

# Car Exhaust Catalysts for Europe

## THE DEVELOPMENT OF LEAD TOLERANT PLATINUM CATALYST SYSTEMS FOR EMISSION CONTROL

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*Although Europe is following the example of the U.S.A. in progressively lowering permitted emission levels of pollutants from motor vehicles, it seems unlikely to introduce lead free petrol in the foreseeable future. Proposed regulations seek a level of emission control which will, in all probability, necessitate the use of oxidation catalysts. In the presence of leaded fuel conventional autocatalysts used in the U.S.A. are susceptible to poisoning after relatively low mileages and there is a potential requirement for a catalyst system capable of functioning satisfactorily on fuel containing substantial concentrations of lead. Following the identification of the nature and the type of the poisoning lead species, Johnson Matthey has developed a first generation lead tolerant catalyst system for emission control. This has demonstrated durability for 50,000 kilometres road usage with leaded fuel.*

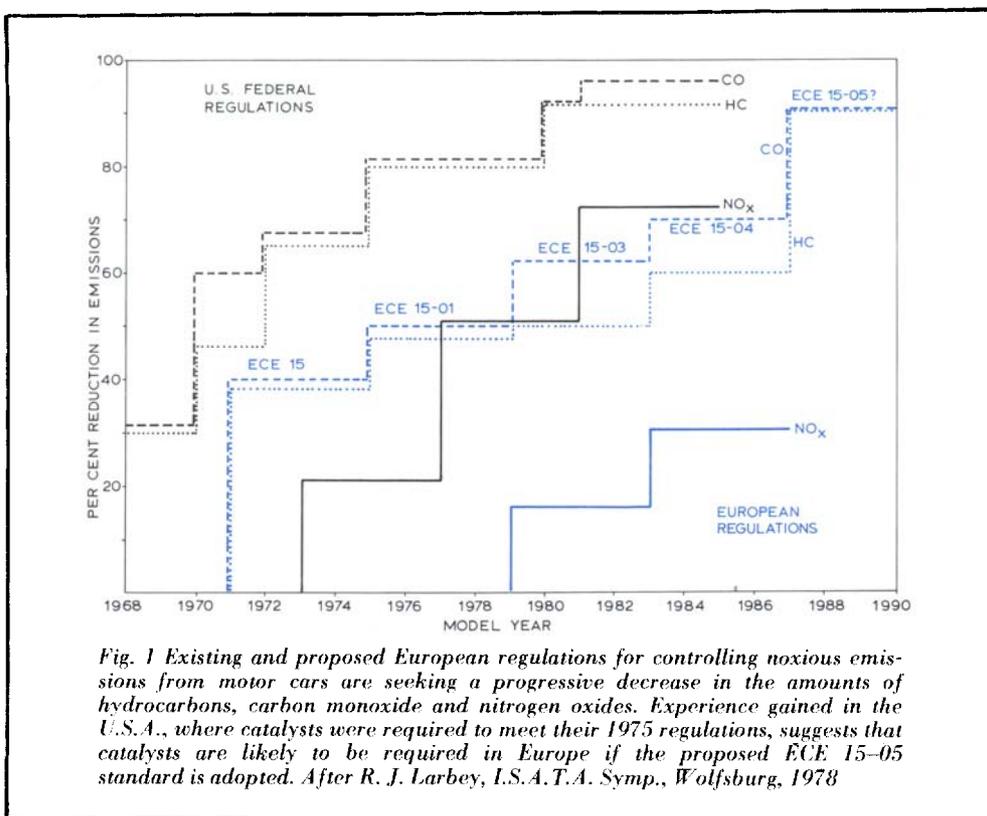
The presence of lead in petrol and potential health hazards resulting from its emission in car exhaust has been the subject of much public debate recently. However, this is really only part of the total scene concerning the contribution of the motor vehicle to atmospheric pollution. Of the major exhaust gases carbon monoxide is a respiratory poison while unburnt hydrocarbons and oxides of nitrogen are known to interact in the presence of sunlight to produce the lachrymatory compounds which

are the components of photochemical smog (1). The dispersion of photochemical smog and its precursors can be retarded by climatic conditions and has become a particular problem in Los Angeles and Tokyo. This situation is by no means confined to the U.S.A. and Japan, however, and photochemical smog and related phenomena have been observed in some European cities, notably in Madrid (2) and Athens (3) and even in London (4).

