuels

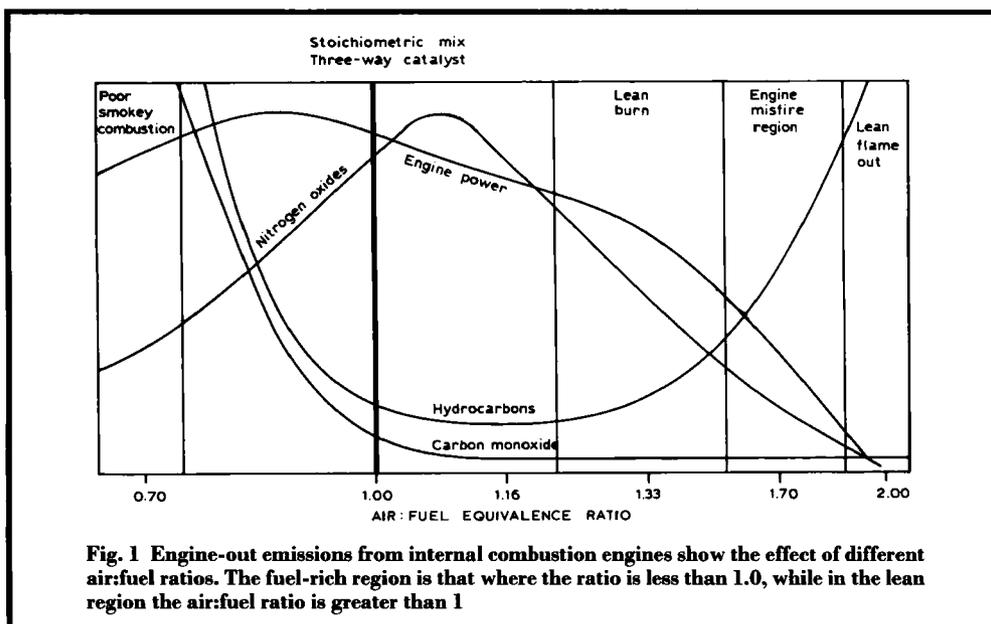
Fuel	Energy, MJ/kg	Energy relative to gasoline	Stoichiometric A/F ratio
Gasoline	42.9	1.00	14.6
Diesel	42.5	0.99	14.5
Natural gas	50.1	1.17	17.2
Propane	46.1	1.08	15.6
Ethanol	26.9	0.63	9.0
Methanol	19.9	0.46	6.5
M85*	23.5	0.55	7.6

