Development Status and Trend Analysis of Four-way Purification Technology for Automobile Exhaust

Wang Ying hui *, Liang Qiu song, Lu Zhen sheng*, Wang Jiu long, Chen Nan xin
Suihua University, Suihua, Heilongjiang, 152061
*sjidan@outlook.com
corresponding author: Lu Zhen sheng; sjidan@163.com

Abstract. Automobile exhaust has seriously polluted the environment and endangered human health, in order to satisfy strict China VI regulation, four-way purification technology will become effective engine after-treatment technology for purifying automobile exhaust. This paper firstly introduces reaction mechanism of purifying particular matter, nitrogen oxide, carbon monoxide and hydrocarbon. Secondly, this paper summaries the development status of four-way purification technology for automobile exhaust from two aspects. Finally, this paper suggests that automobile exhaust can be purified by plasma combined with modified perovskite catalyst technology, this method has the advantages of low back pressure, low cost, simple structure and strong stability compared with the traditional automobile exhaust purifier, which can achieve the purification of PM, NOx, CO and HC simultaneously, and open up a new direction for automobile exhaust purification technology.

1. Introduction
The first paragraph after a heading is not indented (Bodytext style). With the sustained and rapid economic and social development, the number of automobiles in China is on the rise in recent years. According to the statistics of the Ministry of public security, in 2018, the number of cars in China has reached 240 million [1], at the same time, automobile exhaust pollution also has a serious impact on urban air quality safety and human health. Among them, PM and NOx are the main sources of air pollution[2-3]. Haze and photochemical smog produced by PM and NO2 is one of the important factors leading to lung cancer. The combination of CO and hemoglobin can cause headache, fainting and even endanger people's lives. HC have serious damage to human respiratory, nervous and hematopoietic systems [4]. Therefore, on December 23, 2016, the Ministry of environmental protection of the people's Republic of China and the General Administration of quality supervision, inspection and Quarantine of the people's Republic of China jointly issued the latest document of GB 18352.6-2016 《light vehicle pollutants and measurement methods (phase 6 in China)》 , which will be implemented nationwide in July 2020 [5]. In order to meet the stringent national six emission standards, four effect purification technology has become the key development direction of automobile exhaust purification [6].

2. Reaction mechanism of automobile exhaust purification
Automobile exhaust pollution mainly refers to the harmful gases and solid suspended particles produced by the incomplete combustion of automobile fuel. The harmful gases are mainly composed of NOx, CO, HC. Solid particles are mainly composed of heavy metals Pb and PM, in which PM is
composed of soot particles, soluble organic fraction and sulfate [7]. In recent years, in order to reduce the emissions of PM, NOx, CO and HC, domestic and foreign scholars have carried out research on automobile exhaust purification technology.

2.1. Reaction mechanism of NOx purification

There are mainly two kinds of NOx purification methods: selective catalytic reduction (SCR) and NOx storage catalytic reduction (NSR). In the 1990s, developed countries in Europe and the United States put forward SCR units for NOx purification, the main active component of the catalyst is V2O5, which cooperates with urea to selectively reduce NOx [8]. SCR uses the organic compounds in the tail gas as reducing agents to selectively reduce NOx to N2 when the oxygen concentration is more than two orders of magnitude higher than the NOx concentration [9]. NH3, urea, alcohols and various hydrocarbons are usually used as reducing agents in the catalytic process, the NOx content in the tail gas is detected by the sensor, and then urea solution is quantitatively injected onto the catalyst, under high temperature conditions, urea will be decomposed into NH3 and CO2. NH3 reacts with NOx under the condition of catalyst active components to form harmless substances, and the reaction is shown in formula (1-1)~(1-3)[10].

