Catalytic Combustion Applied to Gas Turbine Technology

HIGH TEMPERATURE USE FOR METAL SUPPORTED PLATINUM CATALYSTS

By B. E. Enga and D. T. Thompson

Group Research Centre, Johnson Matthey & Co Limited

The emissions produced by gas turbine engines driven by catalytic combustion can contain significantly lower levels of noxious gases than those exhausted from conventional flame fired engines. The research reported here has demonstrated that efficient power generation, combined with minimum noxious emission, can be achieved by utilising metal supported platinum catalyst combustion systems specifically designed to ensure maximum compatibility with the engine models they are to power. A number of important applications for these systems are suggested.

Catalytic combustion, that is flameless combustion promoted by a catalyst, can take place with a variety of fuels ranging from high octane aviation spirit or propane gas to low BTU waste gases or residue oils. Applications for this type of combustion have been sought, and studied, at Johnson Matthey's laboratories for more than a decade. These studies have included flameless heaters, which are used for a variety of agricultural, industrial and recreational purposes (1), and particularly Honeycat® systems for the combustion of organic fumes which would otherwise cause atmospheric pollution (2). More recently the application of this principal to the production of heat from waste gases has been investigated.

Another recent development of importance has been the successful application of catalytic combustion to direct power production, for example by gas turbine engines. Work carried out over the last few years at the Johnson Matthey Research Centre in collaboration with International Research and Development Co Limited, of Newcastle-upon-Tyne, and with Sunderland Polytechnic, has been designed to demonstrate that a gas turbine may efficiently generate power when utilising a catalyst in the combustion chamber. The major advantage of such a system is that as a direct result of the lower combustion reaction temperature a much lower level of pollution is achieved, compared with that from a conventional flame fired engine. However, it is not sufficient simply to fit a catalytic unit into the combustion chamber of a conventional gas turbine engine. The chamber must be redesigned to ensure a satisfactory fluidic flow system, so as to match the catalyst to the engine.

The gas turbine catalytic combustion system developed by Johnson Matthey consists essentially of three zones through which the gases flow in succession, as shown in Figure 1. Within these zones the principal mechanisms are:

i. The preheat burner. A small fuel-rich flame burner is used to start the combustion process by preheating the inlet gases before they reach the catalyst. The design of the burner ensures that the flame does not burn the fuel completely, but is quenched by air introduced in front of the catalyst thus

Applications for Catalytic Combustion Include the Production, Conservation and Use of Pollution-Free Energy

Energy Conversion	Low grade fossil fuels and industrial waste gases can be converted to electricity or other forms of power
Pollution Avoidance	Undesirable exhaust gases from stationary, aeronautical and automotive gas turbines are inherently lower
Pollution Control	Organic emissions from industrial processes can be economically destroyed
Energy Conservation	Heat generated during the combustion of noxious emissions can be recycled for space or process heating

minimising the nitrogen oxides that are produced by the flame.

ii. The catalyst unit. Secondary fuel is injected into the heated gas stream which then passes through the catalyst unit. The pre-burner conditions are arranged so that the temperature at the catalyst inlet is sufficient to promote the combustion reaction under all operating conditions. Catalytic combustion takes place at a lower temperature than flame fired combustion and, as this is below the temperature at which atmospheric nitrogen reacts with oxygen, very low levels of nitrogen oxides result.

