Gas Turbine Emission Control

PLATINUM AND PLATINUM-PALLADIUM CATALYSTS FOR CARBON MONOXIDE AND HYDROCARBON OXIDATION

By G. W. Cordonna, M. Kosanovich and E. R. Becker

Johnson Matthey, Catalytic Systems Division, Wayne

Platinum-rhodium catalysts have proved to be effective in removing carbon monoxide and hydrocarbons from gas turbine exhaust. A new generation of oxidation catalysts with improved activity and thermal stability has been developed and is now available commercially. The performance of these catalysts in the oxidation of carbon monoxide, hydrocarbons and sulphur dioxide is reported.

Growing concern about the contribution of exhaust emissions from stationary engines to environmental pollution has resulted in increasingly severe limits being proposed for nitrogen oxides, carbon monoxide, hydrocarbons and sulphur oxides emissions. During the past 15 years catalytic control of atmospheric pollutants from automotive engines has been successfully applied in the U.S.A., Japan, Australia and some European countries, and has resulted in a large reduction of atmospheric pollutants. In the U.S.A., for example, the emission of hydrocarbons to the atmosphere has been reduced by over 50 per cent, from approximately fourteen million tons per year in 1976 to less than seven million tons per year in 1988. However, the remarkable success of this pollution control technology has not been mirrored in emission abatement from stationary sources. More than half of the man-made carbon monoxide, hydrocarbons and nitrogen oxides reaching the atmosphere are emitted from stationary sources, such as gas turbines, stationary internal combustion engines, power station boilers, process heaters and industrial boilers (1). Rigorous limits on permissible levels of nitrogen oxides, carbon monoxide and hydrocarbons from such sources have now been imposed by legislation in many American states, in Japan and in West Germany.

Stationary source combustion processes generally operate with a large excess of air to maximise fuel economy, and thereby minimise emissions of partial oxidation products such as carbon monoxide and hydrocarbons. However, one of the strategies for controlling nitrogen oxides involves the injection of water or steam into the combustion zone to lower the combustion temperature. This "wet firing" reduces nitrogen oxide levels by approximately 50 per cent, but results in increased amounts of carbon monoxide and hydrocarbons in the turbine exhaust emissions. Flue gas treatment is therefore required to lower carbon monoxide and hydrocarbon concentrations to permissible levels. Catalytic oxidation technology, developed for mobile source control, has been adapted to reduce both carbon monoxide and hydrocarbon emissions from gas turbines, and has been reported here previously (2). Catalytic reactor design, the use of large metal monolithic honeycomb arrays, and the performance of a Johnson Matthey platinum-rhodium catalyst for emission control were all discussed in detail. This paper presents catalyst performance data regarding the next generation of Johnson Matthey oxidation catalysts.

Catalyst Design

Current commercial requirements necessitate the successful operation of the catalytic element without replacement for 20,000 hours or more, in the temperature range 250 to 650°C. To achieve this the reactor must incorporate a high activity catalyst, to maximise reactor productivity while minimising catalyst volume and

cost. Although some base metal oxides such as cobalt(III)oxide, copper chromite and chromium(III)oxide are active for carbon monoxide and hydrocarbon oxidation reactions, noble metals display the highest intrinsic catalytic activity (3, 4) and are well suited for applications involving high exhaust velocities, such as those found in gas turbine applications. Furthermore, platinum group metals also exhibit higher activity for saturated hydrocarbon oxidation, are less deactivated by fuel sulphur at temperatures below 500°C, and are also more thermally-resistant to loss of low temperature activity than base metal oxide catalysts (5).

When regulations restrict the sulphate particulate content of the flue gas, the oxidation catalyst can be formulated to minimise sulphur dioxide oxidation while maintaining high activity for carbon monoxide and hydrocarbons removal. Rhodium exhibits relatively low catalytic activity for sulphur dioxide oxidation and can be combined with other platinum group metals to suppress sulphur trioxide formation (2). Palladium is also a good oxidation catalyst for carbon monoxide and hydrocarbons, and in addition it displays low activity for sulphur dioxide oxidation (6).

