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Review

Exhaust Gas After-Treatment Systems for Gasoline and Diesel Vehicles

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Abstract: Exhaust gases released from vehicle engines have been a major cause of air pollution, and the emission limits have become much stricter in recent years due to a worldwide concern about the impact of air pollution on public health. These regulations have been complied to minimize the emissions of carbon monoxide (CO), hydrocarbons (HCs), nitrogen oxides (NO_x) and particulate matter (PM) from gasoline and diesel vehicles engines. Different after-treatment systems (ATS) have been developed for the treatment of exhaust gases from gasoline and diesel engines, respectively. The ATS for gasoline engine based on the three-way catalysts (TWCs), as well as the ATS for diesel engines including diesel oxidation catalysts (DOC), selective catalytic reduction (SCR), diesel particulate filters (DPF) and ammonia slip catalysts (ASC), are summarized in this mini-review.

Keywords: after-treatment systems (ATS); three-way catalysts (TWCs); catalysts; zeolite; diesel oxidation catalysts (DOC); selective catalytic reduction (SCR); diesel particulate filters (DPF); ammonia slip catalysts (ASC)

1. Introduction

The health risks of air pollution are extremely concerned around the world now, especially for the harmful air pollutants which are emitted from vehicles. Over the past four decades, several sets of emission standards have been defined for different types of vehicles. As shown in Table 1, the emission limits have become stricter and stricter both in both developed and developing countries [1]. Different after-treatment system (ATS) have been successfully developed for the treatment of exhaust gases from vehicles to meet these emission regulations, of which environment catalysis technologies have play important roles [2].

The classic gasoline engine ATS has built itself well on the three-way catalysts (TWCs) for over 30 years, which convert hazardous carbon monoxide (CO), nitrogen oxides (NO_x), unburnt hydrocarbons (HC) into harmless products carbon dioxide (CO_2), nitrogen (N_2) and water (H_2O) [3]. Unfortunately, the TWCs catalytic technology does not work well for the control of NO_x emissions on lean-burn engine operations with high oxygen/fuel ratios [4]. Therefore, selective catalytic reduction (SCR) of NO_x into N_2 with the reductant such as ammonia, has been commercialized for the abatement of NO_x from diesel engine vehicles [5].

Table 1. Emission standards for diesel engines of heavy-duty vehicles (g kW^{-1} h^{-1}) and gasoline of light-duty vehicles (g km^{-1})[1].

Type	Weight (kg)	Standard	Date	Test	CO	THC	NMHC	NO_x	NO_x + NMHC
Heavy-duty	>3500	Japan 2009	2009	WHTC	2.22	0.17	—	0.40	—
	>2610	Euro 6	2013	WHTC	4.0	—	0.16	0.46	—
	>3500	China 6	2020	WHTC	4.0	—	0.16	0.46	—
	>8500 (lbs)	EPA 2007	2007	FTP	—	—	0.14 (g per bhp h)	0.20 (g per bhp h)	—
Light-duty	<3500	Japan 2009	2009	WLTP	1.92	—	0.08	0.08	—
	<2610	Euro 6	2014[a] (2015[b])	NEDC	1.0[a] (2.27[b])	0.10[a] (0.16[b])	0.068[a] (0.108[b])	0.06[a] (0.082[b])	—
	<2500	China 6	2020 (2023[c])	WLTP	0.7 (0.5[c])	0.10 (0.05[c])	0.068 (0.035[c])	0.06 (0.035[c])	—
	<8500 (lbs)	Tier 3 (bin 1–160)	2017	FTP	0–4.20 (g mi^{-1})	—	—	—	0–0.16 (g mi^{-1})

[a] For passenger cars.

[b] For light commercial vehicles.

[c] For second stage starting from 2023.

WHTC = World Harmonised Transient Cycle. FTP = Federal Test Procedure. WLTP = Worldwide Harmonized Light Vehicles Test Procedure. NEDC = New European Driving Cycle.