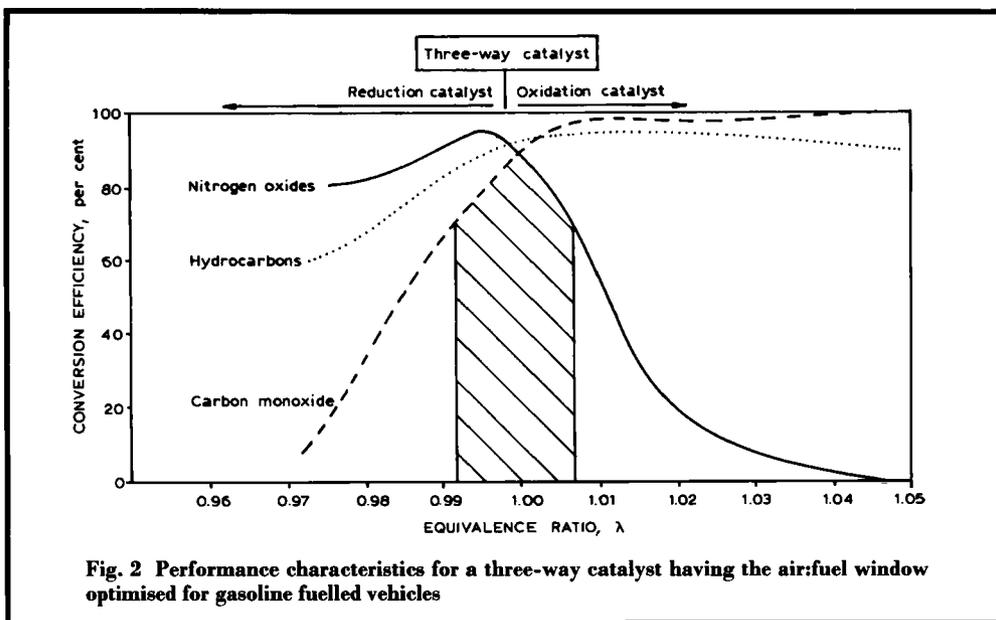
* 85% methanol + 15% gasoline

increase fuel throughput. As a consequence, the potential for oil leaks increases. Such leaks have been known to poison the activity of emission control catalysts, necessitating limitations on the additives that can be used in lubricating oils. Methanol fuel presents an opposite scenario: to support fuel combustion the high fuel concentration requires special fuel pumps and

fuel injection components, which must be compatible with the corrosive nature of methanol. Injector plugging can alter fuel calibration and affect the air/fuel control needed for maximizing emission control catalyst performance.

The usual representation of the emissions from internal combustion engines is shown as Figure 1. The characteristics are generic for all





hydrocarbons and/or oxygenated fuels. Hydrocarbon emissions are high at low air:fuel ratios (fuel-rich), decrease to a minimum at lean-of-stoichiometry and increase again in the lean burn regime as the combustion temperature is lowered by dilution with excess air. Carbon monoxide emission parallels that for hydrocarbons in the fuel-rich regime as it drops at stoichiometry but remains low in the lean burn regime. However, the emission of nitrogen oxides is high on the lean side of stoichiometry

and decreases as the air:fuel mixture becomes fuel-rich, and in the lean burn regime. Gasoline fuelled vehicles are generally calibrated to operate near stoichiometry, with a closed loop system utilising an oxygen sensor and a “three-way” emission control catalyst, which is optimised for the simultaneous oxidation of hydrocarbons and carbon monoxide, and the reduction of nitrogen oxides, see Figure 2.

The primary means of removing gaseous hydrocarbons from exhaust emissions is via

Fuel	Engine-out	Tail pipe (1)
Gasoline	50 –100	3 –10
Ethanol (2)	350	70 –100
Methanol	300	15 – 30
Methanol (3)	270	3 –10

(1) Tail pipe data shown for conventional three-way catalyst technology in underfloor position

(2) Ethanol data for open loop carburetted vehicles

(3) Fuel injected vehicles with close coupled catalysts

complete oxidation to carbon dioxide. The ease of oxidation increases in the order: methane < paraffins < aromatics < olefins < oxygenates. In general when operating near stoichiometry, palladium catalysts are superior to platinum catalysts for the oxidation of methane and unsaturated hydrocarbons, while platinum catalysts are better for the oxidation of paraffins. Rhodium is an excellent catalyst for steam reforming under rich conditions, and in addition it improves the durability of platinum and palladium catalysts and has high activity for the reduction of nitrogen oxides. Therefore, the catalyst formulation can greatly influence the hydrocarbon reactivity in automotive exhaust and allows emission control catalysts to be tailored for specific fuel-vehicle-catalyst systems. This approach has been successfully utilised for suppressing emissions from gasoline fuelled vehicles and appears to be required for the effective control of regulated, as well as unregulated (2), emissions from alternative fuelled vehicles.

Emission Control for Alcohol Fuelled Vehicles

Aldehyde and alcohol emissions are the primary concerns for emission control for vehicles fuelled with methanol and ethanol. Both emissions contribute to the formation of ozone. Also, aldehydes are considered to be toxic materials and the California Air Resources Board has adopted strict emission standards limiting aldehyde emission from light duty vehicles and heavy duty engines. Starting with the 1993 model year, the standard for passenger cars is to be 15 mg/mile and this figure falls to 8 mg/mile for 1997, and subsequent model years. These aldehyde emission levels are similar to tail-pipe emission levels measured on catalyst-equipped gasoline fuelled vehicles, Table II.