### Emission Legislation

The U.S.A. has introduced increasingly severe vehicle emission regulations as a result of the Clean Air Act of 1970 and its subsequent amendments (5). Prior to 1975 ignition and carburation modifications were used in conjunction with secondary air injection and thermal reactors; all of these techniques having a generally detrimental effect on fuel economy. However, since 1975, catalyst technology has been successfully introduced, giving rise to two major side effects. First, most vehicle manufacturers were able to re-tune engines for better economy while relying on the catalyst to control emissions. Secondly, it ensured that the trend to lead-free petrol was maintained as conventional automobile catalysts are poisoned by lead. Thus the overall effect was to tackle both gaseous pollutants and lead. A similar situation has occurred in Japan.

Although emission levels in Europe permitted by legislation are being progressively lowered they are presently far higher than those in the U.S.A., as shown in Figure 1. For the 1981 model year the U.S.A. demands reductions of 90, 90 and 70 per cent, respectively for hydrocarbon, carbon monoxide and nitrogen oxide emissions relative to unregulated 1968



*Fig. 1 Existing and proposed European regulations for controlling noxious emissions from motor cars are seeking a progressive decrease in the amounts of hydrocarbons, carbon monoxide and nitrogen oxides. Experience gained in the U.S.A., where catalysts were required to meet their 1975 regulations, suggests that catalysts are likely to be required in Europe if the proposed ECE 15-05 standard is adopted. After R. J. Larbey, I.S.A.T.A. Symp., Wolfsburg, 1978*

levels. In contrast the E.E.C. amendment presently being introduced calls for only 50, 60 and 15 per cent compared to unregulated levels. However, a further reduction in permitted emissions is due in 1983/4 model years (04 amendment) and proposals for an 05 amendment involving a 90 per cent reduction in hydrocarbon and carbon monoxide levels have been made, notably by the West German Umweldbundesamt (UBA). This level of control is similar to that which is current in the U.S.A. and could become law in Europe in the late 1980s. In addition, Switzerland is independently proposing to introduce legislation equivalent to 1977 U.S. Federal limits in 1986, and Sweden is pursuing a similar course.

As European emission legislation is strengthened, control techniques similar to those used in the U.S.A. until 1975 are available to car manufacturers and it is likely that most vehicles will be able to meet the standards up to ECE 15-04 without catalysts, although, in

some cases, a fuel economy benefit may be possible by re-tuning and using catalysts. However, the ECE 15-05 standards will necessitate catalytic control on the majority of cars. Present evidence suggests that although the lead level in gasoline will be gradually reduced from the current average of around 0.4 down to 0.15 grams per litre (presently used in West Germany and to be introduced in the U.K. in 1985), lead-free petrol is unlikely to be available. Should concerns about lead health effects result in a change in this situation then catalytic converters currently used in the U.S.A. would readily be available for Europe. If not, it follows that any catalysts fitted to cars in Europe should be tolerant of lead, at least at the 0.15 grams per litre level and preferably at the higher 0.4 grams per litre level also.

Johnson Matthey has been involved in the development of ceramic monolith supported automobile catalysts for the U.S. market for over ten years and has supplied more than ten

million units. Oxidation catalysts, capable of removing carbon monoxide and unburned hydrocarbons, were introduced in 1975 (6) and three-way catalysts designed for simultaneously removing these pollutants together with nitrogen oxides were introduced in 1981 (7). In anticipation of legislation necessitating the use of catalysts on European cars, Johnson Matthey has recently undertaken a project aimed at developing lead tolerant catalyst systems. Fuel parameters, systems variables and catalyst parameters have all been investigated in an attempt to identify not merely a catalyst but also the conditions under which it should be used. Initially the work has concentrated on oxidation catalyst technology and has been partly funded by the U.K. Department of Industry. Part of the work has been subcontracted to BL Cars and the U.K.A.E.A. at Harwell, while British Petroleum and Associated Octel have provided fuels and additives.

