

Haren Gandhi 1941–2010: Contributions to the Development and Implementation of Catalytic Emissions Control Systems

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Throughout his long and distinguished career with the Ford Motor Company Haren Gandhi was concerned with controlling tailpipe pollutants from cars, and through his work this article reviews the huge amount of progress made during his four-decade career. His early work with gasoline engines embraced all of the major developments ranging from the first platinum-based oxidation catalysts through nitrogen oxides (NO_x) reduction using platinum-rhodium catalysts and the later introduction of palladium into three-way catalysts (TWCs) via 'trimetal', palladium-rhodium and palladium-only formulations. Gandhi's other work included the interactions of poisons with catalysts as part of maintaining their in-use performance, the potential for using ruthenium in NO_x control in gasoline TWCs and NO_x adsorbing catalysts (NACs) for lean-burn engines, and the use of zeolite-based selective catalytic reduction (SCR) catalysts for effective diesel engine NO_x control. Gandhi received many awards and honours in recognition of his technical achievements and a selection is mentioned here. Haren Gandhi is remembered with tremendous fondness and respect throughout the automotive industry concerned with exhaust gas emissions control and his technical contributions towards improving the quality of the air we breathe will continue to benefit us all.

Haren Gandhi: An Appreciation

Harendra Sakarlal Gandhi (Haren, as he was known) (Figure 1), was born into a large family in Calcutta, India, on 2nd May 1941 and raised in Bombay (Mumbai), the capital of the State of Maharashtra and then the most populous city in the world. He obtained a first class honours degree from the Department of Chemical Technology at the University of Bombay in 1963. Later he moved to the USA where he attended



Fig. 1. Dr Haren Gandhi photographed on 18th July 1996 prior to attending a meeting accompanied by the present author in the House of Lords with Lord Porter and Lord Lewis, to discuss the impact of fuel sulfur levels on catalytic emissions control systems

the University of Detroit, and on 15th February 1967 he joined the Ford Motor Company in Dearborn, Michigan, as a Research Engineer while still studying. In the same year he was awarded a Master of Science degree, and in 1971 he gained a doctorate in Chemical Engineering with a thesis entitled "Adsorption of Nitric Oxide on Transition Metal Oxides". By the time Gandhi had completed his doctoral research, the US 1970 Clean Air Act had been signed. This required a 90% reduction of carbon monoxide (CO) and hydrocarbon (HC) emissions from 1970 model year levels by 1975, and a 90% reduction of nitric oxide (NO) emissions from 1971 model year levels by 1976. These requirements were major challenges to the car industry, and Gandhi joined a research team dedicated to achieving them.

Haren Gandhi spent his working life with Ford, first as a research engineer and, after a number of engineering and scientific positions in the area of automotive catalysis and emissions control, as a Henry Ford Technical Fellow overseeing Ford's worldwide

emissions control projects. He had a keen awareness of the social benefits that result from improved urban air quality delivered by emissions control catalysis, and he wished to extend these benefits around the world and especially to developing countries. He began early in the catalytic control of exhaust pollutants, and his successes were driven by a love and enthusiasm for his work that was supported by always having a creative research group of excellent scientists and engineers at Dearborn, coupled with the strong technical links that he developed with leading scientists from global catalyst manufacturers. He was a visionary leader with openness to new ideas and an ability to translate science and research into innovative products that went to series production. He worked closely with US government organisations such as the Environmental Protection Agency (EPA) and the California Air Resources Board (CARB), as well as national laboratories and universities. Haren Gandhi is shown in **Figure 1** when, on one of his regular European visits, he was invited to the UK House of Lords to help clarify the detrimental effects sulfur from fuel has on the performance of autocatalyst systems.

Gandhi was always a most honourable person with a strong professional appreciation of the ethics associated with commercial dealings. Through his successes he raised the profile of all those who innovate and are responsible for introducing new technologies, especially those associated with transportation. Haren Gandhi passed away on 23rd January 2010 and he will be deeply missed by his many friends around the world, although his technical contributions towards improving the quality of the air we breathe will continue to benefit us all. Above all Haren Gandhi was a family man, and many technical meetings with him started with talk about his most recent family holiday or how his grandchildren were developing; he leaves his wife Yellow, his daughter Sangeeta and his son Anand.