However, gasoline engines emit less aldehydes than alcohol engines and have higher exhaust gas temperatures which promote the catalytic destruction of emitted aldehydes. Existing three-way catalysts, as well as unoptimised catalysts coupled to methanol fuelled vehicles which have been converted from gasoline fuel, show aldehyde emission levels exceed-

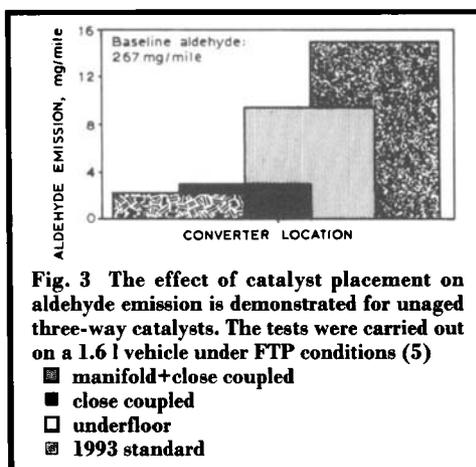


Fig. 3 The effect of catalyst placement on aldehyde emission is demonstrated for unaged three-way catalysts. The tests were carried out on a 1.6 l vehicle under FTP conditions (5)

- manifold+close coupled
- close coupled
- underfloor
- ▣ 1993 standard

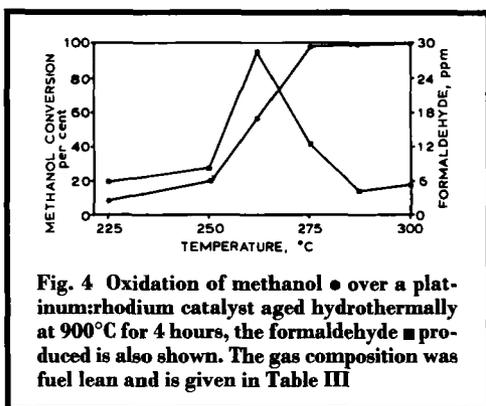
ing those required by the California Air Resources Board standard (3). Consequently, to reduce the levels of aldehydes from alcohol fuelled vehicles to levels similar to those from gasoline fuelled vehicles, and thus meet emission standards, the following are required:

- (a) the catalyst moved closer to the engine to take advantage of a hotter exhaust
- (b) dedicated catalyst technology, and
- (c) optimised fuel control and engine calibration strategy.

The effect of catalyst positioning on aldehyde emission is illustrated in Figure 3 for platinum+rhodium catalysts. Significant reduction in aldehyde emission is achieved by moving the catalyst from an underfloor position to a close-coupled location where the exhaust temperature is higher (4). The addition of a starter catalyst in the manifold further improves the suppression of aldehyde emission.

To illustrate the importance of matching optimised catalyst technology and fuel calibration strategy to minimise aldehyde emission, a review of aldehyde emission and catalytic treatment is warranted. There are two sources of aldehyde emission from alcohol fuelled vehicles equipped with emission control catalysts.

1. Aldehydes are formed from the partial combustion of alcohol fuel post-flame, and in the quench layer and exhaust manifold of the engine combustion chamber. Platinum group metals catalysts are effective for the catalytic decom-



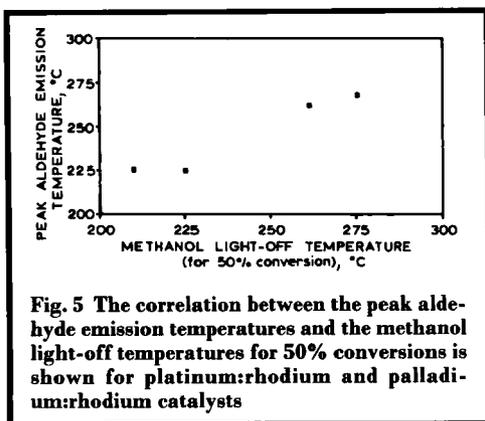
position of aldehydes and their subsequent oxidation to carbon dioxide. The oxidation is enhanced in the presence of excess oxygen in the exhaust (5, 6). This suggests that during a fuel-rich cold start situation, the addition of air to the exhaust gases upstream of the catalyst may be beneficial for the suppression of engine-out aldehydes (those leaving the engine).