### **Fuel Composition and Its Influence on Catalyst Deactivation**

Tetraethyl lead or tetramethyl lead (TEL or TML) are normally added to gasoline as octane improvers to prevent pre-ignition or "knocking" in the engine. In addition to lead, "scavengers" are also included in the fuel in the form of ethylene dibromide (EDB) and ethylene dichloride (EDC). These compounds decompose during the combustion process to form hydrogen bromide and hydrogen chloride, which in turn interact with lead to form volatile lead halides. In this way, lead is effectively removed or scavenged from the cooler parts of the combustion chamber. The quantity of scavengers added to the fuel is expressed in terms of the theoretical amount necessary to interact with all the lead. Thus, the most common additive package, known as "Motor Mix" contains 1 Theory EDC and  $\frac{1}{2}$  Theory EDB, that is, an excess of scavenger. Tricresylphosphate (TCP) has occasionally been added to gasoline as a combustion improver, and sulphur is normally present at a level of approximately 300 ppm.

Thus a wide range of lead species may be formed in engine exhausts, depending on fuel composition and prevailing engine operating parameters such as speed, load and tune. Since lead is a notable poison for both noble and base metal catalysts these factors can also be expected to have a marked influence on catalyst durability.

During the mid-1970s, a number of workers, including those at Chrysler (8, 9), Ford (10, 11), General Motors (12) and Johnson Matthey (13), reported the effect of fuel composition and different lead compounds on precious metal catalyst durability. This work was conducted at the levels of lead then prevailing in U.S. fuel. Although the conclusions reached were in general agreement, a controversy arose over the Chrysler workers' claim that lead did not poison catalysts if ethylene dibromide was not added to the fuel. In view of this it was considered that the first step in approaching the development of lead tolerant catalysts should be to obtain an idea of the lead species likely to be formed under varying operating conditions. A fundamental thermodynamic approach was adopted to develop a model for the system from which concentration of lead compounds such as oxide, chloride, bromide, oxyhalides, sulphate and oxysulphates could be predicted under varying operating conditions. A summary of the results is presented in Table I.

For Motor Mix fuel containing 0.4 g/l lead, the maximum level currently used in most European countries, gaseous halide species predominate at 600°C (assuming no sulphur and an oxygen level of 2 to 4 per cent) while at 400°C a solid oxychloride is the major form of lead. Notwithstanding the possible differences in toxicity of individual lead species, it can be concluded that the nature of lead poisoning of catalysts under the lower temperature condition is likely to be due largely to physical coverage by particulate lead species. In contrast, at the higher temperature, vapour phase lead species are likely to give faster poisoning based on chemical reactions. At 400°C a marginal influence of O<sub>2</sub> potential on the equilibrium is seen, whereas this is not apparent at 600°C.

Fuel additives				Operating conditions		Phases predicted (per cent of Pb)					
Pb (g/l)	EDB (Th)	EDC (Th)	S (ppm)	O <sub>2</sub> conc. (per cent)	Inlet temp. (°C)	Solid			Gaseous		
						PbO	2PbO.PbCl <sub>2</sub>	PbSO <sub>4</sub>	PbO	PbBr <sub>2</sub>	PbCl <sub>2</sub>
0.4	0.5	1	0	2-4	600	19	—	—	1	46	34
				2	400	—	89	—	—	9	2
				4	400	—	91.5	—	—	6.5	2
0.15	0.5	1	0	2-4	600	34	—	—	3	44	19
				2	400	—	85	—	—	13.5	1.5
				4	400	—	88	—	—	10.5	1.5
0.15	0			2-4	600	97	—	—	3	—	—
				2-4	400	100	—	—	—	—	—
0.4	0.5	1	300	1-4	300-600	—	—	100	—	—	—