iii. The thermal region. To obtain

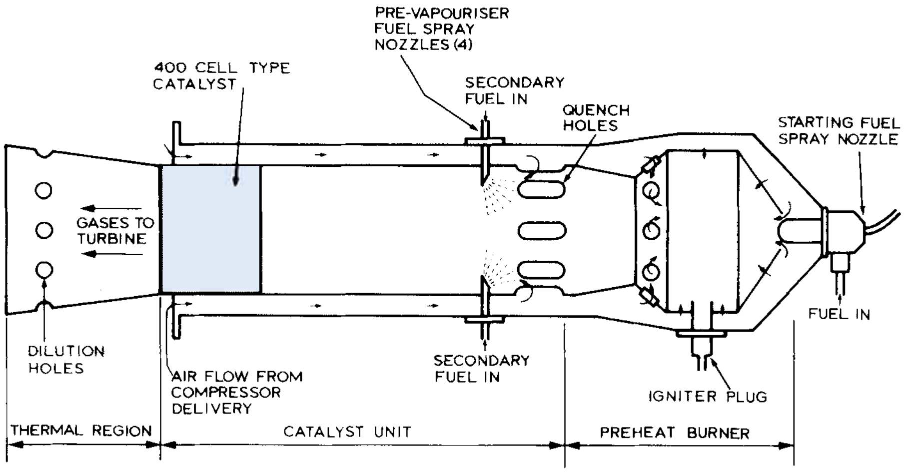

Fig. 1 A simplified schematic diagram of the Johnson Matthey catalytic combustion chamber. In the preheat burner a small amount of fuel is continually being used to preheat the inlet gases. The position of the metal supported platinum catalyst depends on the type used, but in this application 68 per cent of the delivery air flows through it, the remainder entering the system through the dilution holes in the thermal region. If a "bluff body" is required to produce an ultra high temperature this is located axially between the catalyst and the dilution holes

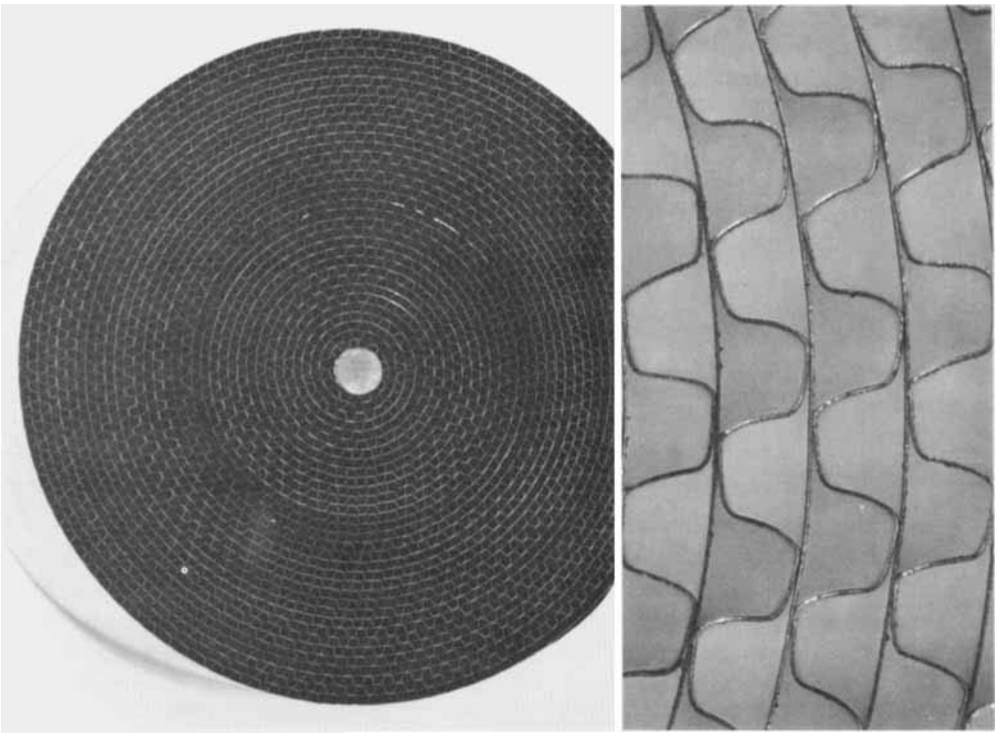

Fig. 2 Metal supports for the platinum catalysts are made from Fecralloy® steel, a material with exceptional oxidation resistance in the temperature range 700 to 1200°C. The thin metal sections have a low thermal mass and therefore a short heat-up time and, despite the high cell density, result in only a low back pressure. Part of the end section of a 400 cell type support is reproduced here at approx. full size and ×10. This type has 400 passages through each square inch of cross-section; thus a 100 mm diameter support 76.2 mm long has a catalyst support surface area of 1,356,480 mm²

temperatures in excess of those at which the catalyst support fails, the system may be modified so that the gas leaving the catalyst still contains some unreacted fuel. In the thermal region, combustion of this fuel is completed by gas phase free radical chain processes, and this may be assisted by the insertion of a "bluff body" to create a zone in which the gases recirculate. Thus the final exotherm of heat is obtained after the catalyst.