2. Aldehydes are also formed over the emission control catalyst from the partial oxidation of unburned methanol exiting the engine. For catalysts containing platinum group metals this reaction occurs at relatively low temperatures (7). Maximum emission typically occurs over

Hill 1 of the Federal Test Procedure (FTP) cycle following cold start, and prior to catalyst light-off which would give complete combustion to carbon dioxide. Figure 4 shows the oxidation characteristics for a platinum+rhodium catalyst in a simulated laboratory test; the conditions are shown in Table III. Peak aldehyde emission arising from partial oxidation of methanol precedes total combustion to carbon dioxide. The relationship is illustrated in Figure 5 for several platinum+rhodium and palladium+rhodium catalyst formulations. Aldehyde emission increases with the air:fuel ratio in the lean regime, is low in the fuel-rich regime, and is lowest near to stoichiometry, as shown in Figure 6. This suggests that stoichiometric air:fuel calibration during cold start would minimise this source of aldehyde emission.

For a particular engine, the need for air to be added to the exhaust is likely to be determined by the relative importance of the two aldehyde sources. The interplay between exhaust temperature (which may be lowered by the addition of air), and fuel and calibration control is a principal factor in this determination. Matching and optimising the air:fuel ratio control and the emission control catalyst appears necessary to effect a net suppression of

Gas component	R Value, molar ratio of reductants:oxidants		
	0.33 Lean	1.00 Stoichiometric	3.03 Rich
Methanol, ppmC ₁	900	900	900
NO _x , ppm	900	900	900
CO, %	0.34	0.34	1.90
H ₂ , %	0.16	0.16	0.16
O ₂ , %	1.12	0.34	0.34
CO ₂ , %	10.0	10.0	10.0
N ₂	Balance	Balance	Balance



aldehyde and alcohol emissions. Such designs also need to be applied to “flexible fuelled” vehicles, these being vehicles which can operate on a variable mixture of alcohol and gasoline. Such vehicles can also operate on gasoline which contains sulphur, thus in addition to controlling hydrocarbons, carbon monoxide, nitrogen oxide, and aldehydes, it is necessary to control simultaneously the emission of hydrogen sulphide (8) by means of a “Five-Way” catalyst.

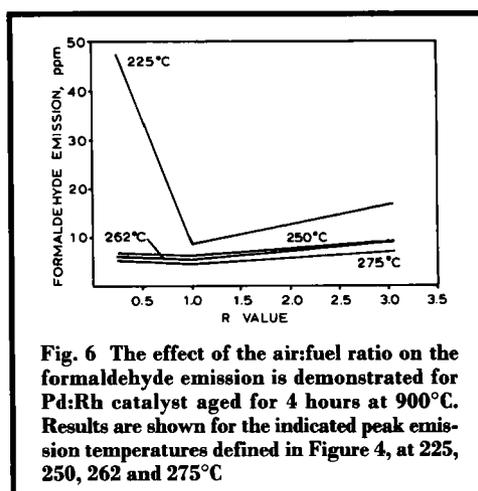
Emission Control for Natural Gas and Propane Vehicles

There is significant interest in the use of natural gas as a clean fuel for urban vehicles (9). Methane is the dominant hydrocarbon component in the exhaust of vehicles fuelled with natural gas; it is also the least reactive hydrocarbon (10) and contributes little to ozone formation, though it is a powerful greenhouse gas. While current and near term emission standards are generally based on non-methane hydrocarbon (NMHC), moves are underway to introduce legislation that will use a total hydrocarbon (THC) standard for all fuels, including natural gas. This will present a significant challenge for both emission control catalysts and engine calibration strategies for natural gas vehicles. The following is an assessment of the effect of engine calibration and catalyst performance characteristics on emission control strategies for both stoichiometric and lean burn engines fuelled with natural gas.

The performance characteristics for three-way emission control catalysts for gasoline fuelled vehicles were shown above in Figure 2. The catalysts are optimised to provide a wide air:fuel window which allows simultaneous oxidation of hydrocarbons and carbon monoxide, and reduction of nitrogen oxide. Fuel control is required to optimise the emission control system and is achieved by maintaining the air:fuel ratio at or near to stoichiometry, by the use of a feedback loop incorporating an oxygen sensor.