\[
\text{catalyzer}\quad 3\text{NH}_3 + 3\text{NO} \xrightarrow{\text{catalyzer}} 3\text{N}_2 + 3\text{H}_2\text{O} \\
\text{catalyzer}\quad 3\text{NH}_3 + 6\text{NO} \xrightarrow{\text{catalyzer}} 5\text{N}_2 + 6\text{H}_2\text{O} \\
\text{catalyzer}\quad 8\text{NH}_3 + 6\text{NO} \xrightarrow{\text{catalyzer}} 7\text{N}_2 + 12\text{H}_2\text{O}
\]

In the 1970s, Japan first proposed the NSR technology [11]. Under the condition of lean burn exhaust, NOx mainly exists in the form of NO. since NO2 is easier to store than NO, NO is first oxidized to NO2 by O2 at the active site of noble metal, and then NO2 is stored on alkali metal or alkaline earth metal in the form of stable nitrate or nitrite, and then the reaction is switched to rich combustion condition, In a short period of time, the concentrations of reducing components HC, CO and H2 in the tail gas increase rapidly, and the stored nitrate or nitrite decomposes to give off NOx. Under the three-way catalysis, NOx is reduced to N2 by HC, CO and H2 [12]. However, the catalyst used in NSR technology is easy to sulfur poisoning, so it is difficult to realize its application according to the current fuel quality in China.

2.2. Reaction mechanism of purifying PM

Diesel particulate filter (DPF) is the most direct way to solve the problem of particulate matter emission, DPF is mainly a kind of ceramic filter, which can be installed in automobile exhaust system. The DPF is coated with soot oxidation catalyst, when the tail gas passes through the DPF, PM in the tail gas will be absorbed and trapped, while, the burner at the tail end of DPF will automatically ignite, and the adsorbed particles will be burned into carbon dioxide and water, and the purification efficiency of PM can reach 85% [13]. However, after the DPF works for a period of time, its internal pores are filled with particles, which increases its back pressure and affects the working performance of the engine. Therefore, DPF needs to be regenerated. DPF regeneration is divided into negative regeneration and active regeneration. Passive regeneration is to use catalyst to reduce the ignition point of soot and make soot burn at a lower temperature, so as to regenerate the catalyst; active regeneration is to improve the DPF The soot particles were heated to make the soot particles burn [14].

2.3. Reaction mechanism of purifying HC and CO

Diesel oxidation catalyst (DOC) can oxidize sof in PM, so as to reduce particulate matter emission, and further reduce the emission of gaseous pollutants CO and HC [15]. DOC usually uses ceramic honeycomb or metal honeycomb as the carrier, and its surface is composed of oxide coating and active metal components. The commonly used active metal components are Pt and PD, which can be used alone or combined with other emission post-treatment technologies, improved fuel, exhaust gas recycling (EGR) and other internal purification technologies [16]. Catalytic oxidation technology uses catalyst to reduce the chemical reaction activation energy of HC, CO and sof in diesel vehicle exhaust,
so that these substances can be oxidized with oxygen in exhaust at a lower temperature, and then converted into carbon dioxide and water. The main reaction is shown in formula (1-4) ~ (1-7)[17]:

\[
\begin{align*}
2\text{CO} + \text{O}_2 &\rightarrow 2\text{CO}_2 \\
4\text{C}_n\text{H}_m + (m + 4n)\text{O}_2 &\rightarrow 2m\text{H}_2\text{O} + 4n\text{CO}_2 \\
[\text{SOF}] + \text{O}_2 &\rightarrow \text{CO}_2 + \text{H}_2\text{O} \\
\text{SO}_2 + \text{O}_2 &\rightarrow \text{SO}_3 \rightarrow \text{SO}_4^{2-}
\end{align*}
\]

Higher exhaust temperature will contribute to the oxidation of SOF and improve the conversion efficiency of PM. However, SO\text{2} in the exhaust gas will be oxidized to sulfate under high temperature conditions, if the fuel contains high sulfur, the proportion of sulfate in PM will be increased, resulting in increased PM emissions. Therefore, this technology is generally not applicable to fuel with high sulfur content [18].