Recognition and Awards

Apart from his earliest research, almost all of Gandhi's work involved the application of platinum group metals (pgms), and his successes were largely based on the tremendous catalytic activity these metals can have in optimally formulated practical catalysts. Gandhi was keenly aware of the unique roles pgms have in controlling tailpipe emissions. He often recalled the experience of visiting a pgm mine and

seeing for himself the very low levels of these metals in the ore that necessitate extensive concentrating and refining to provide pure salts of the metals for the manufacture of catalysts.

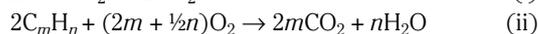
One of the earliest awards Gandhi received was the Chemical Engineer of the Year Award from the Detroit Section of the American Institute of Chemical Engineers (1984). This was followed by others including the UK Institution of Mechanical Engineers (IMechE) Crompton Lanchester Medal (1987), the Society of Automotive Engineers (SAE) Ralph R. Teeter Industrial Lectureship Award (1988–1989), the Technological Innovation Award by Discover Magazine (1990), the Award for Excellence in Catalysis by Exxon (1992) and his election to the US National Academy of Engineers (1999). He heads the list for the number of Henry Ford Technological Awards, with the last, in 2009, being for the development and integration of virtual design and optimisation of global diesel after-treatment systems. But his chief award, and most prized by him, was the US National Medal of Technology for research, development and commercialisation of automotive exhaust catalyst technology and precious metal utilisation that he received from President George W. Bush in 2002.

Contributions to Catalytic Emissions

Control Systems

Oxidation Catalysts

In anticipation of the 1975 implementation of the 1970 Clean Air Act several approaches were investigated to meet the demanding HC and CO tailpipe reduction targets. These included non-catalytic means such as running gasoline engines lean and having post-engine thermal combustion devices. But it became appreciated that the initial requirements could be met by using an oxidation catalyst in conjunction with an air pump to ensure that the exhaust was sufficiently lean (containing excess oxygen) for the oxidation of residual HC and CO to small amounts of carbon dioxide (CO₂) and water (H₂O), Equations (i) and (ii), to take place.



Base metal catalysts were inadequate in terms of long-term activity and poison resistance (1) while platinum-based ones performed well, and once sufficient durability was demonstrated catalytic oxidation was successfully introduced into production. At this

time a profound decision of lasting importance was to use coated monolithic ceramic honeycomb catalyst structures, rather than traditional catalyst pellets initially employed by some other companies which lacked long-term physical durability. The addition of a second pgm to oxidation formulations was used by catalyst manufacturers to provide improved durability, by inhibiting sintering of the very small catalytically-active platinum crystallites under lean conditions. This could be done with either palladium or rhodium and for technical reasons Johnson Matthey used small amounts of rhodium. Later this took on special significance when NO_x emissions also had to be controlled – NO_x refers to the combined NO and nitrogen dioxide (NO₂) emissions. The control of tailpipe emissions with just an oxidation catalyst only lasted a short time because the need to reduce NO_x emissions by 1976 required implementation of more complex catalyst systems.

NO_x Reduction

It was apparent that engine measures alone, such as exhaust gas recycle (EGR) used to moderate the formation of NO_x in an engine by reducing the levels of oxygen present during combustion, would not be sufficient to meet the NO_x emissions limits by 1976. It would be necessary to convert NO_x to inert nitrogen (N₂) to meet the demanding legislation, and a huge amount of research was directed to understanding how this difficult transformation could be achieved. At Dearborn, Ford was in the thick of NO_x reduction research. A key scientist there, who became very important, was Mordecai Shelef with whom Haren Gandhi worked (2, 3). The possibility of using base metal catalysts had appeal and this approach was thoroughly explored; indeed Gandhi's doctoral work was concerned with some of the relevant fundamental chemistry. Although dissociation of NO into its elements is thermodynamically favoured, under practical lean conditions this could not be done. The most active surfaces for NO dissociation are metallic, and dissociative adsorption of NO, Equation (iii), can be followed by rapid desorption of N₂, Equation (iv). However, oxygen atoms remain strongly adsorbed on the catalyst surface, and soon coverage by oxygen is complete and this prevents further adsorption of NO so its dissociation is halted. Effectively the surface is poisoned by adsorbed oxygen atoms, and under the prevailing conditions they can only be removed through reaction with a reductant, for example with