The Basic Approach

Both theoretical and practical approaches were used in the design of this catalytic combustion system, which is being developed initially for industrial gas turbine engines. The simple gas turbine cycle comprises compression of the working fluid, the addition of energy at a constant pressure and then the expansion of the working fluid. Energy losses in the system must include compression and expansion inefficiencies and pressure losses; and above atmospheric pressure work lost in the exhaust:

$$\text{Efficiency} = \frac{\text{Expansion work} - \text{compression work}}{\text{Energy supplied}}$$

A full consideration of all the many components that contribute to this equation showed that most of the variables were in fact controlled by engine design, the combustion efficiency and the pressure drop through the combustion chamber being the only factors which the catalyst process could affect. For an ideal and theoretically acceptable engine the combustion efficiency must be the same for both conventional and catalytic combustion engines, and therefore in a change from flame combustion to catalytic combustion the pressure drop through the combustor must be minimal for maximum efficiency. Accordingly the first step in the experimental work was to measure the pressure drop on a typical gas turbine engine; this was found to be 4.0 per cent under acceptable working conditions.

Having determined the significance of pressure drop many forms of catalyst support were tested on a rig. From this work it was concluded that for a given cell density, cell shape and length, the most important parameter is the open area of the support upon which the platinum catalyst is distributed. As any required open area may be obtained with a suitably designed metal support system, the concept was tested without difficulty. Fecralloy® steel has been developed as a metal support for catalysts (3) and using this material it was shown experimentally that, at the necessary gas velocities, the required pressure drop of less than 4 per cent resulted from open areas of 90 to 96 per cent. Johnson Matthey metal supports of the 400 cell and 200 cell types provide such open areas, see Figure 2.

These experiments also established that the pressure drop through a catalyst support is proportional to the square of the approach velocity, and that the pressure drop of a combination of supports is not the sum of the pressure drop across each individual support. In the turbulent zone of the catalyst the friction factor for the support becomes constant, and is a direct function of the type of washcoat which attaches the platinum catalyst to the support. This is a distinct difference from similarly constructed catalyst systems developed for automobile emission control (4). These operate at lower temperatures and by a heterogeneous mechanism based on surface area—while a homogeneous free radical chain mechanism, in parallel with the surface reactions, is experienced for the systems that are described here.

A series of experimental runs and computer simulation exercises was carried out to provide a better understanding of the problems associated with engine/catalyst interactions. Gas turbine catalytic combustors normally operate under mass transfer controlled conditions, but with the relatively low flows used on start-up the mass transfer rates and kinetic control rates combine to produce a build up of surface temperature. During transient conditions the rate of conversion of the fuel may be only 50 per cent of the rate at which it is arriving at the catalyst surface. As a result the surface temperature could be approximately twice the gas temperature; the controlling parameters being approach velocity, inlet pressure and cell density.

A separate rig was used to determine the activity of the catalyst, as measured by the light-off temperature—that is the temperature at which the catalyst starts to promote the required reaction. It was observed that the activity decreased with each start-up test, and this was considerably larger than the decrease observed when a single start-up was followed by many hours of constant running. In addition it was seen that ceramic supports exhibited a wide variety of failures due to thermal shocks. No ceramic support withstood twenty start-ups and it was therefore decided to concentrate on metal support systems rather than ceramic ones (5).

For a practical engine, the ability to start-up rapidly and reliably under cold conditions is essential. Catalyst light-off requires a temperature of about 200°C, and the addition of a small amount of hydrogen to the delivered air enables the catalyst to light-off instantly, but this requires more equipment and an additional expensive fuel. An alternative is to start-up using a small flame-fired start-up combustor, then, when the engine is running satisfactorily, a catalytic combustor can take over if the compressor delivery temperature is adequate. However, this also requires additional hardware. The system adopted by Johnson Matthey consists of an air preheater located upstream from the catalyst and

