

Development, Application and Direction of Development of Urea-SCR

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Abstract

Nitrogen oxide (NO_x) is one of main pollution sources of air pollution, so it need to be more and more stringent emission regulations limiting pollution. Selective catalytic reduction (SCR) technology is the mainstream technology to remove NO_x at euro V and euro VI stage. By analyzing chemical reaction mechanism of selective catalytic reduction (Urea-SCR) of urea, research actuality of Urea-SCR from the urea storage device, urea supply system, catalytic converter and control unit separately was pointed out. At last development trend of SCR technology based on research that we have was analyzed.

Keywords: Urea-SCR, NO_x, Reaction mechanism

1. Introduction

Recently, with the aggravation of global atmospheric pollution and increase of awareness for environmental protection, the NO_x after-treatment technology is paid more and more attention. NO_x after-treatment technology includes the direct catalytic decomposition technology, selective non-catalytic reduction (SNCR) technology, selective catalytic reduction (SCR) technology, NO_x storage reduction (NSR) technology and plasma-facilitated catalysis (PFC) system. The promotion of practical applications of direct catalytic decomposition technique and SNCR are limited in the field of automobile exhaust purification due to their own limitations. NSR that is also called NSC (NO_x Storage Catalyst) or LNT (Lean NO_x Trap Catalyst) is a technology pathway developed firstly by Toyota in 1996. It is the mainstream technology of light diesel engines to reduce the NO_x, and it is suitable for applications where space is not enough to install SCR system especially. This technology has few applications in China. There are still some key theoretical and technical difficulties of PFC technology to be resolved, so this technology still in the process of theoretical research and experimental studies.

As the mainstream technology of heavy duty diesel engines to reduce the NO_x, SCR refers to the reaction of exhaust and NO_x to N₂ by inhibiting the nonselective combustion of reductant under the action of catalyst. The absolute amount of unburned HC in the exhaust is low, and diesel engines need adding a little more reductant to purify NO_x since they adopt oxygen-enriched combustion technology. According to the differences of additional reductant, SCR can be divided into urea selective reduction of NO_x (Urea-SCR) and HC selective reduction of NO_x (HC-SCR). HC-SCR technology uses hydrocarbon fuel as reductant and zeolite or Ag substrate (Ag/Al₂O₃) as a catalyst generally, so the cost is relatively low. Its advantage is that the catalyst has high initial activity, and the disadvantages are that hydrothermal stability is poor, sulfur resistance is low, low-temperature activity is not ideal, and practical application is limited.

Urea-SCR technology is developed based on the ammonia selective reduction of NO_x (NH₃-SCR) technology. But as one kind of gas, ammonia is too difficult to move and store. Ammonia is also highly corrosive, prone to leakage, while urea is characterized by non-toxic, clean, odorless, non-flammable, no explosion danger and low corrosive. So we

use an aqueous solution of urea instead of ammonia generally in the current, and use the ammonia from urea thermal decomposition to restore the NO_x of diesel exhaust. Urea - SCR has been used all over the world widely as the most mature post-processing technology of NO_x.

2. Structure of System and Chemical Reaction of SCR

A typical Urea-SCR system includes urea storage device, urea supply system, catalytic converters and control unit, as shown in Figure 1.