hydrogen, as illustrated in Equation (v), or with CO as in Equation (vi), to provide a clean active surface for further NO dissociation.



These equations do not imply intimate mechanistic detail, but rather highlight the key role that surface oxygen plays in inhibiting NO dissociation. Consequently, at that time NO emissions could only be controlled by reduction to N_2 in the absence of oxygen. It was found that the most catalytically active metals for this process were rhodium and ruthenium, and Gandhi explored the use of both. The reduction of NO became a theme that ran throughout his career.

Reduction/Oxidation Systems

The first practical system to control all three pollutants from cars used two separated pgm-based catalysts (4), the first reduced NOx to N_2 and the second oxidised HC and CO to CO_2 and H_2O . The upstream catalyst operated under rich conditions and, after addition of excess oxygen from an air pump, HC and CO were oxidised over the downstream catalyst. However, chemical complexities including over-reduction of NOx to ammonia (NH_3) by the first catalyst took place with some formulations, with remarkably high selectivity and high conversion (5–7). NH_3 emissions are undesirable, although any

NH_3 formed in this way would almost certainly be oxidised back to NOx over the downstream oxidation catalyst, causing significant overall NOx conversion inefficiency. To combat this, catalyst selectivity was improved and operating conditions were kept not too rich. Such two-catalyst systems for controlling HC, CO and NOx emissions were effective, but the catalyst and ancillary equipment cost was high.

Three-Way Catalysts (TWCs)

Fundamental work on conversion of the three pollutants HC, CO and NOx as a function of air to fuel ratio showed that if a gasoline engine is operated around the stoichiometric point (an air to fuel ratio of about 14.7) a single platinum-rhodium catalyst could remove the three pollutants simultaneously, and this concept was christened the three-way catalyst (TWC) (8, 9) (Figure 2). There was a strong drive for the introduction of TWCs because of improved cost and efficiency, but well-controlled stoichiometric engine operation required implementation of three new key technologies:

- An oxygen sensor to determine whether the exhaust gas is on the lean or rich side of the stoichiometric point;
- An electronic fuel injection (EFI) system to permit metering precise amounts of fuel into the engine to maintain stoichiometric operation;
- A microprocessor for closed-loop feedback control of the overall system.

By the late 1970s all these features had become available, and TWCs came into production in the early



Fig. 2. Photograph of a cut-away three-way catalyst (TWC) converter of a type much used by Ford in Europe in the early 1990s. For a long time two separate TWCs were contained in one can, perhaps reflecting the time when two separated catalysts were used with different functions

1980s as the technology of choice for controlling all three legislated pollutants (10). Subsequent addition of components such as air mass flow meters, better oxygen sensors, substantial microprocessor and software improvements and major catalyst improvements enabled TWCs to be integrated into the engine management system with remarkable results. Today TWCs can achieve almost complete removal of the three legislated pollutants, while an onboard diagnostics (OBD) system monitors their performance *via* measurement of oxygen storage capacity with two oxygen sensors. **Table I** illustrates trends in Californian

emissions limits set by CARB since 1993 (11) that for several years guided the direction of Gandhi's work. The SULEV hydrocarbon limit after 120,000 miles' driving ($0.010 \text{ g mile}^{-1}$, and significantly less than this when the vehicle is new) can be compared with the 15 g mile^{-1} of a typical mid-1960s US car to provide a direct indication of the progress made in controlling emissions over three and a half decades. Indeed, tailpipe emissions of regulated pollutants from gasoline cars can now be less than levels in ambient air, so it could be said that during Gandhi's career the situation moved from these emissions being a major