2. The After-Treatment Systems for Gasoline Vehicles

2.1. The Origin of Three-Way Catalysts (TWCs)

The first generation of automobile catalyst was developed by Engerlhard (acquired by BASF in 2006) in the 1970s. The main active ingredients were platinum (Pt) and palladium (Pd) which were used to eliminate CO and HC. NO_x was reduced by a mechanical system called exhaust gas recirculation (EGR). However, because of the widespread addition of lead (Pb) additives in gasoline that was adopted to improve the octane number of gasoline, the catalyst had been plagued by Pb poisoning. Fortunately, a legislation was called for the elimination of Pb additives due to environmental concerns and mounting evidence of its toxicity.

With the implementation of NO_x regulations in 1980s, it was found that EGR systems cannot meet the requirement of 90% conversion of NO_x. Research on catalytic materials led to the discovery of Rhodium (Rh) as a highly active and stable catalyst for the reduction of NO/NO_2. [6] Pt and Pd were the best solution for the oxidation reactions of both CO and HC, while Pt/Pd/Rh became the main active components of the TWCs system. However, Farrauto et al. [7] reported different air-to-fuel feed ratios can affect the performance of TWCs. When there is fuel rich or deficient in O_2, it will facilitate the reduction of NO_x to N_2. On the other hand, when there is fuel lean or excess air, the oxidation reactions will be favourable. In order to control the engine operation and provide the narrow air-to-fuel ratio ($\lambda = 1$) , the first automotive Lambda sensor, also known as an oxygen sensor, was developed by Bosch in 1976 to monitor the amount of unburnt oxygen present in the exhaust pipe. Since then, TWCs exhibited their great advantage for the reduction of NO_x as well as the oxidation of CO and HC simultaneously.

2.2. The Composition of TWCs

Currently, about 98% of the cars with gasoline engines have been equipped with TWCs. As shown in Figure 1, Bahaloo-Horeh [8] described that a TWCs catalytic converter mainly consists of cordierites or metal base integral substrates with honeycomb structures, which are coated with high surface area oxide supports, catalytic active metals, as well as oxygen storage materials as the promoters.

Two kinds of monolith for TWCs have been used with cordierite ($2MgO{\cdot}2Al_2O_3{\cdot}5SiO_2$) and metal, respectively [9]. Cordierite monoliths have been widely used due to their advantages of low costs, good thermal shock resistance, remarkably low thermal expansion coefficients and high melting temperatures [10]. Such advantages make cordierites a good material that can withstand the harsh conditions of TWCs. On the other hand, a major advantage of metallic monoliths is their low heat capacity and high thermal conductivity, which allows the close-coupled catalyst (CCC) to be heated fast during the phase-in of the engine, and results in the minimization of the light-off time [11].

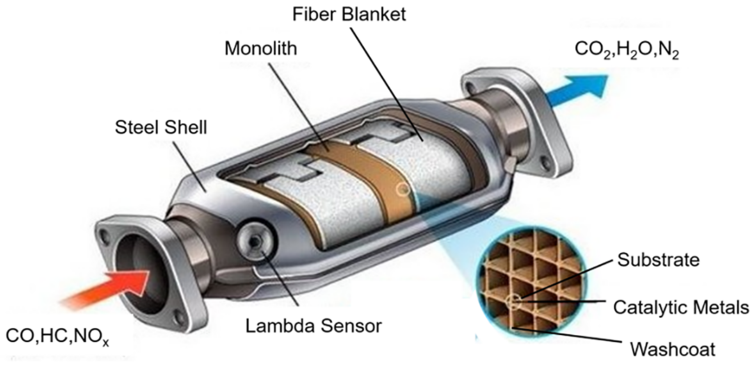

Figure 1. The composition of after-treatment systems (ATS) for gasoline vehicles.