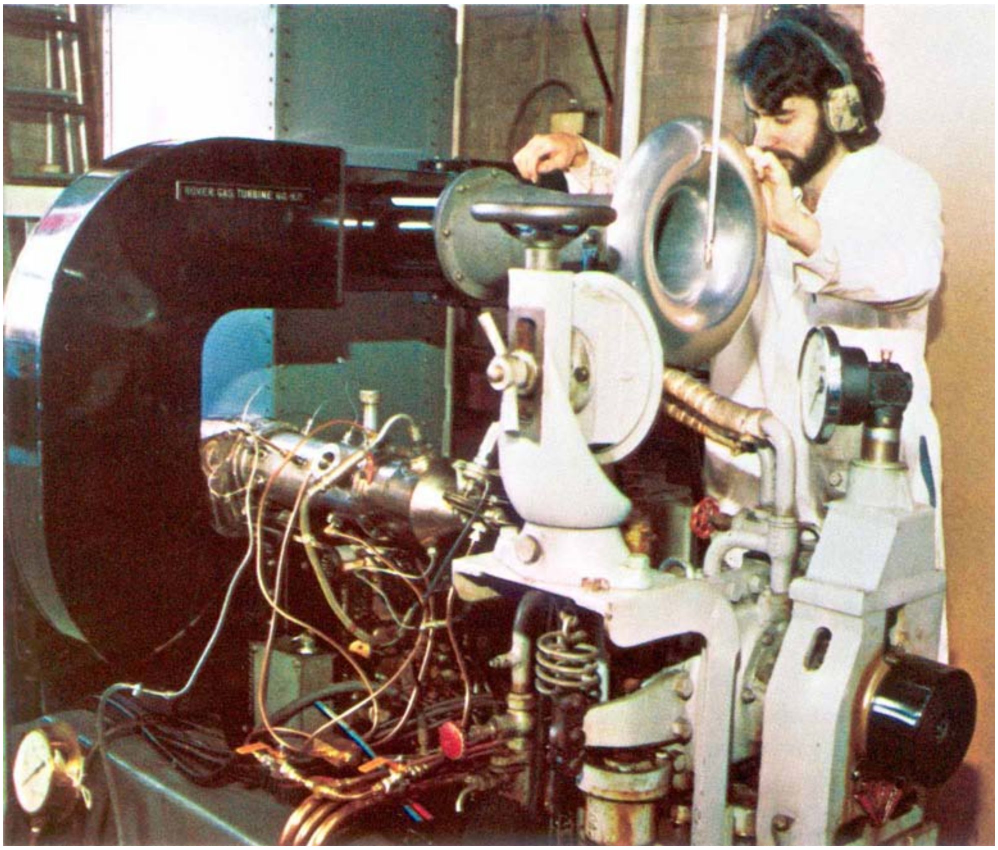

Fig. 3 The catalytic combustor mounted on a 60/65 BHP Rover engine undergoing evaluation tests at Sunderland Polytechnic. The dynamometer in the foreground enables the load to be measured while other equipment determines engine speed, temperature, pressures, fuel consumption and working fluid mass flow

operating continuously on a small amount of fuel. The main air flow is then dumped into the pre-burner flow to quench the flame and heat the feed air to the catalyst. The main fuel supply is then added and the pre-mixed fuel is passed through the catalyst, as shown in Figure 1.

Engine Tests

A Rover 60/65 BHP automotive and aircraft auxiliary generator engine was selected to test the Johnson Matthey gas turbine system. Standard test equipment was used to measure the emission levels from the engine both during normal operation and after the catalytic combustion system was fitted, see Figure 3, and the data are given in Table 1.

The range of metal and metalloid catalyst supports fabricated for the test programme consisted of two distinct classes:

Type 1. A low cost support capable of running continuously at temperatures up to 925°C. Most industrial engines do not exceed this temperature, and the majority of the test work was done using this system.

Type 2. An expensive support which may be subjected to a continuous running temperature of 1425°C. Both the construction material and the support were developed to enable advanced technology engines to be powered by catalytic combustion.

Initially the system had problems with fuel delivery but, despite rather high carbon monoxide emissions, nitrogen oxides emis-