Platinum group metal catalysts possess high reactivities for methane (10, 11) and are the most effective catalysts for the treatment of natural gas exhaust, as they are for gasoline exhaust. The corresponding three-way characteristics for an aged platinum+rhodium catalyst developed for natural gas vehicles are shown in Figure 7 as a function of R, the redox value (defined as the molar ratio of reductants:oxidants). Gas compositions are shown in Table IV.

For natural gas, the observed air:fuel window is narrower than for gasoline, and the optimum is shifted toward the fuel-rich side of stoichiometry. This behaviour appears to be correlated with the reduced reactivity for methane conversion at, and on the lean side of stoichiometry. The high hydrocarbons conversion under fuel-rich conditions can be attributed to the catalytic steam reforming of methane (12). Figures 8 and 9 show the effect



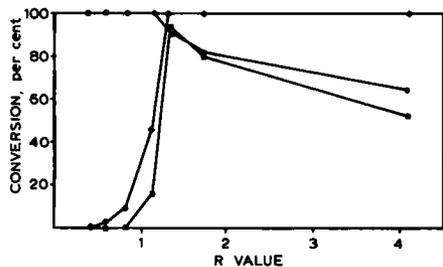


Fig. 7 Three-way performance characteristics for hydrothermally aged platinum:rhodium catalyst. Tests were carried out at 500°C and gas compositions are shown in Table IV

- methane
- carbon monoxide
- ◆ nitrogen oxides

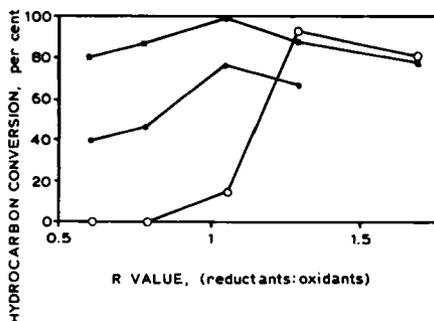


Fig. 8 Hydrocarbons conversions over a platinum:rhodium three-way catalyst, near the stoichiometric air:fuel ratio, where $R = 1$, are shown. Measurements were made at 500°C and on equal CI basis:

- propane
- ethane
- methane

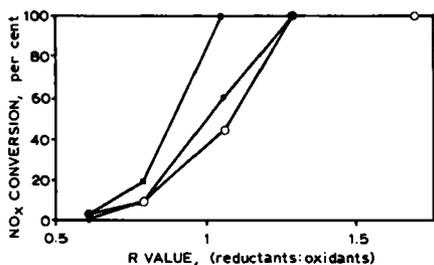


Fig. 9 The percentage of nitrogen oxide converted corresponding to the conversion of methane, ethane and propane shown in Figure 8. The conversions occur over platinum:rhodium three way catalyst and the stoichiometric air:fuel ratio is at $R=1$

- propane
- ethane
- methane

of the reactivity of three particular hydrocarbons on the conversion of both hydrocarbons and nitrogen oxides; propane, being the most reactive in the series, exhibits the widest air:fuel window. The matched suppression in activity for hydrocarbons and nitrogen oxides at stoichiometry appears to highlight the role of hydrocarbon activity in the catalytic reduction of nitrogen oxides.

The reduced performance at the stoichiometric air:fuel ratio highlights the need for matching the fuel control strategy of the engine/vehicle with the three-way characteristics of the catalyst, and for maintaining calibration for optimum emission control. This matching can be aided by the use of a proportional (as opposed to a switching) oxygen sensor to fix the air:fuel control set-point, at a rich-of-stoichiometry value, and to minimise air:fuel swings outside the optimum air:fuel window of the catalyst. Such a strategy has been demonstrated to be effective in minimising emissions (13).

The data in Figures 8 and 9 suggest that the emission control characteristics for propane fuelled vehicles are similar to those for gasoline fuelled vehicles and that propane vehicles therefore can utilise conventional three-way catalyst technology for emission control. However, nitrogen oxides emissions may be higher for propane fuelled engines (as is the case with natural gas) and may require exhaust gas recycling and also high activity catalysts for effective control (14).