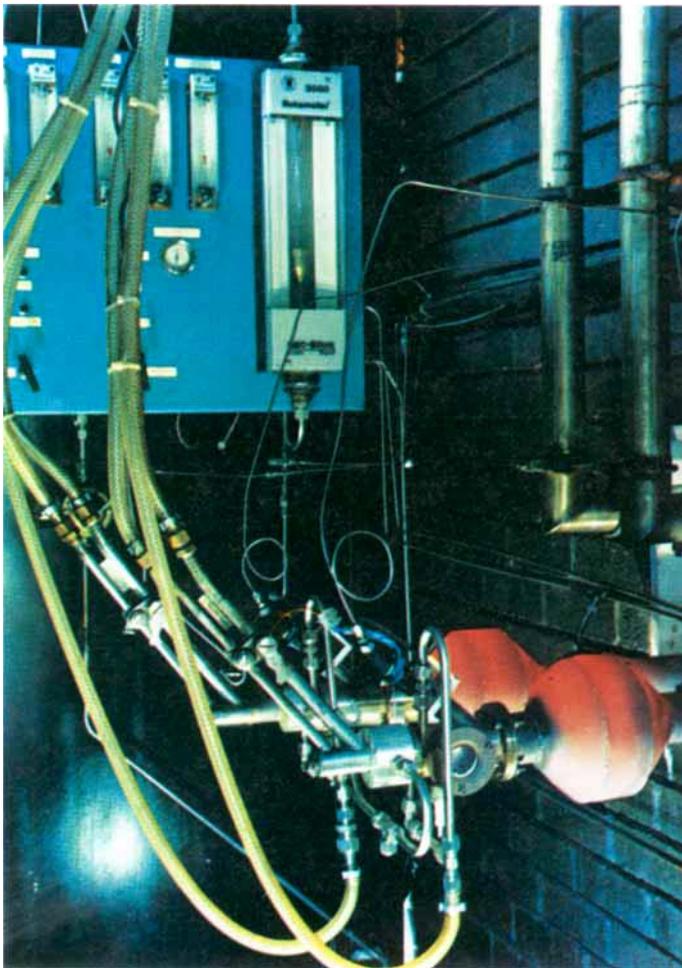
At the level of 0.15 g/l lead (again as Motor Mix) as currently used in West Germany, and about to be adopted in some other European countries including the U.K., a higher proportion of lead is in the solid phase at 600°C suggesting that a combination of the two poisoning effects should be expected.

The influence of the scavenger additives can be seen from the prediction that all exhaust lead would be in the solid phase at 400°C and 97 per cent would be solid at 600°C, for 0.15 g/l lead fuel without scavengers.

In reality all commercial gasoline contains sulphur largely derived from the original crude oil and when a typical level of sulphur, 300 ppm, is present in fuel containing either 0.4 or 0.15 g/l lead as Motor Mix, the model predicts a totally different situation with solid phase lead sulphate being the thermodynamically stable phase from 300 to 600°C at 1 to 4 per cent O<sub>2</sub>. If all of the lead were converted to solid lead sulphate in practice, one might expect scavengers to be totally ineffective; one might also expect physical poisoning of catalysts to occur to the complete exclusion of chemical poisoning. Experience, however, is to the contrary,

indicating that PbSO<sub>4</sub> formation involves a slow step, the oxidation of SO<sub>2</sub> to SO<sub>3</sub>, prior to interaction with lead compounds. It would be expected, therefore, that the composition of the lead species in the exhaust would be similar for fuel containing 300 ppm sulphur to that for sulphur-free fuel. However, over platinum-containing catalysts, and particularly in the presence of the high levels of oxygen that are often obtained when secondary air injection is used, SO<sub>2</sub> oxidation to SO<sub>3</sub> followed by reaction with PbO to form PbSO<sub>4</sub> would be considerably more favoured. This would explain the practical observation that catalysts which have run on leaded, sulphur containing fuel are rarely found, in analysis, to contain lead halides or oxide halides, but normally are contaminated with either lead sulphate or lead oxide sulphates. It is tempting to suggest that lead reaches the catalyst in the form of halide, oxide halide or oxide and then reacts with SO<sub>3</sub> to form the stable lead oxide sulphates or lead sulphate which are difficult to remove.