The New Generation Catalysts

Catalyst samples were prepared on metal honeycomb monoliths having a diameter of 1 inch, a length of 3.5 inches and a cell density of 200 cells per square inch. The substrates were then coated with a high surface area alumina and catalysed with solutions of platinum, palladium and rhodium. These samples were dried and calcined prior to testing.

Catalyst performance was measured in an integral, tubular reactor with a simulated turbine exhaust gas consisting of 10 ppm propane, 45 ppm carbon monoxide, 15.0 per cent oxygen, 4.5 per cent carbon dioxide, 40 ppm nitric oxide, 10 ppm sulphur dioxide, 5.0 per cent water and the balance nitrogen. Sophisticated analytical instruments monitored the low ppm concentrations of the inlet and outlet gases. Carbon monoxide and propane concentrations are measured by non-dispersive infrared and
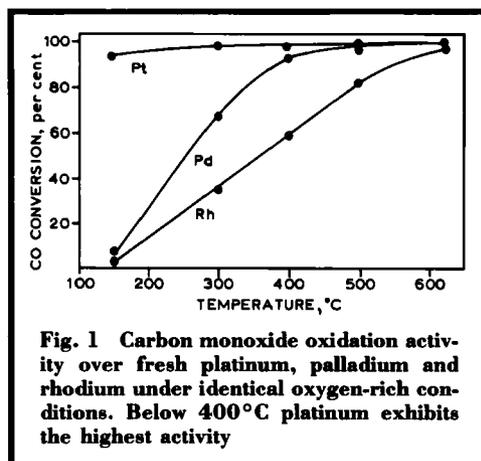

Fig. 1 Carbon monoxide oxidation activity over fresh platinum, palladium and rhodium under identical oxygen-rich conditions. Below 400°C platinum exhibits the highest activity

flame ionisation, respectively. Catalyst efficiency was measured as a function of the catalyst metal, metal loading and thermal ageing.

Carbon Monoxide Oxidation

Platinum, palladium and rhodium are the noble metals most frequently used in exhaust gas treatment catalysis. Metal oxide volatility and availability preclude the use of ruthenium, iridium and osmium. Carbon monoxide oxidation activity for comparable loadings of platinum, palladium and rhodium catalysts under identical reactor operating conditions is contained in Figure 1. For these oxygen-rich conditions, platinum exhibits the highest activity for carbon monoxide oxidation at temperatures below 400°C. The light-off temperature, defined as the temperature required to achieve 50 per cent conversion, occurs below 150°C for a fresh platinum catalyst. To achieve light-off the palladium catalyst requires a temperature of 250°C, and the rhodium catalyst in excess of 350°C. Both palladium and rhodium display lower activation energies and lower intrinsic catalytic activities than platinum for carbon monoxide oxidation; however, the palladium catalyst performs as well as the platinum catalyst at temperatures above 400°C. While studies utilising catalytic wires have shown palladium to be a better carbon monoxide oxidation catalyst than platinum (5), these results were obtained with relatively
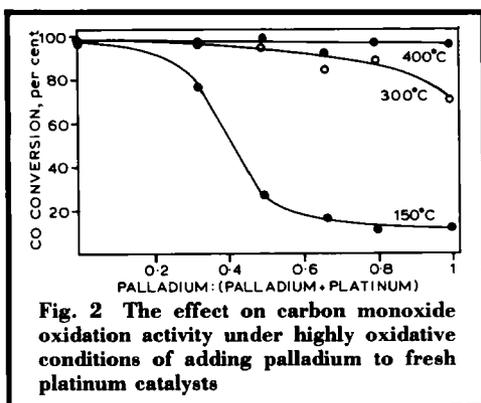
Fig. 2 The effect on carbon monoxide oxidation activity under highly oxidative conditions of adding palladium to fresh platinum catalysts

high carbon monoxide concentrations of 1 volume per cent, 2 volume per cent oxygen, and in the absence of water. Under the simulated conditions of a gas turbine flue gas described here, the platinum catalyst is by far the best carbon monoxide oxidation catalyst, provided that it has not been degraded by high temperature operation or by poisons.