Lean burn engines fuelled with natural gas require an oxidation catalyst for reducing the emissions of hydrocarbons and carbon monoxide. Nitrogen oxides emissions are inherently low for this calibration and are not affected significantly by the oxidation catalyst. Since methane is the dominant hydrocarbon in the exhaust, and the least reactive, its catalytic oxidation is characterised by high light-off temperatures. Oxidation catalysts containing platinum metals are the most effective for the oxidation of methane (10, 11) and have been optimised to suppress hydrocarbons and carbon monoxide in the exhaust of lean burn engines. Efforts are underway to develop catalysts to suppress nitrogen oxides emissions under

the lean burn environment. Such catalysts appear necessary to meet future, more stringent emission standards for nitrogen oxides.

The combustion of methane in engines produces a significant concentration of CH₃ radicals, which are precursors to aldehydes. Additionally, the exhaust contains some particulates – though substantially less than from diesel engines – arising from uncombusted lubricating oil. With the exception of two-stroke engines, exhaust gas temperatures for most natural gas vehicles are generally higher than for gasoline fuelled vehicles. This provides favourable conditions for platinum metals catalysts to suppress both aldehyde and particulate emissions.

Emission Control for Diesel Engines

The development of emission control technologies for diesel engines has been the subject of numerous publications, see for example (15), and is not reviewed in this paper. The emerging emission control requirements, however, are reviewed from technical perspectives and are classified as follows:

1. Meet particulate emission standards. The most stringent of these standards are those

adopted in the U.S.A. for heavy duty engines, namely 0.1 g/bhp-hour for 1994 trucks and 1993 urban buses, with the possibility of the adoption of 0.07 g/bhp-hour standard for 1994 urban buses. Several strict standards are also being adopted for light duty passenger cars, particularly in Europe where a significant and growing number of vehicles are fuelled with diesel. Engine manufacturers usually set an engineering target of about 75 per cent of the levels dictated by the standards in order to provide a safety margin for engine manufacturing variability and durability.

2. Reduce exhaust odour and gaseous hydrocarbons. Aldehydes and ketones characterise the odorous emissions from diesel engines but oxidation catalysts are usually effective in suppressing both these odours and the hydrocarbons emissions from diesel exhaust. While gaseous hydrocarbons emission from diesel engines is low, the requirement for meeting a combined hydrocarbons+nitrogen oxide emissions standard for Europe presents a need to limit hydrocarbons emissions, since currently it is difficult to reduce nitrogen oxides emissions under lean conditions from mobile sources via catalytic means, and without sacrificing the

Table IV
Gas Composition for Synthetic Gas Test to Characterise Three-Way Performance of Catalysts for Natural Gas Vehicles
Measurements at 500°C and 65,000/h gas hourly space velocity

Gas component	R Value						
	0.400	0.612	0.787	1.062	1.300	1.701	4.051
HC ppmC ₁	850	850	850	850	850	850	850
CO ppm	1000	1500	2000	2500	3000	3500	4000
H ₂ ppm	1000	1500	2000	2500	3000	3500	4000
NOx ppm	1700	1500	1450	1400	1350	1300	1100
O ₂ ppm	5906	4482	3979	3256	2941	2406	857
CO ₂ %	9.45	9.45	9.45	9.45	9.45	9.45	9.45
H ₂ O %	15.0	15.0	15.0	15.0	15.0	15.0	15.0
N ₂	Balance						

control of particulate emissions via nitrogen oxide-particulate emissions trade-off.