At the Johnson Matthey Research Centre, the pulse flame reactor test rigs shown in Figure 2 have been used to study the effect of lead and

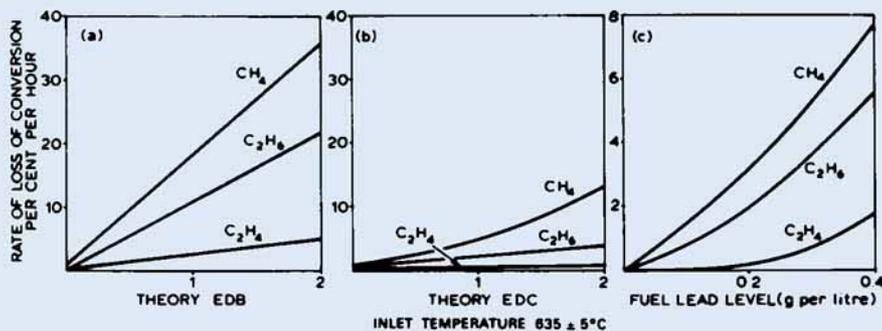


*Fig. 2 Pulse flame reactor test rigs enable large numbers of catalysts and fuel additives to be evaluated rapidly. Standardised fuel and air mixtures are burnt in the combustion chambers and the resulting gases pass through the catalyst. A comparison of inlet and exhaust gas concentrations enables the efficiency of the catalyst to be determined*

other fuel additives on catalyst deactivation. For this purpose, 2" diameter  $\times$  3" long cores of a standard, platinum-containing, U.S. type, monolithic oxidation catalyst were used. Petroleum, doped with the additive mixture under investigation, was injected into a combustion chamber using a standard fuel injector and combustion was initiated by a propane/air pilot flame. The exhaust, together with added secondary air, was passed through the catalyst sample, and inlet and outlet gases were analysed. This type of procedure was used as a short-term eight hour screening test and although a wide range of hydrocarbons and also carbon monoxide were present in the exhaust gas only methane, ethane and ethylene were regularly monitored, because, under the test

conditions employed, no catalyst deactivation was observed for higher hydrocarbons, or indeed for carbon monoxide, over this interval. Catalyst deactivation for oxidation of these three lower hydrocarbons was therefore used as an indicator of likely performance for the removal of total exhaust hydrocarbons over a longer timespan. Unless otherwise indicated, steady state ageing conditions, involving a catalyst inlet temperature of 630 to 650°C, were used.

Experimentation has shown that, in the presence of a fixed concentration of lead of 0.4 g/l and the absence of EDC, a linear relationship exists between the rate of catalyst deactivation and EDB concentration, as shown in Figure 3. The effect of EDC in the presence of lead



**Fig 3** These data demonstrate how different levels of lead and scavenging additives in the fuel affect the rate at which a standard catalyst is deactivated. With fuel containing 0.1 grams per litre of lead, a linear relationship exists between EDB concentration and the rate of deactivation for methane ( $\text{CH}_4$ ), ethane ( $\text{C}_2\text{H}_6$ ) and ethylene ( $\text{C}_2\text{H}_4$ ) (a), but the effect of EDC concentration is considerably less (b), while in the absence of scavengers deactivation was extremely slow. In the presence of a Motor Mix additive package the rate of catalyst poisoning increases with lead concentration (c)

and absence of EDB was markedly less. This is in accordance with the thermodynamic model which predicts a higher gas phase concentration of  $\text{PbBr}_2$  than of  $\text{PbCl}_2$  under these test conditions. This enables faster transport of lead to the catalyst, which results in faster chemical poisoning. The fact that increased quantities of either scavenger, above 1 Theory, continued to affect poisoning rate, illustrates the equilibrium which exists between solid lead oxide and the gaseous lead halide in each case. With no scavengers present in the fuel, extremely slow deactivation was observed, owing to the drastically reduced concentration of gaseous lead species—most of the available lead being present as solid  $\text{PbO}$ —with only a small amount of vapour phase  $\text{PbO}$ . As expected, catalyst poisoning rate increased with increasing lead concentration when lead was incorporated in the presence of Motor Mix scavenger additives. Sulphur was found to have little effect on catalyst deactivation when added to Motor Mix doped fuel, providing evidence that sulphur plays little part in the transport of lead to the catalyst surface.

Following these rig tests, static engine durability tests were conducted on similar catalysts of 4" diameter  $\times$  6" long, using leaded fuels containing zero or low levels of scavenger.