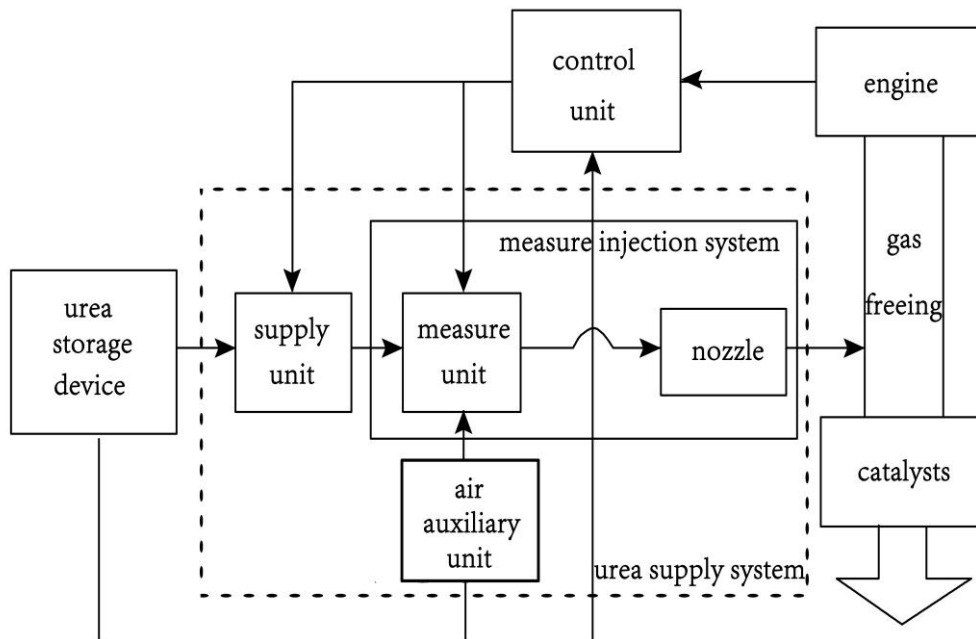


Figure 1. Schematic Diagram of Typical Urea-SCR System Structure

When SCR is working, the control unit performs the predefined control strategy. Then the urea solution in the storage device is sucked by the supply pump and injected into the exhaust pipe in front of the catalysts through the nozzle of urea supply system. The urea reduces into reductant NH₃ by heating, as shown in formula (1). On the surface of the SCR catalyst, the NO_x is reduced to N₂ by NH₃, its main formula of reaction as shown in (2) and (3):



In high-temperature environment (higher than 300 °C), NO_x in the exhaust gas can quickly react with ammonia (NH₃) to form N₂ and H₂O under the action of the catalyst. The temperature of diesel engine exhaust is generally in the range of 300 ~ 500 °C, it meets the temperature conditions that NO_x reduction needs.

3. Present Status of Urea-SCR Technology

The most ideal working conditions of Urea-SCR system is to ensure a low escape rate of reductant NH_3 on the premise of achieving a high conversion of NO_x . To meet these two requirements, we must ensure the following three performance indicators: good level of droplet evaporation and pyrolysis, reasonable space velocity distribution, reasonable concentration distribution of the reductant. SCR supply system should ensure that the liquid droplet can be reacted completely for evaporation and pyrolysis before entering the inside of the catalytic converter carrier. And reasonable space velocity distribution is to ensure that reductant and reactant have plenty of contact and reaction time. Moreover reasonable distribution of reductant is the determinant conditions that avoid excess ammonia coefficient too high or too low. The current studies are going largely around the urea storage device, urea supply system, catalytic converters, control unit and three performance indicators that are mentioned later in this article.

3.1 Urea Storage Device

Urea storage device, which is used for storing the urea solution, mounts transducer, heating device and ventilation device at the top and the screw holes to release the residual liquid at the bottom. Transducer includes temperature sensor and level sensor, which can sense the temperature and height of the urea level.

It consists of two parts to research urea storage device: the stability of solution and the adaptability to the environment. Urea solution is alkaline, the PH of which is 8.8. And its corrosion for copper, cast iron, solder and aluminum is serious. So we must use special materials such as austenitic steel to make its storage, pipeline transportation and filling. The urea storage device generally uses urea aqueous solution which concentration is 32.5 %, the most important reason is the lowest freezing point of the solution (-11°C). Even if the concentration of the urea aqueous solution is in a semi-frozen state, the solution concentration remains unchanged. Not only that, the concentration of solution can still keep unchanged for a long time in normal storage conditions. So it is suitable for long-term preservation in process of transport or storage. And it also meets the requirement that vehicle is not used for a long time.

When the temperature was lower than the -11°C , urea solution began to crystallize. The storage container, pipeline, pump and nozzle need to be heated in the environment temperature less than -11°C to prevent crystallization. Urea tank, which has the low temperature heating device, uses engine circulating water heating in general.

In addition, there are a lot of alternatives of urea solution, including the solid urea, ammonium salts (such as ammonium carbamate), chloride salts (such as magnesium chloride) storage technologies of ammonia [1-2]. Part of the solution can make the freezing point of urea solution to -30°C .

FEV company in 2009 showed solid selective catalytic reduction (SSCR) system on the Dodge Ram Pickup Truck [3], which declared the SSCR technology has entered a practical stage. Ammonia's solid precursor using ammonium carbamate. In the container, ammonium carbamate decomposition heated cyclically by oil produces ammonia. Working state of heating device was adjusted by control system according to pressure and temperature in the container. Generated ammonia was sprayed into exhaust pipe through quantitative injection device.

Patent of Germany's MAN commercial vehicle company in China [4], provides a means and device for solid urea pellets with grinding and pyrolysing to produce ammonia. U. S. patent of Theodore J, *et al.* [5], provides an apparatus and method for production of ammonia by solid urea pellets.

However, it still need further study about additional heat source, influence of fuel economy, the response characteristics of the release of NH_3 and other issues.