3. Development status and trend analysis of four-way purification technology

In order to purify PM, NO\text{x}, CO and HC in automobile exhaust at the same time, domestic and foreign scholars have carried out research on four-way catalytic technology of automobile exhaust. Yoshida first proposed the concept of simultaneous removal of NO\text{x} and PM by catalytic method in the oxidation atmosphere of diesel vehicle exhaust [19]. Four way catalytic technology can be divided into integral four-way catalytic technology and combined four-way catalytic technology, integral four-way catalytic technology Three way catalysis is a technology to realize simultaneous purification of PM, NO\text{x}, CO and HC by using catalysts; combined four-way catalysis is an integrated exhaust purifier formed by optimizing the combination of two or more post-treatment devices. Based on the characteristics of the two four-way catalytic technologies, the key is the optimal combination of four-way catalytic converter and the development of four-way catalyst [20].

3.1. Four-way catalytic converter

At present, a variety of four-way catalytic conversion systems have been proposed for purifying vehicle exhaust PM and NO\text{x}, which mainly include the combination of DOC and SCR, , the four-way catalytic converter products have been commercialized in Germany, Japan, the United States and other developed countries [21].

The quad cat four-way catalytic converter is adopted by Cerix company of the United States, which is composed of heat exchanger, diesel vehicle DPF or diesel vehicle doc, fuel injection system, control system and LNC [22]. Quad CAT four-way catalytic converter can reduce CO and HC emissions of motor vehicles by more than 95%, PM by 90% and NO\text{x} by 44%. At present, the converter has been put into operation and can be used by motor vehicles. Due to the low NO\text{x} purification efficiency, the technology still needs further optimization design [23].

In recent years, Toyota company has proposed a four-way catalytic system with its structure as shown in Figure 1. The system is coupled with DPF and NSR technologies, and its purification efficiency of soot particles and NO\text{x} reaches more than 80% [24]. The dparm system adopts a wall flow ceramic filter element, and applies NO\text{x} adsorption and reduction catalyst on the filter element to achieve reduction effect. In addition, NO\text{x} storage and soot combustion can be realized in lean burn state After switching to the concentrated combustion state, the NO\text{x} stored in the catalyst is reduced, and the catalyst still has certain soot combustion activity [25].
Mitsubishi Automobile Co., Ltd. of Japan has developed a new generation of four-way catalytic converter for automobiles. The structure of the converter is shown in Fig. 2. The Pt based selective reduction catalyst adsorbing HC is added to the front support, which can efficiently purify NOx, and Pt based oxidation catalyst is used in the rear stage, which can oxidize the sof in CO, HC and PM. When the catalytic converter is combined with EGR, the catalytic converter can be used to oxidize the sof in CO, HC and PM When used together, the emission of NOx and PM in vehicle exhaust can be less than 0.4g/km and 0.08g/km [26]. The four-way catalytic converter has been basically commercialized and is now being popularized in Europe.

3.2. Four-way catalyst
At present, the three-way catalyst which is suitable for automobile exhaust purification has been widely used, and entered the industrialization stage. It mainly purifies three kinds of harmful substances, NOx, CO and HC in the automobile exhaust at the same time. There is no mature four-way catalytic technology applied to the automobile for PM. In order to make the automobile exhaust emission meet the standard, the key is the four-way catalytic converter and four-way catalyst In recent years, the research and development of four-way catalysts for simultaneous purification of PM, NOx, CO and HC has become a hot spot at home and abroad, including noble metal four-way catalyst and non noble metal four-way catalyst.