Table I

California Gasoline Emissions Standards Set by CARB (11)

Year	Category	Emissions (g mile^{-1} , FTP Test)			
		HC	CO	NOx	PM
1993	–	0.25 ^a	3.40	0.40	–
1994	Tier 1	0.25 ^b	3.40	0.40	–
2003	Tier 1	0.25 ^c	3.40	0.40	–
2004	TLEV ₁ ^d	0.125	3.40	0.40	0.08
	LEV ₂ ^{e, f}	0.075	3.40	0.05	0.01
2005	LEV ₁ ^d	0.075	3.40	0.40	0.08
	ULEV ₂ ^{e, f}	0.040	1.70	0.05	0.01
2006	ULEV ₁ ^d	0.040	1.70	0.20	0.04
	SULEV ₂ ^{e, f, g}	0.010	1.0	0.02	0.01
2007	ZEV ₁	0	0	0	0
	ZEV ₂	0	0	0	0

^aNMHC = non-methane hydrocarbons, *i.e.*, all hydrocarbons excluding methane

^bNMOG = non-methane organic gases, *i.e.*, all hydrocarbons and reactive oxygenated hydrocarbon species such as aldehydes, but excluding methane. Formaldehyde limits (not shown) are legislated separately

^cFAN MOG = fleet average NMOG reduced progressively from 1994–2003

^dLEV₁ type emissions categories phased out 2004–2007

^eLEV₂ type emissions limits phased in 2004 onwards

^fLEV₂ standards have same emission limits for passenger cars and trucks <8500 lb gross weight

^gSULEV₂ onwards 120,000 miles durability mandated

Note: TLEV = transitional low emission vehicle, LEV = low emission vehicle, ULEV = ultra low emission vehicle, SULEV = super ultra low emission vehicle, ZEV = zero emission vehicle,

CARB = California Air Resources Board, FTP = Federal Test Procedure

contributor to atmospheric pollution to one in which car use could help to reduce their levels in the air! And this was achieved with no more pgm per car than originally employed (**Figure 3**), thanks to the tireless efforts of Gandhi and other catalyst scientists in the automotive industry.

Ruthenium Catalysts

Although platinum-rhodium-based catalysts were discovered to be effective in NO_x reduction, during the search for NO_x reduction catalysts it was found that ruthenium was particularly effective and had the advantage of being cheaper than rhodium. However, the major problem encountered with ruthenium (and also to a lesser extent with iridium) catalysts is metal loss *via* volatile higher oxidation state oxides. Gandhi with his colleagues undertook research aimed at stabilising ruthenium catalysts against metal loss by forming stable ruthenates, and this was the subject of some of Gandhi's first patents (12–16). There was some success (17–19), although real-world performance and durability proved insufficient to displace rhodium from platinum-rhodium TWC formulations.

Palladium Catalysts

The pgms have outstanding catalytic activity and ruthenium was one of the cheapest for NO_x reduc-

tion, but because of durability concerns it was inappropriate for autocatalyst applications. The next most cost-effective metal was palladium, but it is very sensitive to poisoning by lead and sulfur. Unleaded gasoline was introduced to allow the use of catalysts, and when fuel sulfur levels were reduced in the early 1990s, it became feasible to incorporate palladium into platinum-rhodium catalysts to form commercially successful 'trimetal' TWCs (20). These still contained a high proportion of platinum. Palladium-rhodium formulations were later developed that actually outperformed the older, more costly platinum-rhodium TWCs. Gandhi became a strong advocate for the development of 'palladium-only' formulations that provided very good HC control and sufficient NO_x control to meet the legislative requirements of the time. Gandhi pushed catalyst manufacturers to develop palladium-only TWC formulations with exceptional thermal durability and this was achieved, allowing durable palladium-only products to enter series production in the mid-1990s (21). The increased demand for palladium caused its price to increase and this, together with more demanding legislation that required tighter NO_x control, took rhodium back into TWC formulations (see **Table I**). However, the experience and knowledge obtained in the development of palladium-only catalysts was