3.2. Urea Supply System

The main function of the urea supply system is that urea is dispensed from the storage device in accordance with the requirements of the controller and transported to the nozzle with a certain pressure. But the urea will be withdrawn in the line to urea tank when the system to be turned off. Urea supply system consists of the following parts: the urea supply unit, the air assist unit, measuring injection system, connecting pipes, all kinds of sensors and heating devices *etc.*

There are two main types of urea supply unit: one is represented by Grundfos company, another is represented by Bosch company. The first unit uses a stepper motor to drive a constant volume of plunger reciprocating motion and achieves the control of supply by controlling the speed of the motor. The second unit achieves the control of supply by controlling the solenoid valve, which is controlled by a PWM. But it must adopt a urea pump to build a constant pressure in the line before the solenoid valve.

Urea supply system is divided into two types: air-assisted and non-air-assisted. Urea supply system that used the compressed air-assisted was relatively common at the beginning of researching the automotive SCR technology, such as Bosch, Grundfos and Albonair. Then the urea supply system which had non-air-assisted appeared gradually, and it was putted into business operations successfully. Air-assisted system uses compressed air, and it uses metering pump for controlling the injection amount of the urea solution commonly. Experiment and simulation results show that the effect of air-assisted spray atomization is good [6]. And air-assisted promotes the decomposition of urea. It can save the cost at the same time because the original car brings air compressors. Non-air-assisted supply system establishes pressure through the urea pump. Urea solution does not need compressed air because it can achieve spray under the high pressure. In addition, the advantages of non-air-assisted include simple system principle, fewer parts, tight structure, low failure rate and precise control. Their disadvantages are that the effect of spray is poor. And nozzle needs to be cooled down because of its complex structure.

Figure 2 shows a set of non-air-assisted urea supply system. Under the action of the motor in the urea pump, urea solution in the urea tank is sucked up, and reaches the urea injection unit after the urea pump, and the end injects into the exhaust channel through the nozzle. Dosing Control Unit (DCU) is used to control the injection quantity and injection timing of urea solution. First the rotation of the urea pump motor is controlled by DCU, so that the pipeline pressure before urea injection unit is stable in the numerical value of calibration. Excessive urea solution goes back to the urea tank through backflow inlet. Then according to the current state of exhaust to determine the required amount of urea, by adjusting the pulse width of PWM signal to control the opening time of urea injection unit, the corresponding amount of urea solution sprayed into the exhaust channel, in order to achieve the measurement and atomization of urea. The cooling of urea injection unit is accomplished by the engine cooling liquid, and the working temperature is not more than 140°C.

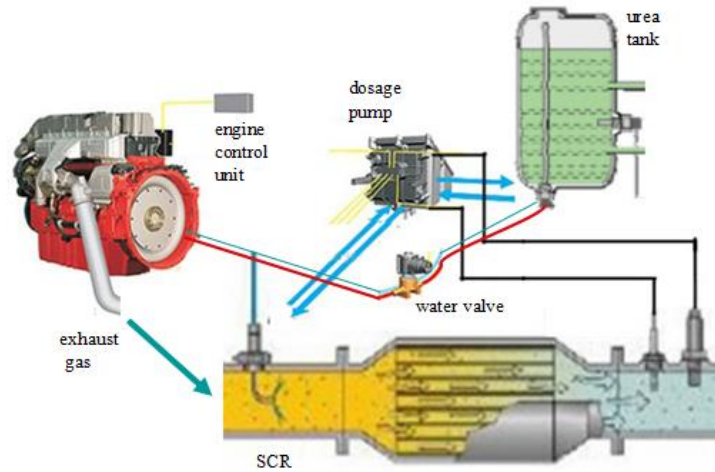


Figure 2. Non-Air-Assisted Urea Supply System

Figure 3 shows a set of air-assisted urea supply system. The metering of urea solution is accomplished by the diaphragm pump. Stepping motor through the belt drive eccentric wheel, then the eccentric wheel drive the plunger reciprocating movement, and the diaphragm is finished under the action of the plunger to the suction and extrusion process of urea solution. Diaphragm working chamber volume is fixed, so the stepper motor each push a diaphragm, quantitative 0.8ml urea solution is supplied. According to the measurement and control unit of instruction, in different speed of motor work, the corresponding demand for urea is provided. The urea solution from diaphragm reaches the mixing chamber through a one-way valve, and is mixed with the high pressure air supplied by the air compressor. The final urea solution in the compressed air blowing reaches atomizer to achieve atomization. A sensor for monitoring the pressure of the pipeline is arranged on the urea pipeline in the mixing chamber. Under normal operating conditions, the pressure of compressed air so that the air valve urea return port blocked. If the pressure sensors to monitor the urea solution pressure is too high, the control unit send instructions to air solenoid valve to reduce the air pressure before executing valve, thereby opening a reflux inlet, excessive urea goes back to the urea box by reflux inlet.