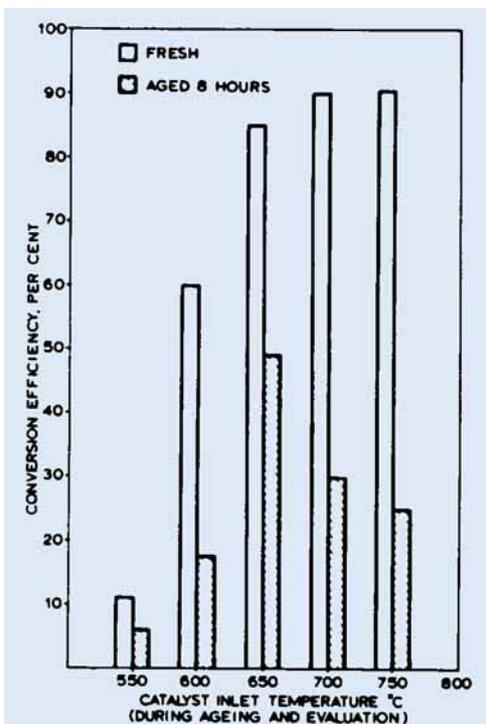
As in the rig tests, lower deactivation rates for hydrocarbon oxidation were observed as compared with cases where Motor Mix fuel was used. However, a heavy build-up of deposited lead compounds, which plugged a high proportion of the monolith cells, caused a rapid loss in performance for carbon monoxide removal.

Thus, although it is possible to demonstrate that a reduction in the level of scavengers, particularly ethylene dibromide, present in leaded fuel can cause a reduction in the rate of chemical poisoning by lead species, it is clear that physical contamination is likely to become more prevalent. This limits the potential of the use of such fuels as a means of minimising catalyst deactivation.

### Emission System Design

Although catalyst design is of primary importance in the development of a lead tolerant system, the conditions under which the catalyst must operate also play an important role with regard to activity and durability. One of the critical factors affecting the performance of a catalyst is the inlet temperature, since this influences light off, activity, stability and, as discussed above, the nature of the lead species reaching the catalyst.

The effect of inlet temperature was



*Fig. 4 Data established at temperatures up to 750°C, using fuel containing lead in a Motor Mix additive package, show that although some reduction in conversion efficiency for methane was observed in all cases this was minimised at 650°C, demonstrating the advantage that would be gained by operating an emission control catalyst close to this temperature when lead is present in the fuel*

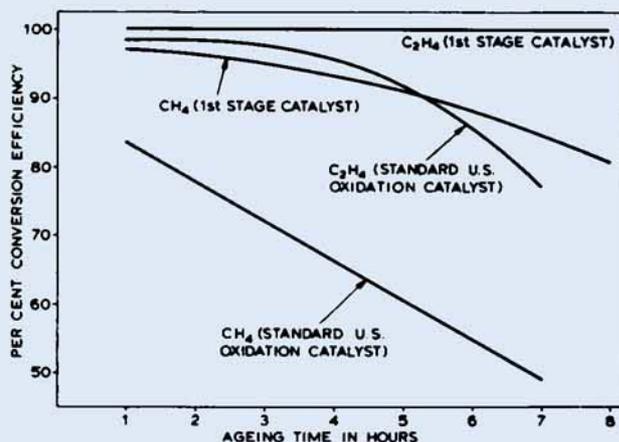
investigated by performing pulse flame reactor tests in the range 550 to 750°C, again using U.S. type oxidation catalysts, in conjunction with fuel doped with 0.4 g/l lead (Motor Mix). One catalyst was both evaluated and aged at each of five different temperatures between 550 and 750°C and the results, for methane conversion only, are shown in Figure 4. The five fresh catalysts effectively demonstrated light off for methane, with catalyst activity increasing as temperature increased up to a conversion efficiency of 90 per cent at 750°C. After ageing for eight hours, a different trend emerged and an optimum in performance was observed at a catalyst inlet temperature of 650°C. It was also noticed that poisoning had shifted the effective

light off temperature for methane to a higher level.