Fig. 3. A photograph illustrating where a three-way catalyst (TWC) can be mounted directly on the engine exhaust manifold in a modern car. In this position the catalyst can be rapidly heated when the engine is started with a suitable start-up strategy. Since it runs hotter than if it were located further from the engine, less pgm is required than otherwise would be the case. However the high temperatures mean that catalyst thermal durability is essential

transferred by catalyst manufacturers to developing advanced palladium-rhodium formulations (22) that led to remarkable TWCs for super ultra low emission vehicles (SULEVs) with astonishing performance and durability using relatively low pgm loadings (23).

Sources and Effects of Catalyst Poisons

Throughout his career Gandhi was involved with understanding the effects of poisons on automotive catalysts. In fact, the introduction of catalytic control of exhaust pollutants was only possible once unleaded gasoline was made available (for many years significant levels of toxic tetraethyl lead (TEL) were used to enhance the octane rating of gasoline). Lead is a powerful catalyst poison that blocks surface active sites and forms inactive alloy phases with pgms (24). Sulfur, a common catalyst poison, originates from gasoline and also lubrication oil (25). Other poisons derived from lubrication oil include calcium, zinc and phosphorus compounds. Phosphorus can form glassy surface layers on the catalyst that prevent gases reaching the active components (26) as well as being responsible for other problems (27, 28). Some TWCs are also susceptible to forming redox-inert cerium phosphate that degrades washcoat structure and depletes its vital oxygen storage capacity (29). Addressing these problems, which forms part of ensuring that in-use catalyst activity is maintained, concerned Gandhi. In this area he was also much troubled by the use of the lead-substitute octane enhancer methycyclopentadienyl manganese tricarbonyl, because manganese oxide deposits which form from it degrade engine performance by fouling the spark plugs and engine internals (30). In the exhaust system these deposits can inhibit the behaviour of the oxygen sensors which are instrumental in the proper running of the engine and OBD measurements. Catalyst performance can also be impaired through a surface coverage/blocking process (31–37). Recently the manganese fuel additive was reported to form ultra-fine nanoparticle tailpipe emissions (38, 39) that may themselves give rise to health concerns (40).

Lean-Burn Gasoline Engines

Over the last decade or so there has been growing interest in the reduction of greenhouse gas emissions. Widespread use of lean-burn engines, especially diesel engines (see below), with improved fuel economy could make significant contributions to lowering

emissions of the main greenhouse gas, CO₂, and starting in the mid-1990s lean-burn gasoline engines had a revival for this reason. The possibility of operating gasoline engines under lean conditions with direct injection of fuel into the cylinder has a number of attractions, including increased power and fast response as well as some fuel economy benefits. Several lean-burn direct injection gasoline engines in cars followed Mitsubishi's 4G93 1.8 litre engine which was first used in Japan and came to Europe in the Mitsubishi Carisma model in 1995. The fuel mixture is stratified in the cylinder of these engines so that it can be ignited by the spark plug, and progressively the mixture is leaner further away from the ignition point, so overall the combustion is lean. Thus the main emissions challenge was to control NO_x emissions under lean conditions. Because of these considerations, controlling pollutants from lean-burn engines became the focus of some of Gandhi's latest work at Ford.

The typical catalytic emissions control system for a direct injection lean-burn gasoline engine, illustrated