In emission control and chemical process catalysis, it is quite common to use combinations of metals to provide the optimum stabilisation or activity enhancement. Examples include the well known use of platinum-rhodium catalysts for the simultaneous control of carbon monoxide, hydrocarbons and nitrogen oxide from automotive engines (7); rhenium and iridium additions to platinum reforming catalysts to increase selectivity and decrease coke formation (8,9); and a cobalt-molybdenum catalyst which offers enhanced poison resistance in hydrodesulphurisation applications (10).

Early work on the development of pelleted automotive oxidation exhaust catalysts has demonstrated that the best performance is obtained with a combination of platinum and palladium (11). For both hydrocarbon and carbon monoxide oxidation, the platinum-palladium configuration gave the best initial high temperature performance, during both steady-state and light-off conditions. The effect of palladium additions to platinum catalysts for carbon monoxide oxidation under the highly oxidative conditions of a simulated gas turbine

exhaust is shown in Figure 2. All samples tested contained identical total noble metal weight loadings, with varying proportions of the two parent metals. At 150°C, platinum and platinum-rich catalysts produce the highest levels of carbon monoxide removal, due to the higher intrinsic activity of platinum for carbon monoxide oxidation. Palladium-rich catalysts behave more like palladium, and exhibit higher light-off temperatures. At 300°C or higher the effect of palladium is minor, and platinum-palladium catalysts give carbon monoxide conversion comparable to that of platinum-only catalysts. Consequently, to achieve high carbon monoxide removal rates when using platinum-palladium catalysts, an operating temperature in excess of 300°C is necessary.

The effect of total noble metal loading on carbon monoxide oxidation is illustrated in Figure 3. Increasing the platinum loading four-fold has no significant effect on the activity of fresh catalyst, at 300°C or higher. Catalyst activity is high enough to cause the reaction to be mass transfer-limited; that is the reaction rate on platinum is much faster than the rate of diffusion of carbon monoxide through the boundary layer separating the solid catalyst from the gas phase. However, as will be shown below, platinum catalytic activity degrades significantly during high temperature thermal treatment, necessitating the use of higher platinum
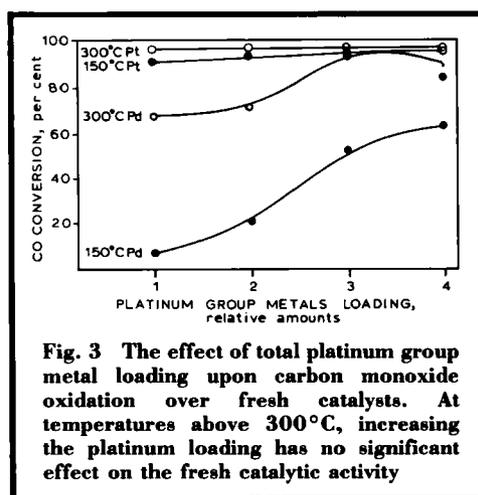

Fig. 3 The effect of total platinum group metal loading upon carbon monoxide oxidation over fresh catalysts. At temperatures above 300°C, increasing the platinum loading has no significant effect on the fresh catalytic activity

loadings to compensate for this loss of catalytic activity.

The dependence of carbon monoxide conversion on palladium metal loading is also shown in Figure 3. Palladium, being less active than platinum, shows a greater carbon monoxide conversion dependence on metal loading, and the mass transfer-limited operating regime is not reached until nearly 400°C. It will be shown that palladium can also suffer loss of catalytic activity upon exposure to high temperature, but not to the same extent as platinum.

Hydrocarbon Oxidation

Hydrocarbons are inherently more difficult to oxidise than carbon monoxide. The non-methane hydrocarbons emitted in the exhaust of turbines fuelled with natural gas are predominantly ethane and propane, with minor amounts of ethylene, propylene and butanes. As emission control requirements become more stringent, the oxidative removal of these hydrocarbons becomes more crucial. For the purposes of this study, and for simplicity sake, propane was selected as a compromise between hard-to-oxidise ethane and easy-to-oxidise ethylene, propylene and butane.