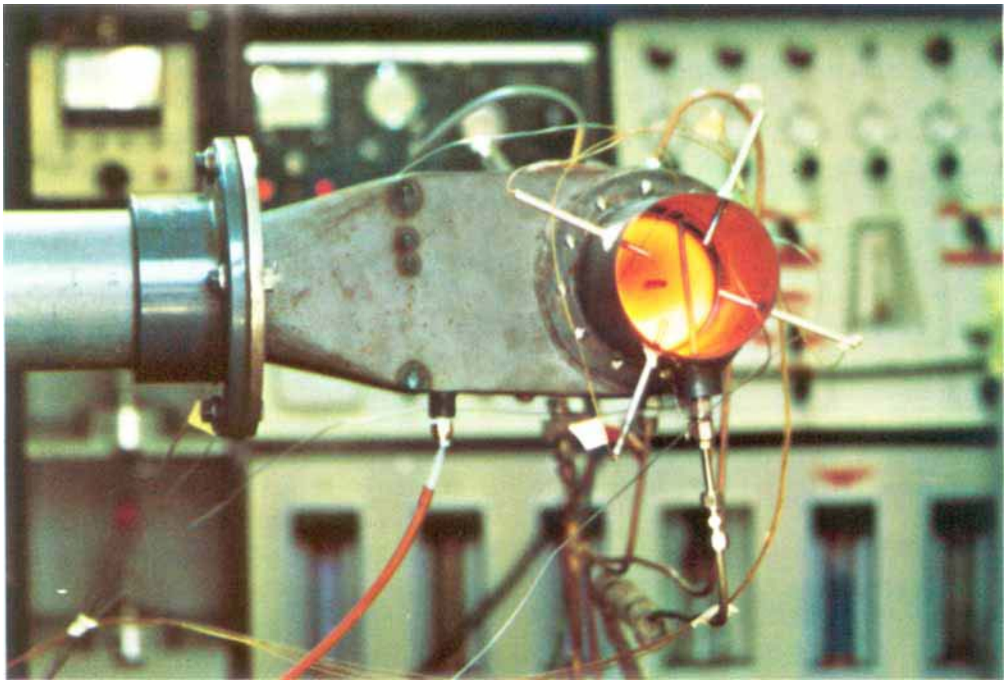

Fig. 4 A computer controlled blowing rig undergoing test at International Research and Development Co Limited. Related apparatus analyses the gas content of the emission, although here the exhaust has been removed to display the catalyst at work at a temperature of about 1020°C. Four thermocouples inserted through the dilution holes enable the heat profile of the gases leaving the catalyst unit to be established

sions as low as 0.44 ppm were obtained, compared with 58 ppm for a conventionally fired engine.

The catalyst employed was 101.6 mm diameter by 76.2 mm long; other relevant operating conditions were: gas velocity into catalyst 60 m/s, pressure drop through the catalyst 4.2 per cent, and the pre-burner delivery temperature was 420°C with 68 per cent of the delivery air flow passing through the catalyst.

Experiments were performed using a wide variety of fuels, including diesel, wide cut gasoline, kerosines, natural gas, and low BTU gas-air mixtures containing carbon monoxide-hydrogen-methane. The catalysts performed well with all of them, the only adjustments required being to the preheat burner output. It is extremely difficult to combust very low BTU fuels by conventional methods, especially if they are diluted with nitrogen, but catalytic combustion took place readily and this method is clearly an advantageous way of combusting such fuels. Low emissions of carbon monoxide, hydrocarbons and nitrogen oxides were obtained from all these tests, and this conforms with the surface initiated gas phase free radical chain mechanism shown to occur during the combustion reaction. The results with the 200 cell catalyst clearly demonstrate the difference between surface controlled catalysis, as experienced in autocatalyst systems (4), and free radically controlled combustion.

Rig Experiments

Two series of tests were run on a blowing rig, shown in Figure 4, the first used the ultra-high temperature support with a stabilised high temperature catalyst at an exit temperature of 1225°C. The performance of this unit was the same as for a normally supported catalyst, but in addition it was able to run at high exit temperatures without damage. This support was also found to be very tolerant of variations in pre-mix fuel quality, and suffered no damage when a fine spray of liquid fuel was used instead of vapourised fuel.

The second series of tests involved running a standard catalyst unit to provide continuously an exit gas at a temperature of 825°C, and then adding further fuel before passing the gases through an extremely small high temperature support. In this mode the high temperature support was essentially acting as a bluff body and the textbook phenomenon of hot body ignition could be observed.

This hot body ignition led to an interesting series of experiments during which this small support was fed with exhaust from a standard catalyst with, or without, additional fuel. The basic findings confirmed the results of work carried out both by the Surface Combustion Company and Von Elbe from the 1930's onwards (6), and in addition the application of Johnson Matthey's technology contributed the feature of low surface temperature surface-induced gas phase reactions. The fundamental principle demonstrated right from the outset of this work was "all surfaces at incandescence are equally catalytic". During the present work it was established by quenching techniques that this phenomenon is due to catalytic surface and radiation-induced oxidation of hydrocarbons giving rise to free radical precursors, such as aldehydes and carbon monoxide.

By determining the flow characteristics and fuelling patterns the most suitable position for the bluff body was determined. This position was found to be critical, malpositioning increasing the output of nitrogen oxides from the combustor.

A confirmation run of the small combustor rig was carried out. An inlet temperature to the combustor of 290°C gave an exit temperature from the conventional catalyst of 920°C, while the exit from the bluff body was at a temperature of 1370°C. This condition was held for 14.3 hours and the experiment was terminated only when, despite intensive film cooling and water injection, the high temperature caused rig damage which resulted in failure of the rig guide nozzles, and destroyed the rig pressurising valve and exhaust.