To meet control requirements for particulate emission, an after-treatment device such as a flow-through catalyst appears necessary. Platinum group metals catalysts are known to be effective oxidation catalysts and are used to suppress gaseous emissions from various vehicle engines. The catalysts can also produce significant reductions in particulate-bound hydrocarbons, known as the soluble organic fraction (SOF), originating from uncombusted lubricating oil and fuel (16). The combustion of the soluble organic fraction must occur at temperatures below 300°C since diesel exhaust is generally characterised by low temperatures, particularly for direct injection engines. Particulate condensation and trapping by the catalyst must be minimised to prevent device plugging, which can lead to an increase in back pressure and reduced engine performance. This problem has plagued the use of particulate traps and has brought into focus the potential use of a flow-through catalyst as the after-treatment device of choice (17), particularly since prototype engines are being developed with low particulate emission. For these engines to meet emission targets safely, the catalyst is needed to "trim" the particulate emissions sufficiently to overcome production variability and engine wear.

Sulphate emission from diesel engines is produced during the combustion process in the engine cylinder. Approximately 2 per cent of fuel-bound sulphur is converted to sulphur trioxide and is emitted as sulphuric acid or metal sulphate. Their contribution to particulate emission is further increased by their hygroscopic properties which lead to the absorption of water to approximately 130 per cent of their weight. The emission of sulphate from diesel engines equipped with oxidation catalysts can be significantly higher than that in uncatylased baseline engine-out emissions (17, 18). The catalyst must therefore exhibit minimum activity in converting exhaust sulphur dioxide to sulphur trioxide, which leads to the emission of sulphuric acid and increases particulate emissions.

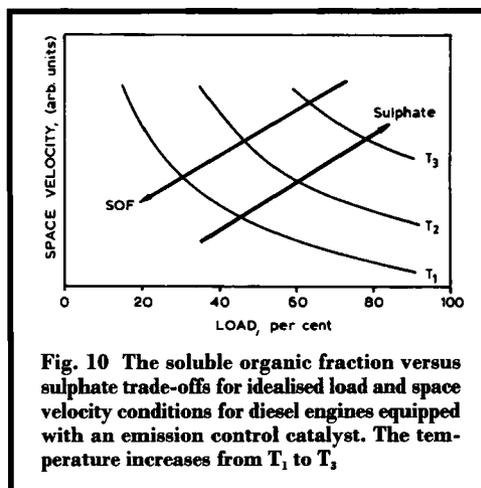


Fig. 10 The soluble organic fraction versus sulphate trade-offs for idealised load and space velocity conditions for diesel engines equipped with an emission control catalyst. The temperature increases from T_1 to T_3

As the conversion of sulphur dioxide to sulphur trioxide increases with temperature below the thermodynamic limit, at about 600°C, the catalyst is required to limit the reaction in a temperature range of about 300 to 600°C.

The previous discussion suggests that when coupling a catalyst to a diesel engine, significant reductions in the soluble organic fraction can occur at the expense of substantial increases in both sulphuric acid and particulate sulphate emissions, particularly for active catalysts and under high temperature exhaust conditions. This trade-off between the reduction in the

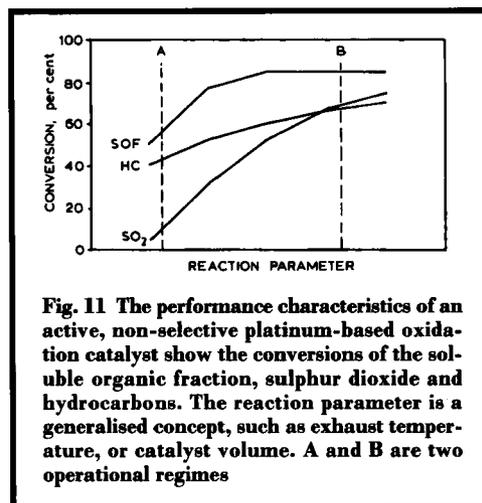
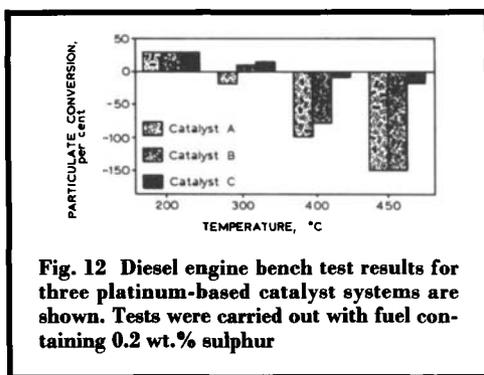