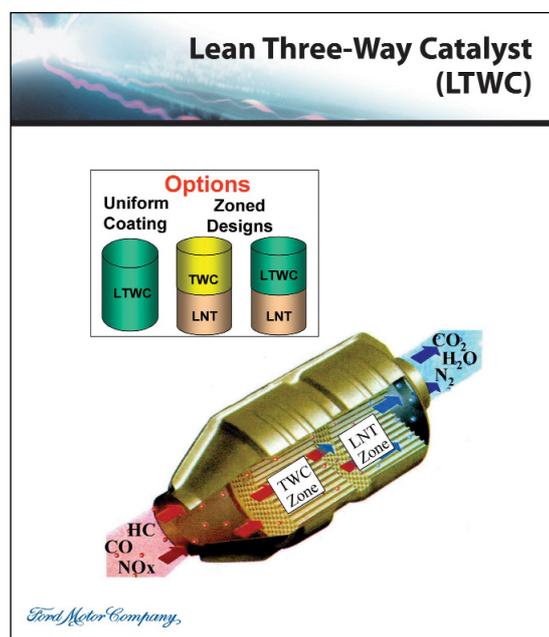
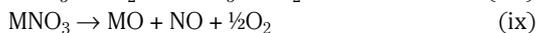
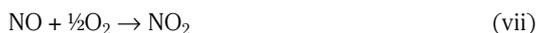


Fig. 4. A lean three-way catalyst (LTWC) system. The upstream three-way catalyst (TWC) is used when the engine runs stoichiometrically (typically when starting and during acceleration), and when the engine operates lean the TWC oxidises hydrocarbons and carbon monoxide while nitrogen oxides (NO_x) are removed by the downstream lean NO_x-trap (LNT) (Courtesy of Ford Motor Company)

in **Figure 4**, has in practice a close-coupled TWC for use when the engine runs stoichiometrically (usually when starting and during acceleration) and an oxidation catalyst to control HC and CO emissions when the engine runs lean (41). NO_x is then removed by a cooler downstream NO_x absorbing catalyst (NAC) (also known as a lean NO_x trap (LNT)) through a process involving oxidation to NO₂ over a platinum component followed by its reaction with an alkaline component to form a nitrate phase, as illustrated in Equations (vii) and (viii). Before the absorbing catalyst is saturated with nitrate the exhaust is enriched, causing the nitrate to destabilise and decompose, Equation (ix), liberating NO that is reduced to N₂ over a rhodium component in much the same way as in a TWC. Under operating conditions the alkaline component (M), usually the alkaline earth metal barium, when not nitrated is present as a carbonate, Equation (x). The overall process is called 'regeneration' and typically takes place every minute or so, with enrichment lasting around a second (**Figure 5**) (42).



In a typical Gandhi way he embraced these concepts with energy and vigour (43–45), and even returned to his desire to use ruthenium-based catalysts – one of his recent patents uniquely uses ruthenium in NAC formulations (46). He considered lean operation of gasoline engines under a variety of situations, including when idling, as a way to help improve real-world fuel economy. However, none of these approaches have yet gained widespread

acceptance, perhaps because of technical complexity and the cost of direct injection. However, the combination of an upstream NAC over which NH₃ can be formed during regeneration if the exhaust is sufficiently rich, followed by an SCR catalyst (see the section on Diesel Engines) that uses this NH₃ to reduce residual NO_x, may well gain acceptance in diesel engine NO_x control (47) as evidenced by several recent publications (48–51).

Diesel Engines

The ultimate lean-burn concept is the diesel engine, and during the last decade Gandhi gave attention to the control of diesel engine emissions, especially NO_x (52). In the 1990s major diesel engine developments took place in Europe. The bottom-end torque and driving characteristics of new turbocharged direct injection diesel-powered vehicles, coupled with their outstanding fuel economy (low CO₂ emissions) resulted in them becoming very popular. Around half of all new passenger cars in Europe are currently diesels. The two most difficult to control diesel engine emissions are particulate matter (PM) or soot, and NO_x. By 2000 diesel particulate filters in combination with fuel-borne catalyst (to enhance periodic burning of PM retained in the filter) were introduced on some European cars (53, 54). This was followed by the introduction of catalysed particulate filters that do not need the fuel-borne catalyst and its associated equipment. By 2005, small compact catalysed filters had been developed that fit directly on the engine's turbocharger and perform all the catalytic and filtration functions necessary to control HC, CO and PM (55).