Oxidation activity for propane over platinum, palladium and rhodium is compared in Figure 4. The platinum catalyst demonstrates the best propane activity of these monometallic catalysts at temperatures up to 500°C. Palladium
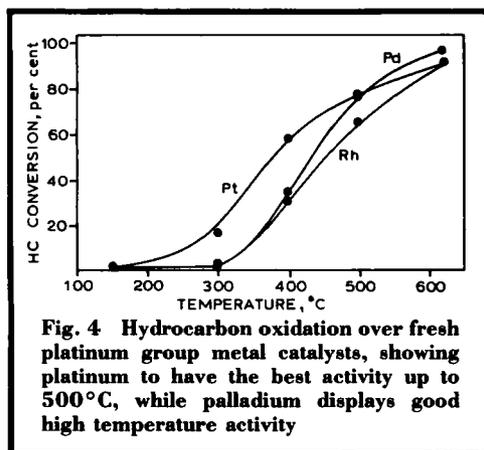
Fig. 5 **Hydrocarbon oxidation data show that combinations of platinum and palladium offer superior activity to their parent metals at the given temperatures**

displays relatively poor propane oxidation activity below 400°C, but possesses good high temperature activity. Palladium by itself is a good candidate for high temperature turbine exhaust clean-up, and is reported to be more active than platinum for the oxidation of ethane, methane and olefinic compounds (5,12). Platinum is said to be more effective for the oxidation of saturated hydrocarbons of carbon number 3 or greater (5), a conclusion which is supported by the propane conversion data presented here. Rhodium exhibits the lowest specific activity for propane oxidation but, as will be shown later, can be combined with platinum to produce an exceptionally active saturated hydrocarbon oxidation catalyst. Finally, at temperatures above 600°C these three platinum group metals all achieve high levels of propane removal.

For a gas engine or turbine exhaust containing ethane, propane and carbon monoxide, and operating at 500 to 600°C, a good catalyst choice would be a bimetallic platinum-palladium catalyst. The propane oxidation activities of four platinum-palladium catalysts and also for monometallic platinum and palladium catalysts, all at identical total noble metal weight loadings, are shown in Figure 5. For hydrocarbon oxidation, fresh catalysts containing both platinum and palladium are superior to their parent metals at each of the three operating temperatures. The combination of platinum and palladium also offers excellent

Fig. 4 **Hydrocarbon oxidation over fresh platinum group metal catalysts, showing platinum to have the best activity up to 500°C, while palladium displays good high temperature activity**
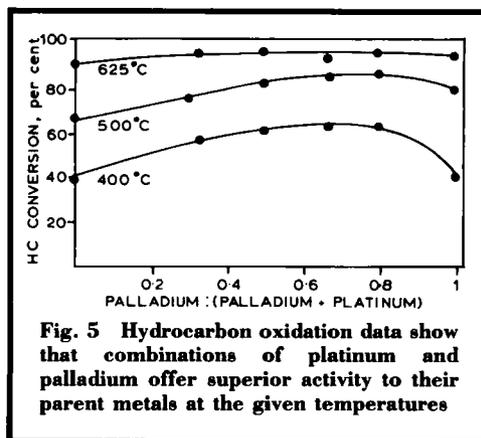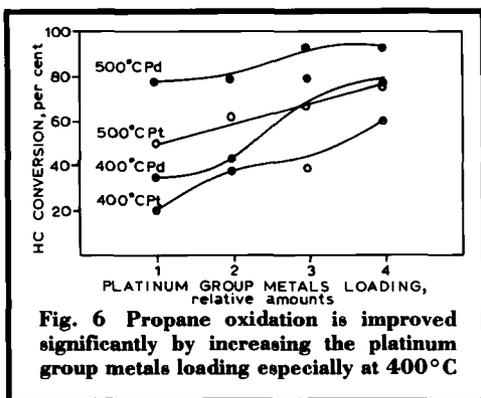
Fig. 6 Propane oxidation is improved significantly by increasing the platinum group metals loading especially at 400°C

oxidation activity for carbon monoxide and propane removal at temperatures greater than 400°C.

Hydrocarbon removal efficiency has been determined as a function of catalyst metal loading, and the data are collected in Figure 6. Propane oxidation, a kinetically slower reaction than carbon monoxide oxidation, is improved significantly by increasing the active metal loading, especially at the relatively low operating temperature of 400°C. Even at 500°C propane conversion is improved by increasing the noble metal loading, with mass transfer-limited conversion not encountered until 600°C. Experiments such as these, when combined with durability studies, allow optimisation of a catalyst formulation for a given pollution reduction requirement.