Fig. 11 The performance characteristics of an active, non-selective platinum-based oxidation catalyst show the conversions of the soluble organic fraction, sulphur dioxide and hydrocarbons. The reaction parameter is a generalised concept, such as exhaust temperature, or catalyst volume. A and B are two operational regimes



soluble organic fraction and the increase in sulphate is controlled by engine speed and engine load conditions, as depicted in the generic representation of Figure 10 for operational regimes characterised by the engine exhaust temperatures, which are shown increasing from T_1 to T_3 . Catalysts with high selectivity are required to possess high activity for reducing the soluble organic fraction content at low exhaust temperatures, and also to have low activity for the conversion of sulphur dioxide to sulphur trioxide.

In general, the emission control system of which the catalyst is a component can be engineered for maximum effectiveness using the known exhaust characteristics of the engine and the catalytic properties of the device. Figure 11

illustrates the characteristics for an active platinum-based oxidation catalyst, where performance is plotted as a function of a generalised reaction parameter such as exhaust temperature or catalyst volume. For such a catalyst less sulphate is produced in operational regime A as compared with regime B. This is achieved at the expense of some reduction in catalyst capability for the soluble organic fraction conversion. Such a trade-off in activity may be acceptable in controlling emissions for some engines. In general however, the device must be effective for the combustion of the soluble organic fraction-rich particulate at low temperatures, and may be required simultaneously to have low activity for converting sulphur dioxide to sulphur trioxide at high temperatures. This will minimise the resulting sulphuric acid emission and the sulphate contribution to particulate emissions (19).

Total (net) particulate conversion over a range of steady-state modes represented by exhaust temperatures measured at the catalyst inlet, and given in Table V, is shown in Figure 12. The region of negative conversion is attributed to the tail-pipe particulate emission being dominated by sulphate particulates generated by the catalyst. The new Eurocycle for certification of future passenger cars in Europe requires, simultaneously, high soluble organic fraction and

Speed, rpm	Torque, Nm	Exhaust temperature, °C	Particulates, g/h	HC, ppm	NOx, ppm	CO, per cent
2500	29	200	4.1	87	137	0.2
2500	44	250	6.1	91	181	0.2
2500	57	300	7.1	118	221	0.1
2500	72	350	7.5	103	291	0.2
2500	88	425	8.3	85	380	0.2
2500	100	500	9.2	69	407	0.2

gaseous hydrocarbons conversions at low temperatures (characteristic of the ECE component of the cycle), and low sulphur dioxide conversion at high temperatures (characteristic of the EUDC component of the cycle). The results for catalyst C in Figure 12 suggest that it has performance characteristics which can meet these requirements.

Catalyst development could be aided significantly if the amount of sulphur in the fuel was to be dropped below 0.05 weight per cent, the level currently being adopted by several countries. The use of low sulphur fuel would circumvent the soluble organic fraction-sulphate trade-off, and very active catalysts could then be developed to suppress both the soluble organic fraction and the hydrocarbons emissions (20).

Summary

Emission characteristics, fuel control strategies, and emission control systems for alternative fuelled vehicles have been considered; some of the most notable features are:

* Control of aldehyde emission from alcohol fuelled vehicles is needed to meet emission control requirements. "Five-Way" catalysts placed close to the engine manifold to make use of higher exhaust gas temperatures are effective

for suppressing aldehyde and alcohol emissions from flexible fuelled vehicles.

* Emission control for "stoichiometrically" calibrated natural gas vehicles has been shown to be optimum at a rich-of-stoichiometric air:fuel calibration, to match the air:fuel window for dedicated three-way catalysts. Oxidation catalysts with improved light-off for methane combustion can be utilised to control hydrocarbons and carbon monoxide emissions from lean burn engines.

* The principle emission control issues for diesel engines are the reduction of the soluble organic fraction of diesel particulates and the gaseous hydrocarbons emissions at low temperatures, coupled with the simultaneous suppression of sulphate generation at high temperatures by means of emission control catalysts. Lowering the level of sulphur in fuel will aid further the development of more effective catalysts.

Acknowledgements

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