Diesel NO_x control has all the problems associated

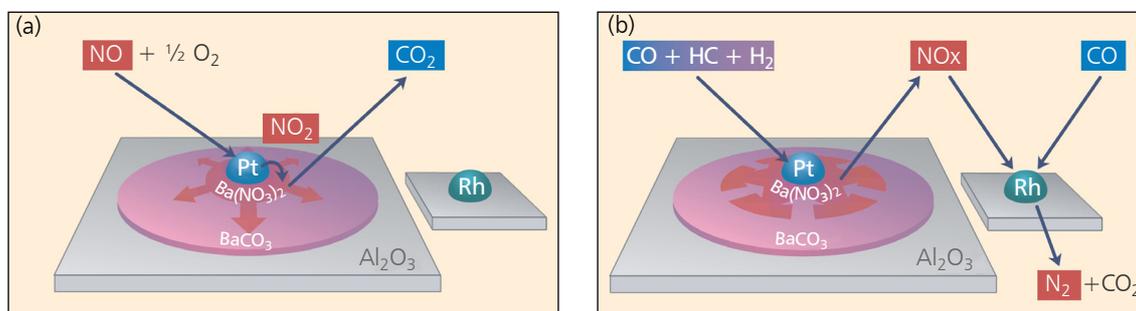
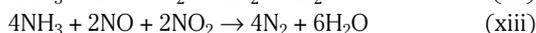
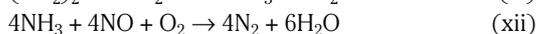
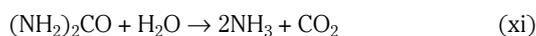


Fig. 5. Schematic operation of a lean NO_x-trap (LNT) or NO_x adsorber catalyst (NAC): (a) NO_x is removed from the lean exhaust gas by adsorption onto platinum sites where it is oxidised to NO₂ and then converted to a solid nitrate phase; (b) When the exhaust gas is enriched, the NAC NO_x capacity is recovered by releasing NO_x that is reduced to nitrogen over rhodium catalyst sites (42)

with lean-burn direct injection gasoline engines, exacerbated by lower-temperature operation. Formulation modifications enabled NACs to be used on diesel engines (56), but Gandhi generally favoured selective catalytic reduction (SCR) of NO_x with NH₃ derived from hydrolysis of urea supplied as an aqueous solution, Equation (xi). NO and NO₂ react differently with NH₃, Equation (xii) and Equation (xiii), and equal amounts of both react faster than either individually, so the SCR catalyst is usually located downstream of an oxidation catalyst to adjust the NO:NO₂ ratio for optimum NO_x control performance.



Since the 1970s SCR technology has been used to remove NO_x from the flue gas of power stations and the off-gases from industrial processes such as the manufacture of nitric acid (HNO₃) from NH₃. Here the catalyst is vanadium-based and requires quite high temperatures, close to those available on heavy-duty trucks, and over recent years these catalysts have been increasingly used in these mobile applications. However, the lower temperatures on cars mean that catalysts of much higher activity are required to be effective. Gandhi over several years pushed catalyst manufacturers to achieve this remarkably difficult target through the use of new zeolite-based SCR catalysts whose performance is remarkable in the light of previous technologies, and they are being introduced into series production (57).

Conclusions

Throughout his entire career Haren Gandhi was deeply engaged in the technical aspects of emissions control, and he led the catalyst manufacturers to continually produce technically better products to meet increasingly demanding legislative requirements. He saw car tailpipe emissions go from being relatively out of control (compared to today) and a cause of major environmental problems, to being well controlled by innovative catalytic technologies. However, and as he appreciated, there are always new challenges and demands in this area, as evidenced by the recent growth of advanced diesel engines in cars in Europe with their unique emissions control requirements, and the growing need to reduce CO₂ emissions.

Unlike most in the automotive industry, Gandhi remained in his specific chosen area, and saw a series

of major objectives achieved. Because of his wide experience and depth of knowledge he became known as a practical emissions control guru. In many ways Gandhi was unique in the industry and all of those who knew and worked with him mourn the loss of a very special friend.

Acknowledgements

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