Sulphur Dioxide Oxidation

Oxidation catalysts intended for the removal of carbon monoxide and hydrocarbons should be designed to minimise the downstream formation of sulphate particulates resulting from sulphur dioxide oxidation. Therefore catalytic activity for sulphur trioxide formation should be reduced. Normally, gas turbines are fuelled with natural gas which contains only minor amounts of organosulphur compounds; however when refinery gas, which typically contains 50 to 300 ppm sulphur, or liquid fuels such as # 2 fuel oil or jet A fuel are burned, the exhaust emissions include significant quantities of sulphur dioxide.

Sulphur dioxide oxidation activity on noble metal catalysts has been reported by numerous authors. For example, Hammerle and Truex demonstrated that the oxidation activity of platinum-rhodium automotive catalysts decreased when the rhodium content was increased (6). A similar rhodium effect has been observed with a Johnson Matthey platinum-rhodium catalyst (2). Kummer, in his review of automotive catalytic converters, gives the following activity ranking for sulphur dioxide oxidation:platinum>palladium>rhodium (5).

Sulphur dioxide oxidation data obtained under gas turbine exhaust conditions is displayed in Figure 7, for monometallic noble metal catalysts of equivalent metal loading. Clearly, platinum displays the highest intrinsic activity, exhibiting nearly 80 per cent conversion at 400°C, while rhodium and palladium convert less than 10 per cent. At 500°C, rhodium achieves a sizeable level of sulphur dioxide conversion, but palladium is still relatively inactive. For high temperature catalyst operation at 625°C, all three noble metal catalysts have sufficient kinetic activity to achieve a high level of sulphur dioxide conversion, and are limited only by the thermodynamic equilibrium of the reaction. For catalyst operation at 550°C or lower, platinum-based oxidation catalysts can be formulated to include rhodium and/or palladium to reduce their affinity for sulphur dioxide oxidation.

Comparative Performance of New and Conventional Catalysts

In preparing effective oxidation catalysts for emission control purposes, the choice of noble metal precursors, raw materials, and fabrication technology can significantly affect both catalyst distribution and dispersion, and hence catalytic activity and durability. Johnson Matthey has been a leading contributor to progress in the environmental catalyst field for many years, and conducts an active research programme to develop new and more efficient oxidation systems, which help it to maintain its leading technological position. In the Table the performance data of the Johnson Matthey

| | | Conversion, per cent | | | | | | |
| Formulation | Relative metal loading | At 300°C | At 400°C | | | At 500°C* | | |
		CO	CO	C_3H_8	SO_2	C_2H_6	C_3H_8	SO_2
Johnson Matthey Platinum-Palladium	2.5	69	93	58	20	68	81	54
Johnson Matthey Platinum-Rhodium	1.0	98	99	83	62	37	75	39
Johnson Matthey Platinum	1.0	97	98	59	90	7	18	—
Conventionally Impregnated Platinum	1.0	98	99	51	79	10	28	63
Conventionally Impregnated Palladium	2.0	57	90	35	13	53	66	16
Conventionally Impregnated Rhodium	1.0	48	78	52	12	—	—	—

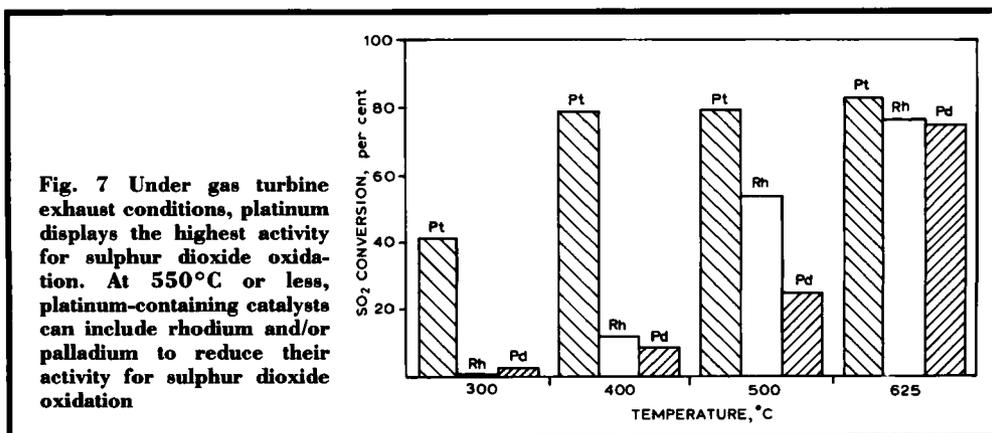*Catalysts thermally aged at 625°C for 168 or 1032 hours