3.2.1. Noble metal four way catalyst
Platinum (PT), rhodium (RH) and palladium (PD) with strong selectivity and high activity are mainly platinum (PT), rhodium (RH) and palladium (PD), which have a strong pr-omotion effect on the purification of PM and NOx. The fresh Pt catalyst has good reduct-ion performance at 180 ~ 210 °C, and its activity decreases after aging test, and shows h-igh activity in the high temperature range of 260 ~ 280 °C [27]. In 2006, Matarrese et al. Studied the catalytic activity of noble metal Pt involved bimetallic catalysts for simultan-eous removal of NOx and soot particles, and the reaction activities of pt-k / Al2O3 and Pt BA / Al2O3 catalysts, and found that the former showed higher activity in soot oxidati-on and NOx storage capacity [28].
Engelhard company of the United States has developed a new four-way catalyst. The catalyst is composed of zeolite and the second component of noble metal and non-catalytic porous zeolite. Its catalytic characteristics are affected by temperature. When the temperature reaches the normal working temperature of the catalyst, the gas components adsorbed by zeolite are desorbed and released. Under the action of Pt, HC and Co are oxidized to CO2. When the temperature does not reach the temperature required for the normal operation of the catalyst, the catalyst can use the adsorption characteristics of zeolite to absorb HC, SOF and NOx in the exhaust gas, so as to achieve the purpose of four-way catalysis. This catalyst can reduce the mass concentration of NOx in the exhaust gas at 20 mg/m³ [29].

Wu et al. developed a four-way catalyst with noble metal Pt as the main component. The catalyst was supported by Al2O3 with Pd content of 2.47 kg/m³. The European test system results show that the catalyst can effectively degrade 90% CO and HC, and the purification rate of PM can reach 40%, but the purification rate of NOx is only 10% [30]. At the same time, He Hong et al. have developed a catalytic system which can selectively remove HC, CO and NOx in the atmosphere containing SO2, mainly including inorganic porous carriers and precious metals Pt, Ag and Au. The purification rate of NOx is more than 90%, but the purification capacity of soot particles is limited, so further optimization design is needed [31].

The noble metal four-way catalyst has low ignition temperature and high activity, but it is easy to sinter at high temperature, which results in the loss of active components due to sublimation, which reduces the activity. The required air-fuel ratio window is very narrow, which is easy to be poisoned and inactivated by sulfur (S), lead (Pb), phosphorus (P) and other elements in fuel oil and lubricating oil. At the same time, precious metal four-way catalyst is limited in resources and expensive in price. It cannot be used on a large scale [32]. At present, a kind of cheap catalyst which can replace the traditional precious metals is being studied at home and abroad.

### 3.2.2. Non noble metal four way catalyst

Up to now, the active components of perovskite type four-way catalysts which are studied more include rare earth elements plus Mn, Co, Fe and Cu, which are Mn based, Co based, Fe based and Cu based active components [33]. The perovskite type four-way catalyst can be supported by molecular sieve HZSM-5, cordierite honeycomb ceramics, alumina (Al₂O₃), titanium oxide (TiO₂) and metals. The activity of the catalyst can be improved by using the characteristics of "chemical tailoring" of perovskite type composite oxides. The perovskite type composite oxides are orthonormbic oxides with molecular formula of ABO₃, and their cubic structure is shown in Fig. 3. It is shown that an ion is located in the center of the cube, and oxygen ion is located at the edge. There are 12 oxygen coordination sites. Metal ions at B site are surrounded by adjacent octahedral oxygen. These oxygen ions are shared by eight BO₆ octahedrons. A site is rare earth metal or rare earth metal with alkali gold or rare earth metal with alkaline earth metal, and B site is one or two kinds of metal such as CO, Mn, Fe and Cu combination [34]. In the perovskite type catalyst In LaCoO₃ structure, K⁺ with larger ionic radius is used to replace La³⁺ at a site with smaller ion radius, which makes lattice oxygen expand and deform, resulting in the increase of oxygen vacancies. In this way, the valence of CO will be changed and co exists in +3 and +4 valence states. The doping and substitution of new B-site metal Mn can also produce doped Mn valence, thus forming a valence state with higher oxidation performance and perovskite type La₁₋ₓKₓCo₁₋ₙMnxO₃ can further improve the redox performance of the catalyst. The conversion process is shown in Fig. 4. When x = 0.2, the catalytic oxidation efficiency of PM reaches 93.8%, and when y = 0.3, the catalytic oxidation and reduction of CO, HC and NOx are the best, reaching 100%, 99.9% and 75.8%, respectively. From the perspective of kinetics, La₀.₈K₀.₂Co₀.₇Mn₀.₃O₃ is the best. It can accelerate the catalytic oxidation-reduction rate of PM, NOx, CO and HC, and improve the four-way catalytic efficiency [35].
Zhao Zhen et al. studied the four-way catalytic activity of aluminum oxide supported on different specific surface areas for automobile exhaust gas. It was found that when supported with Mn, the catalyst had good catalytic performance, which could reduce the light off temperature of soot particles, and the selectivity to CO$_2$ generation could reach 100%. At low temperature, the catalyst also had high activity for hydrocarbon conversion, but it decreased. The selectivity of NO$_x$ to N$_2$ is reduced [36]. Liu et al. developed a double-layer four-way catalyst. Under the condition of four-way catalysis, the combustion temperature of soot particles can be reduced to 421 °C, the purification rate of NO$_x$ can reach 74%, the temperature of complete conversion of HC is 357 °C, and the purification rate of CO can reach 99% [37].