Platinum (Pt), palladium (Pd), and rhodium (Rh) are the common clusters of platinum-group metals (PGMs), which are dispersed onto the surface of oxide supports and employed as the active components in TWCs designs [12]. Pt and Pd are used as oxidizing components for three-way catalysis, while Rh is necessary for the control of NO_x emissions. Currently, Pd-only, Pd/Rh, Pd/Pt/Rh, and Pt/Rh have been all commercialized for TWCs formulations [13]. Alumina (Al_2O_3) is a primary material in most TWCs washcoat formulations due to its high surface area, mechanical strength and good thermal stability [14]. In most of the cases, γ-Al_2O_3 is used as the first coating layer on the monolithic cordierite as a support of noble metal catalysts. Alkaline, alkaline earth metals and lanthanides are usually combined with Al_2O_3 as structural stabilizers to inhibit the conversion of Al_2O_3 from γ to α phase [15]. The introduction of CeO_2 as an Oxygen Storage Capacitor (OSC) enables a constant air-fuel ratio around the stoichiometry resulting in an enhancement on the catalytic activity and the durability for Pd-based catalysts [16]. Barium (Ba) has been reported to play an important role for the inhibition of the aggregation Pd active sites, which is indispensable for high performance catalysts [17]. Jing et al. [18] reported that Pd/La/Al_2O_3 can promote the catalytic reduction of NO efficiently, while Pd/Ba/Al_2O_3 exhibits high activity for the oxidation of CO and C_3H_6. In addition, TWCs formulations are often influenced by economic factors, regulatory standards, fuel quality and different types of exhaust composition.

2.3. Prospects of TWCs

As discussed above, TWCs have shown a quite mature and highly effective performance for pollution abatement. However, there are some inherent limitations which require further improvement and development. One of the biggest issues for TWCs is the degradation of the catalytic activity caused by the agglomerating of noble metals. Understanding the influence of the temperature and atmosphere (on the state of noble metals and the noble metals-carrier interaction) is the key to realize the stabilization of noble metal active sites. For example, Chen et al. [19] reported that the composition of aging gases impacted the particle size of Pd. Aging treatment under oxygen-rich conditions with the co-existence of steams at a high temperature led to much more serious agglomerating of noble metals. In contrast, particle growth is minimal in a mixture of depleted (O_2) gases below the decomposition temperature of PdO, and the sintered Pd particles could be modestly re-dispersed by treating in O_2. Therefore, it is very important to develop advanced TWCs synthesis methods and engine control technologies, which undoubtedly plays a critical role for the reduction of the usage of noble metals and the cost of TWCs.

The low activity at low temperatures (start-up of the engine) is another challenge for TWCs systems. The majority of the tailpipe emissions are emitted under low temperature operating conditions of the vehicle, especially during the cold start periods. The first possibility technology to reduce the cold start emissions is HCs traps, and typically HCs trap materials consist of zeolites and TWCs component. In an optimal trap, HCs emissions are trapped on zeolites at low temperatures. As the temperature is increased to above 250–300 °C, the stored HCs are released from zeolite components and subsequently converted on TWCs component. However, a major technical hurdle is that the temperature of HCs release is still relatively low comparing with the light-off temperature of TWCs. Moreover, the temperature of the under-floor TWCs catalysts may reach 850–900 ℃. Thus, a high stability under hydrothermal conditions is necessary for HCs trap materials w, but remains a challenge for zeolite-based systems currently [20]. As a result, the HCs trap technology has been employed only in some limited commercial applications so far. In addition, another approach is to develop new catalysts with ultra-high conversion efficiency at a low temperature in order to meet the stringent emission regulations in the future.

3. The After-Treatment Systems for Diesel Vehicles

3.1. Principles of the Operation

Diesel vehicles emissions are significantly dependent on the performance of the ATS. As shown in Figure 2, Beale et al. [21] described that a typical ATS is composed of diesel oxidation catalysts (DOC), selective catalytic reduction (SCR), diesel particulate filters (DPF) and ammonia slip catalysts (ASC) coated on honeycomb substrates such as cordierite, which is now used to meet Euro 6.