Areas of Application

Applications for catalytic combustion systems arise from two of the major demands of industry, the requirement for energy and the need to avoid atmospheric pollution. Conventional gas turbine generators produce noxious emissions as they convert chemical energy into electricity, but this does not occur with catalytic combustors. It will therefore be possible to site such generators in populated areas, where the electricity is required, thus avoiding a substantial part of the cost of power transmission. As a wide range of fuels can be combusted by catalytic means, including some which are very difficult to combust thermally, it seems probable that electrical energy will in future be generated from chemical fuels which are not considered suitable for this purpose at present.

The ease with which the Johnson Matthey catalytic combustor system can be adjusted to accommodate a wide variety of fuels offers the possibility of producing power plants capable of running on whatever fuel is available.

Underground gasification of shale oil or low quality coal tends to produce a low BTU gas. Catalytic combustion is an ideal technique for burning such gases and its development may hasten the utilisation of the world's vast reserves of these low grade fossil fuels.

In addition to converting energy into a required form without producing pollution, catalytic combustion offers the possibility of reducing some forms of industrial atmospheric pollution and at the same time producing heat to be used for process or space heating. The power from the turbine can also be used to run air compressors or similar plant. For example, energy recovery from waste solvents has been demonstrated in processes used for

Table I

Emission Levels Before and After Fitting Catalyst Combustion Systems to a Rover 60/65BHP Engine

Combustor	Equivalence Ratio φ *	Carbon Monoxide g/kg fuel	Hydrocarbons g/kg fuel	Nitrogen Oxides g/kg fuel
Standard engine	0.34	5.1	12.0	3.8
with flame combustor	0.38	4.9	9.2	4.2
Catalytic combustor	0.33	0.1	1.9	0.03
400 cell type	0.38	0.04	0.9	0.03
Catalytic combustor	0.34	7.5	2.2	0.03
200 cell type	0.40	0.04	1.1	0.06

* φ = fuel:air ratio divided by stoichiometric fuel:air ratio

paper coating, printing, wire enamelling, and petrochemical manufacture.

Growing environmental concern around major airports is producing a requirement for pollution free aircraft engines. The development of such engines is currently being pursued using catalytic combustion.

The elimination of noxious gases from engine emission means that it may become practicable to operate such engines in enclosed spaces such as warehouses, factories and mines where pollution cannot be tolerated.

Conclusions

The Johnson Matthey catalytic combustion system (7) can be used to generate power from a 60/65 BTU gas turbine engine. This has been confirmed by running the engine at full power for a period of sixty hours.

The use of catalytic combustion in gas turbines reduces considerably the level of carbon monoxide, hydrocarbons and nitrogen oxides emissions, compared with those produced during traditional flame fired combustion. This is a result of the lower temperature of combustion, which eliminates the nitrogen oxides formation, and the surface catalysed free radical chain mechanism for the combustion, which lowers the levels of all the pollutants.

The metal supported catalyst systems specially developed for this study have the advantages of low pressure drop, high temperature stability and high surface to volume ratios.

A wide variety of problems concerned with energy conversion and pollution control are likely to be solved, or reduced, using metal supported platinum catalyst combustors.

It is emphasised that the catalytic combustion system within the turbine must be designed for each particular model engine. Simply to place a catalyst within the combustor is not the best way to getting efficient catalytic combustion.

Acknowledgements

We gratefully acknowledge the helpful collaboration received in this work from Norman Searle and Derek Smith, of International Research and Development Co Limited, and from Sunderland Polytechnic.

References

1 J. D. MacConnell, *Platinum Metals Rev.*, 1972, **16**, (1), 16
2 G. J. K. Acres, *Platinum Metals Rev.*, 1970, **14**, (1), 2
3 A. S. Pratt and J. A. Cairns, *Platinum Metals Rev.*, 1977, **21**, (3), 74
4 G. J. K. Acres, *Chem. & Ind.*, 1974, (22), 905
5 B. E. Enga, "Catalytic Combustion in Actual Engines: a Summary of Engine and Rig Tests", Proceedings: Third Workshop on Catalytic Combustion, EPA-600/7-79-038, U.S. Environmental Protection Agency, Washington, February 1979, pp. 491-512
6 "Combustion, Flames and Explosions of Gases", B. Lewis and G. von Elbe, Academic Press, New York, 1961
7 B. E. Enga and A. S. Pratt, *German Offen.* 2,809,407; 1978