proprietary catalysts and those prepared by conventional methods are compared.
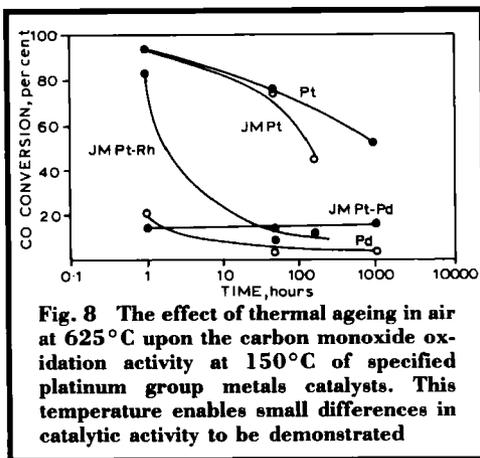
The Johnson Matthey platinum-palladium catalyst offers superior activity for ethane oxidation at 500°C when compared to conventionally-prepared oxidation catalysts, with only moderate levels of sulphur dioxide conversion. For a saturated hydrocarbon such as propane, the Johnson Matthey platinum-rhodium catalyst offers exceptional oxidation ability at 400°C, while activity for carbon monoxide oxidation is equivalent to the platinum-only catalyst systems. Furthermore, this platinum-rhodium catalyst offers relatively low sulphur dioxide oxidation behaviour, especially when thermally aged. The performance of these Johnson Matthey proprietary catalyst systems following thermal deactivation is discussed below.

Catalyst Stability

In industrial practice, several mechanisms operate to decrease catalyst activity. These include mechanical vibration, particulate abrasion, blocking of catalyst pores, thermal sintering, and poisoning by trace chemical

Fig. 7 Under gas turbine exhaust conditions, platinum displays the highest activity for sulphur dioxide oxidation. At 550°C or less, platinum-containing catalysts can include rhodium and/or palladium to reduce their activity for sulphur dioxide oxidation

Fig. 8 The effect of thermal ageing in air at 625°C upon the carbon monoxide oxidation activity at 150°C of specified platinum group metals catalysts. This temperature enables small differences in catalytic activity to be demonstrated
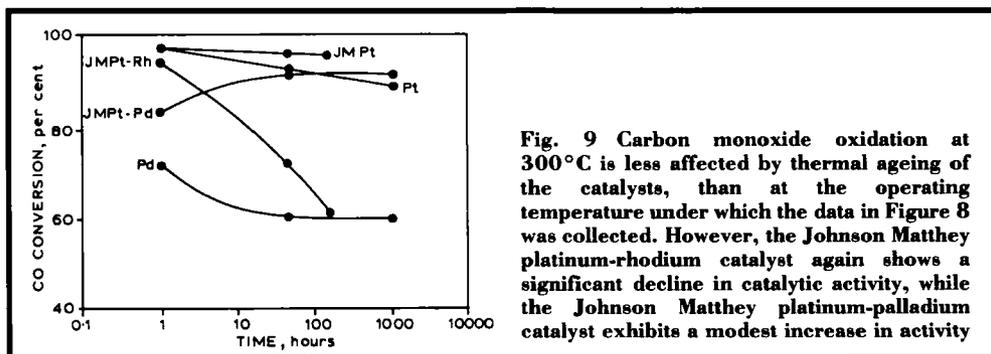
impurities. For normal turbine operation the main causes of catalyst deactivation are thermal sintering, fouling of the catalyst pores, and poisoning of the active noble metal catalyst particles.