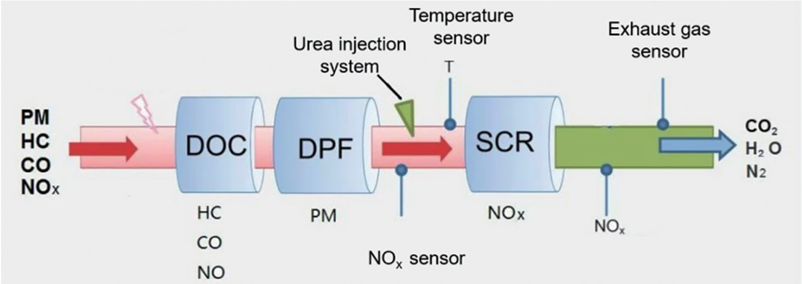

Figure 2. Layout of ATS for diesel vehicles.

TWCs is an effective NO_x reduction system without the presence of oxygen while it does not work well for the control of NO_x emissions on lean-burn engine operatiosn with high oxygen/fuel ratios. Thus, selective catalytic reduction of NO_x into N_2 (where ammonia is used as the reductant) has been commercialized for the deNO_x treatment of diesel vehicles [22]. The main reactions on SCR catalysts are shown as follows:

$$4NH_3 + 4NO + O_2 \rightarrow 4N_2 + 6H_2O \tag{1}$$

$$2NH_3 + NO + NO_2 \rightarrow 2N_2 + 3H_2O \tag{2}$$

$$2NH_3 + 2NO_2 \rightarrow NH_4NO_3 + N_2 + H_2O \tag{3}$$

$$4NH_3 + 4NO + 3O_2 \rightarrow 4N_2O + 6H_2O \tag{4}$$

$$4NH_3 + 5O_2 \rightarrow 4NO + 6H_2O \tag{5}$$

$$4NH_3 + 7O_2 \rightarrow 4N_2O + 6H_2O \tag{6}$$

As NO_x in diesel exhaust usually consists of >90% NO, the main reaction of SCR with NH_3 is the standard SCR reaction (SSCR) (1). When NO and NO_2 are ideally in equimolar amounts, especially at low temperatures, the conversion of NO_x by the reaction with NH_3 proceeds through the fast SCR reaction (FSCR) (2) [23]. In addition to the standard and fast SCR reactions, there are several side reactions occurring simultaneously. In the low-temperature regions, NH_4NO_3 will be accumulated through reaction (3). In the high-temperature regions, the non-SCR and catalytic oxidation of NH_3 will occur through reactions (4-6). Therefore, good NH_3-SCR catalysts and their systems should be developed to promote the reactions of (1) and (2) and inhibit the reactions of (4)~(6) [24].

3.2. Diesel Oxidation Catalyst (DOC)

The DOC unit is typically the first component in the Diesel ATS with platinum-group metal (PGM) as catalytic active species. The DOC unit serves in commercial emission control systems with two major functions. First, it converts hydrocarbon (HCs) and CO into CO_2 and water. Second, NO can be oxidized to NO_2 on DOC, while a higher proportion of NO_2 is beneficial to improve the NO_x conversion efficiency of SCR [25]. Moreover, it can create exothermic heat which can be used to regenerate DPF due to the exothermic oxidation reaction on DOC units.

3.3. Selective Catalytic Reduction (SCR)

Selective catalytic reduction with ammonia (NH_3-SCR) is a technology used to abate nitrogen oxide emissions from diesel engines effectively [26]. Vanadium-based oxides, copper zeolite, and iron zeolite are three general families of SCR catalysts for commercial use today [27]. Copper zeolite has been the leading choice due to the high activity under a wide temperature window, among which small-pore zeolites such as CHA have shown especially good hydrothermal stability and excellent resistance to HC poisoning [28]. As a result, Cu/SSZ-13 zeolite has been employed for the high-performing SCR systems as the most popular SCR catalyst [29]. Comparing with copper zeolite, iron zeolite exhibits a better catalytic performance at a high temperature, as well as a lower affinity for sulphur. However, the inferior low-temperature activity of iron zeolite has hampered its applications to meet stringent emission limits with a low exhaust temperature [30]. Vanadium-based oxides catalysts are generally preferred for diesel engine using high-sulphur fuels due to their greater sulfur resistance. However, the key drawback of these catalysts is their poor stability at a high temperature [31].