The non noble metal four-way catalyst has the advantages of sufficient activity, low price and excellent thermal stability. It is considered as an ideal catalyst for automobile exhaust gas purification and has been paid attention to by people [38]. Therefore, the development process of catalytic materials is an alternative process, that is to use materials that meet the requirements of catalytic performance, have lower cost and have a wide range of sources.

4. Conclusion and Prospect

With the promulgation of the China VI regulation, automobile exhaust purification has become an urgent problem to be solved. Due to the low level of engine production in China, it is difficult to meet the emission standard by single internal purification method [39]. Therefore, in order to meet the requirements of the regulations, external purification has become the focus of research on automobile exhaust purification, and four effect purification technology is considered as one of the most ideal external treatment technologies. There are some problems such as high requirements for catalyst, incomplete purification of exhaust gas and low selective reduction of NOx Plasma) purification technology of automobile exhaust has the advantages of good treatment effect, wide treatment range, simultaneous treatment of multiple pollutants, complete purification without secondary pollution and no requirement for sulfur content of fuel [40]. Therefore, in order to improve the activity and selectivity of perovskite catalyst, this paper proposes a plasma four-way purification technology. The purification device is shown in Fig. 5. The device consists of a shell and a catalyst. The shell is used as the outer electrode of the plasma generator, the catalyst carrier is used as the discharge medium, and a plurality of wires around the carrier are used as the central electrode. The above three constitute a dielectric barrier discharge reactor to generate NTP. The central electrode is connected with the high-voltage power supply outside the catalyst shell through the wire, and the shell is composed of left and right shells. The sealing performance of the purifier is ensured by the sealing flange connection. In order to make the catalyst load evenly, the porous honeycomb ceramic carrier with bottom diameter of 110mm, height of 100mm, hole type of square hole and average inner diameter of square hole of 400 mesh is selected, which has the characteristics of thin hole wall, large surface area, impact resistance and high temperature resistance.
Although the plasma combined with modified perovskite catalyst can achieve simultaneous purification of PM, NO\textsubscript{x}, CO and HC, the perovskite catalyst can not exist in the ideal perovskite structure, which will produce defect structure to a certain extent, that is, the structure of the catalyst will be distorted and distorted. Therefore, this paper proposes that the plasma combined with the modified perovskite catalyst which will exist in the ideal perovskite structure. At present, plasma and modified perovskite catalyst combined with four-way purification technology is a potential technology. This technology has the advantages of low exhaust back pressure, low cost, simple structure and strong stability, but it has energy consumption due to low temperature plasma collaborative catalysis. For example, COMSOL is used to simulate and analyze the electric field, flow field and chemical field of the plasma four effect automobile exhaust purifier. On this basis, we can explore the influence of structural parameters such as applied voltage amplitude, high-voltage electrode diameter, dielectric layer relative dielectric constant on the discharge device, so as to facilitate the plasma four effect automobile exhaust gas The optimization design of purifier provides theoretical reference.

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