These phenomena can be explained in terms of the lead species present at each ageing temperature. The faster deactivation observed at 700 and 750°C can be explained by the onset of PbO volatilisation at temperatures above about 670°C, thus increasing the chemical poisoning of the catalysts aged at these temperatures. At lower temperatures, poisoning is largely caused by lead halides alone, since PbO is non-volatile. It appears, therefore, that an optimum temperature exists at which lead halides are unstable with respect to solid PbO, but where PbO has no significant vapour pressure. These results have been confirmed in three engine tests where catalysts were positioned in the exhaust train to give inlet temperature ranges of 455 to 860°C, 245 to 720°C and 155 to 540°C, respectively. The coolest catalyst position was most detrimental to catalyst performance and resulted in conversion efficiencies of < 30 per cent for both carbon monoxide and hydrocarbons after only 100 hours running on a 5 mode ageing cycle. Although the other catalyst positions resulted in similar performance (> 90 per cent carbon monoxide, 35 to 55 per cent hydrocarbon conversion) after 200 hours ageing, the position closest to the exhaust manifold (455 to 860°C) was slightly inferior to the intermediate position (245 to 720°C), providing further evidence of the effect of volatile PbO at high temperatures.

A second parameter which can affect the lead tolerance of catalysts is that of engine tune, since this can influence the type of lead compound in the exhaust stream. It would be expected that tuning rich of stoichiometry would favour the stability of lead halides and hence would enhance chemical poisoning by these species. Lean tuning, on the other hand, would favour lead oxide formation, which, provided it was not volatilised, would result in slower physical poisoning. Pulse flame reactor tests tended to confirm these predictions, since at air : fuel ratios of 12.9, 14.6 and 16.1, rates of loss of conversion for methane of 9.4, 6.6 and

*Fig. 5 A first stage lead tolerant catalyst, after ageing at 650°C in a pulse flame reactor burning a fuel containing 0.1 grams per litre lead and Motor Mix, is substantially superior to a standard U.S. oxidation catalyst*



4.1 per cent per hour, respectively, were observed.

In the successful operation of oxidation catalysts it is important to ensure overall oxidising conditions at the catalyst inlet at all times. This is normally accomplished via secondary air injection from an air pump or a Pulseair system (14). In the case of lead tolerant oxidation catalysts it is again necessary to provide sufficient secondary air to cope with peaks in emissions, but excessive use of secondary air can be detrimental. High levels of secondary air promote the conversion of lead halides to lead oxide in the exhaust and therefore may reduce chemical poisoning but increase the possibility of physical blocking or plugging. The conversion of sulphur dioxide to sulphur trioxide is also promoted by excess oxygen and hence lead deposited on the catalyst is more readily "fixed" in the form of stable lead oxy-sulphates or lead sulphate. Finally, secondary air can also exert a "flash cooling" effect on the exhaust at the point of injection, which may enhance the condensation of particulate lead, which in turn can lead to plugging. White and Zakrajsek have also reported the effect that secondary air has on the increase in weight of the catalyst as lead is deposited (15).

Thus, it is important, in developing a lead tolerant catalyst system, to optimise the design in terms of catalyst inlet temperature range, catalyst position, engine tune and secondary air

injection, as well as the catalyst itself, in order to maximise durability.

## Lead Tolerant Catalysts

In seeking to improve on the lead tolerance of standard oxidation catalysts, the approach adopted by Johnson Matthey has been to study each of the important catalyst parameters in turn in order to establish the effect on performance in a leaded environment. These parameters include substrate type, for example monolith or pellet, monolith material and cell density, washcoat type and loading, presence of washcoat stabilisers, base metal components, noble metal components and loading, and also dispersion of active sites. By optimising these parameters, a first generation lead tolerant system was identified which is based on a pure platinum catalyst supported on a Fecralloy® metal substrate. This catalyst exhibited an excellent result in a pulse flame reactor test compared to that obtained for a standard U.S. oxidation catalyst, as is evident in Figure 5.