Thermal sintering of noble metals can be significant for some modes of turbine operation, particularly when the duct burners are fired to make more steam, as then the catalyst temperature can reach 650°C. Prolonged exposure to high temperature causes agglomeration of the noble metal particles (sintering), with a consequent loss of catalytic activity due to the reduced number of catalytic sites. This loss of catalytic activity is irreversible and is most evident at low temperatures, where conversion is controlled by the rate of surface chemical reaction.

Several new Johnson Matthey catalysts and also conventionally-impregnated noble metal catalysts, all of which have been calcined in air at 625°C, are compared for carbon monoxide conversion at a temperature of 150°C in Figure 8. Although these catalysts are not designed to operate at 150°C, this temperature was selected to elucidate small differences in the catalytic activity of the various samples. At 150°C, conversion of carbon monoxide is controlled by the surface reaction rate, and losses in catalytic surface area affect the observed conversion level. This is most evident for the Johnson Matthey platinum-rhodium catalyst which sinters appreciably during thermal treatment, as witnessed by the drop in conversion from 85 per cent to approximately 10 per cent. Both the Johnson Matthey platinum and the conventionally-impregnated platinum catalyst also suffer loss of catalytic activity following thermal treatment. The conventionally-impregnated palladium catalyst, which shows only 22 per cent fresh carbon monoxide conversion, shows a rapid drop in the first 48 hours and then stabilises at a low conversion level of 5 per cent. The Johnson Matthey platinum-palladium catalyst offers the lowest fresh carbon monoxide conversion at 14 per cent, but shows no sign of thermal deactivation.

In general, emission control catalysts are designed to operate at temperatures where small changes in catalyst activity do not significantly affect conversion. At higher operating temperatures, where external mass transfer of the carbon monoxide and hydrocarbons to the catalyst surface controls the overall

Fig. 9 Carbon monoxide oxidation at 300°C is less affected by thermal ageing of the catalysts, than at the operating temperature under which the data in Figure 8 was collected. However, the Johnson Matthey platinum-rhodium catalyst again shows a significant decline in catalytic activity, while the Johnson Matthey platinum-palladium catalyst exhibits a modest increase in activity

conversion, the drop in conversion due to loss of catalytic activity will be much less. This is illustrated in Figure 9, which shows carbon monoxide removal activity at 300°C for the selected catalyst systems. With the exception of the Johnson Matthey platinum-rhodium catalyst, all the thermally treated catalysts display only a modest decline in catalytic activity at 300°C, due to the thermal treatment. The palladium-only catalyst exhibits a fresh carbon monoxide conversion of 72 per cent, which declines to 60 per cent after more than 1,000 hours of thermal ageing. It is expected that palladium should be more resistant to thermal sintering than platinum; the interaction between palladium(II)oxide and the alumina surface being sufficient to give considerable thermal stability to palladium/alumina catalysts in an oxidising atmosphere (11,13).

Interestingly, the Johnson Matthey platinum-palladium catalyst actually improves its carbon monoxide oxidation activity, possibly due to surface enrichment of alloy particles with platinum. Following thermal stress, the Johnson Matthey platinum-palladium catalyst shows equivalent carbon monoxide oxidation activity at 300°C to the conventionally-prepared platinum catalyst. Both platinum-only catalysts experience some loss in catalytic activity at 300°C, but not as severe as that exhibited by the platinum-rhodium catalyst. Although the Johnson Matthey platinum-rhodium catalyst is susceptible to a substantial loss in low temperature carbon monoxide oxidation activity, it maintains excellent activity for saturated hydrocarbon removal following thermal treatment, as described below.

The effect of thermal ageing on catalyst performance for hydrocarbon removal is illustrated in Figure 10. Both the new Johnson Matthey and the conventionally-prepared platinum catalysts suffer extensive deactivation for propane removal during the first 48 hours of thermal treatment. Johnson Matthey platinum-palladium and platinum-rhodium catalysts, along with the conventionally-prepared palladium catalyst, demonstrate excellent durability for propane oxidation following thermal ageing. These durability experiments, when conducted for different times and temperatures, form the basis of a thermal deactivation model which allows catalyst performance to be projected during commercial operation.