3.4. Diesel Particulate Filters (DPF)

The combustion patterns of diesel engines usually result in high emissions of particulate matter (PM). Therefore, DPF have become a necessary configuration to meet increasingly stringent PM emission standards [32]. Modern DPF is a wall-flow ceramic monolithic filter, which mainly consists of cordierite, silicon carbide or aluminum titanate. Engine exhaust gas is forced to flow through a porous wall to adjacent outlet channels of the DPF, then most soot agglomerates accumulate on the filter [33]. To keep the filter clean, the collected soot must be burnt via DPF regeneration.

3.5. Ammonia Slip Catalyst (ASC)

Generally, excess ammonia is injected to ensure the high conversion efficiency of NH_3-SCR deNO_x systems. However, some unreacted ammonia may slip through the SCR units. ASC catalysts are normally coated on the honeycomb substrate with PGM catalyst, which can oxidize slipped ammonia to N_2 [34]. Recent developments in ASC catalysts technology is a dual-layer structure in which the PGM catalyst coating is located below the SCR catalyst coating. The bottom PGM layer oxidizes NH_3 to N_2 with the byproduct of NO_x, while these latter compounds diffuse back to the above layer in which they are also transformed into N_2 with the excess NH_3 typically stored by the SCR catalyst. Thus, this solution shows great advantages in terms of NH_3 overall conversion and N_2 selectivity [35,36].

3.6. Prospects of After-Treatment Systems (ATS) for Diesel Vehicles

Despite SCR is the leading lean deNO_x technology today, there are still plenty of opportunities for the improvement on SCR systems for the ATS of diesel vehicles. One significant opportunity is to improve the catalytic performance at low temperatures below 200 ℃. The cold-start deNO_x performance remains a great challenge considering the kinetic limitations of SCR catalysts and the decomposition of urea at low temperatures. PNNL, Cummins Inc. and Johnson-Matthey Inc. have collaborated to develop a sustained low-temperature NO_x reduction (SLTNR) system capable of removing 90% of NO_x at SCR inlet temperatures of 150 °C. The key of this new technology is the utilization of fast SCR instead of standard SCR so as to boost NO_x conversion. As a result, a diesel oxidation catalyst (DOC) bed is required to oxidize NO to NO_2. [37] A second solution to solve the cold-start problem is the usage of passive NO_x adsorbers technology (PNAs), of which the adsorbent materials are invented to store all NO_x emitted after the engine start-up until the exhaust temperature is high enough for the SCR reaction. For example, the group of Tronconi showed a new two-layer monolithic AdSCR system (AdSCR=adsorption + selective catalytic reduction), which was constituted by Cu/CHA (top layer) and BaO/Al_2O_3 (bottom layer), to improve the $deNO_x$ performances of ATS for diesel vehicles at low temperatures [38]. Another opportunity is to further improve the stability of SCR systems. Especially, the hydrothermal stability of Cu/SSZ-13 zeolite can be considered as a critical criterion for the evaluation of the long-term stability for practical applications [39]. As the dealumination and the loss of surface area/pore volume of zeolites, which lead to the transformation of Cu active species into inactive forms, do occur during the hydrothermal aging, new zeolites should be designed and synthesized to withstand harsh hydrothermal aging tests.

4. Concluding Remarks

In this review, a number of after-treatment devices have been summarized. It can be found that no single setup is sufficient to eliminate exhaust pollutant completely. New catalytic materials and control technologies remain as impending demand to meet much stricter limits and rules such as Euro 7. Scientific understanding on the catalytic mechanism, rational design of materials and ATS, as well as precise powder coating should be further investigated and developed not only for ATS of gasoline vehicles but also for that of diesel vehicles with high decontaminating efficiency. Besides, ATS of automobiles should also have compact design with low costs to benefit both procurements and operations.

It has been well acknowledged that anthropogenic emissions of CO_2 are the main cause to the continuous increase of the global temperature since the industrial revolution. Many CO_2 mitigation strategies have been under investigation in the past years with the goal of net-zero emissions by 2050 [40]. New engine systems and their exhaust gas after-treatment systems should be developed with the usage of zero-carbon fuels and biofuels such as ammonia, biogas and synthetic hydrocarbon with carbon capture.

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