Following this result, the catalyst was subjected to static engine durability testing, using a slave test vehicle to carry out the European ECE 15 test schedule at various points. This test was also successful with the lead tolerant catalyst giving 90 per cent carbon monoxide and 45 per cent hydrocarbon conversion after 300 hours ageing on 0.4 g/l lead Motor Mix fuel, compared to 75 per cent and 15 per cent,



*Fig. 6 Lead tolerant catalyst systems have been tested for road durability and catalyst activity on a BL Cars' 1.7 litre "O" series Morris Marina. Here a vehicle is undergoing the ECE 15 test procedure on a rolling road dynamometer where the exhaust emissions produced during a specified driving schedule are continuously measured. Test vehicles fitted with the first stage lead tolerant catalyst have been run on fuel containing 0.4 and 0.15 grams per litre of lead, the maximum and minimum amounts permitted by European Economic Community regulations*

Photograph courtesy of BL Cars Limited

respectively, for a standard U.S. catalyst after only 150 hours ageing. On the basis of this result it was decided to submit the catalyst to road durability testing.

Two such trials were conducted at BL Cars using 1.7 litre "O" series Morris Marina cars running on fuel containing 0.4 and 0.15 g/l lead as Motor Mix, Figure 6. The Automotive Manufacturers Association (AMA) cycle was used for endurance, with ECE 15 emissions testing being conducted at specific intervals. The European emissions test (the ECE 15 test), like the U.S. Constant Volume Sampling (C.V.S.) test, simulates an urban driving cycle, and the exhaust gas is continuously sampled and collected while the vehicle is driven on a rolling road dynamometer. Unlike the American test, the driving schedule is simpler and involves a maximum speed of only 40

m.p.h., compared to 55 m.p.h. for the C.V.S. test, resulting in relatively low exhaust temperatures. The exhaust gases collected are analysed for hydrocarbons, carbon monoxide and oxides of nitrogen and the results are expressed as grams per test emissions. Thus, the legislation proposed by West Germany would require carbon monoxide emissions of <30 grams per test and combined hydrocarbon and nitrogen oxide emissions of <10 grams per test for vehicles in the 750 to 1,250 kg weight range. No durability limit for emission control devices has yet been set although it is assumed that at least 50,000 km will be required.

Catalyst inlet temperatures on the two vehicles were adjusted to 400 to 450°C by the use of a single insulated downpipe which allowed the catalyst (4" diameter x 6" long) to be fitted immediately after the manifold. The

Fuel additives	Distance km	Per cent conversion		ECE 15, g/test	
		HC	CO	(HC + NO <sub>x</sub> )	CO
0.40 g/l lead Motor Mix	37,000	52	57	10.53	30.76
0.15 g/l lead Motor Mix	50,000	60	74	9.10	27.73
UBA proposed limits				10	30

trial using 0.4 g/l lead Motor Mix successfully completed 48,366 km before termination due to catalyst blocking. Deposition of particulate lead compounds appeared to be aided by a high secondary air flow rate from the air pump. However, the trial was very encouraging with both carbon monoxide and hydrocarbon conversion efficiencies levelling off at around 50 to 55 per cent after 37,000 km, the last point of measurement. ECE 15 g/test figures were very close to those proposed by the West German UBA for future legislation.

The second road trial, using 0.15 g/l lead fuel, completed 50,000 km without monolith cell plugging occurring and the final catalyst performance over the ECE 15 test at 50,000 km was very encouraging. Hydrocarbon and carbon monoxide conversions of 60 and 74 per cent, respectively, were achieved and the levels of hydrocarbons, nitrogen oxides and carbon monoxide were inside the UBA proposed limits, illustrated in Table II.

These results on a Stage I catalyst system show that there is no insuperable technical barrier to the production of catalysts capable of meeting proposed European legislation for 50,000 km road usage on leaded fuel. This catalyst system is presently being evaluated by the leading European motor companies.

Meanwhile Johnson Matthey has embarked upon a further research programme aimed at incorporating new catalyst technology into a Stage II system and engine tests are underway. Critical parameters such as catalyst position, inlet temperature, engine tune and secondary air flow rates are being optimised in conjunc-

tion with BL Cars. Areas requiring special attention include the blockages observed at the higher lead level and the position of the catalyst in relation to the low temperature ECE 15 test as well as the high temperature conditions sometimes encountered in countries with no motorway speed limits.

#### Acknowledgements

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