Conclusions

Oxidation catalyst technology, originally developed to control automotive exhaust pollutants, has been refined and adapted to eliminate carbon monoxide and hydrocarbon emissions from gas turbines and other stationary engines. Catalysts formulated from platinum, palladium and/or rhodium can be tailor-made to maximise carbon monoxide and hydrocarbon removal, minimise sulphur dioxide oxidation and maintain catalytic activity for thousands of hours of high temperature operation. Currently, Johnson Matthey has more than seven years commercial operating experience with large catalytic arrays for the treatment of gas turbine exhaust. Increasing concern about, and the regulation of, atmospheric pollution will provide further opportunities for applying this reliable platinum group metals technology in Europe, the United States of America and other industrialised areas of the world.
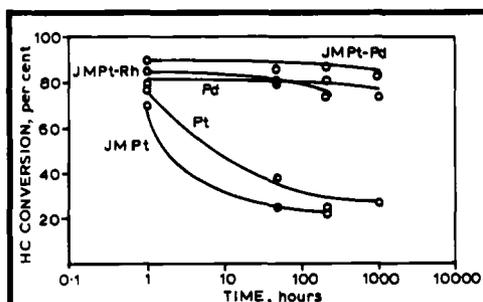

Fig. 10 The effect of thermal ageing in air at 625°C upon the hydrocarbon oxidation activity of various platinum group metals catalysts at 500°C. Excellent durability for propane oxidation can be obtained after thermal ageing with both the Johnson Matthey platinum-palladium and platinum-rhodium catalysts, and also with conventionally prepared palladium

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Magneto-Optical Data Storage Materials

Extensive work is being undertaken on a variety of thin film materials which may be suitable for magneto-optical recording applications. During the information storage process small magnetic domains in a perpendicularly magnetised layer have their direction of magnetisation reversed, while subsequent retrieval is facilitated by the optical readout of changes occurring in the linear polarisation of light reflected from the various domains on the magnetic surface.

The necessary properties of materials to be used for this purpose include: a suitably high polar Kerr rotation, high intrinsic anisotropy perpendicular to the film plane, sufficiently high coercive force, good reflectivity and corrosion/oxidation resistance. A platinum-manganese-antimony intermetallic compound with a $C1b$ phase has attracted considerable interest for this application, partly because it displays high Kerr rotation at room temperature. The preparation of suitable homogeneous films has proved to be difficult, however, but a recently reported Japanese investigation has shown that the properties of PtMnSb films can be improved significantly by varying both the preparation and the annealing conditions ("The Influence of Deposition Conditions on the Magnetic Properties of PtMnSb Films", S. Ohnuma, A. Kunimoto and T. Masumoto, *IEEE Trans. Magn.*, 1988, **24**, (6), 2551–2553).

Their samples were prepared by radio frequency sputtering a 1 micrometre film onto a silica substrate, the target being pieces of platinum supported on a manganese-antimony backplate. The magnetic properties of the as-deposited films were strongly related to the pressure of the argon sputter gas, and to the temperature of the substrate during the deposition stage. Too high argon gas pressure, or too low substrate temperature failed to produce the $C1b$ structure. A suitably high readout signal requires a high Kerr rotation angle, and for all the samples this could be increased by annealing them at temperatures above 350°C, which also increased the coercive field.

Nuclear Fusion Reported

As this issue of *Platinum Metals Review* was being prepared for the press it was reported that Professor Martin Fleischmann, of the University of Southampton, England, and Professor Stanley Pons, of the University of Utah, U.S.A., had claimed to have achieved controlled nuclear fusion within a simple electrochemical cell. The process apparently depends upon the notable ability of palladium to absorb hydrogen/deuterium. Their experiment was carried out in an insulated flask containing deuterium oxide (heavy water). Electrolysis of the heavy water using a palladium cathode and a platinum anode splits the heavy water into oxygen and deuterium, and the latter is absorbed by the palladium. In fact it is claimed that so much deuterium enters the palladium lattice structure that the deuterium nuclei begin to fuse together, releasing large amounts of heat, which could be used, for example, for the generation of electricity.

The efforts currently being made to repeat the experiment described by Professors Fleischmann and Pons will be followed with the greatest interest, as their work seems to offer the possibility of a major new source of energy, which should be environmentally compatible.