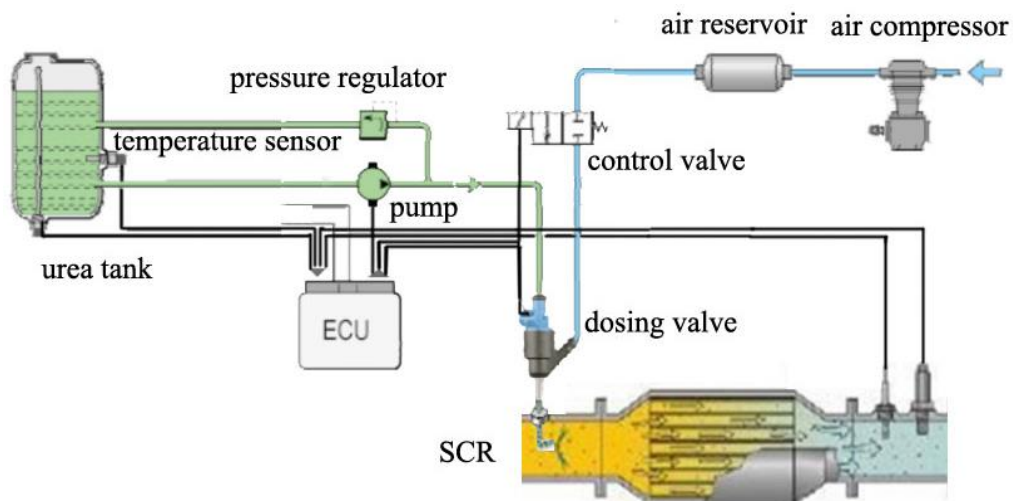


Figure 3. Air-Assisted Urea Supply System

Injection system of measurement is an important research domain in Urea – SCR. Its function is that urea solution can be sprayed into the exhaust pipe by fog according to the demand of Urea under the control of the Data Controlled Unit (DCU). Generally this system improves the quality of urea injection through improving nozzle structure and strengthening its atomization characteristics. It also improves the atomization results by improving the pipeline shape, position and angle of the spray. This way can promote the decomposition of urea, and it also can reduce the formation of deposits [7].

There are two methods to the mature technology of decomposing urea into NH₃: pyrolysis and hydrolysis. Currently the SCR of vehicles takes pyrolysis technology. This technology supposes that urea pyrolysis and catalytic going at once in an ideal world. The urea solution can be decomposed completely before entering the catalyst, and NH₃ mixes with the engine exhaust. But in actual condition, the distance between the urea aqueous solution injection and the catalyst entrance is very short, the decomposition of urea at the entrance is not sufficient. It also cannot guarantee that reductant and exhaust could be mixed uniformly, and it generates other by-products. The test indicates that, a mixture of NH₃, HNCO and urea enters into the catalyst due to the more undecomposed urea at the entrance of catalyst. HNCO has a polymerization with urea to biuret easily at higher temperature (about 150~200°C) because its activity is very high. It may form cyanuric acid and react with NH₃ to ammelide, ammeline, melamine and so on [8-10].

The rate constants of the quasi first order reaction of HNCO can be calculated by the following formula:

$$k = \frac{V}{W} \ln (1 - \eta) [\text{cm}^3 / (\text{g.s})] \quad (4)$$

In the formula, k is rate constant; V is gas flow velocity under the actual temperature and pressure; W is the quality of catalyst. Figure 4 is the relationship between the reaction rate constant and temperature of HNCO in 4 different catalysts. From the diagram, at low temperatures, the reaction rate constants of HNCO under different catalysts are different, but the gap is gradually decreased with the increase of temperature.

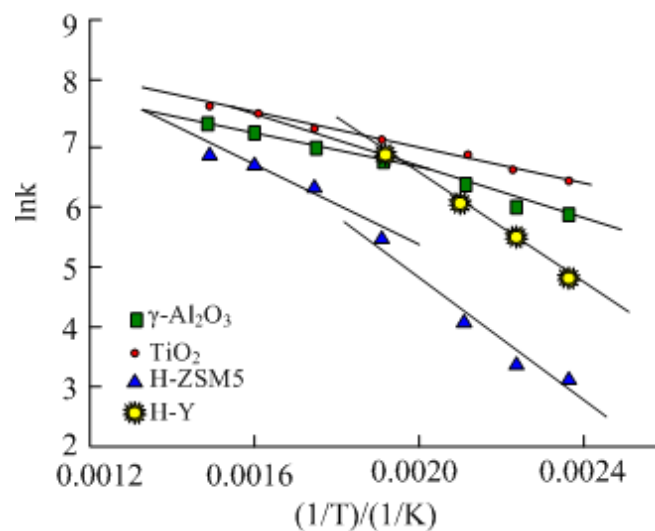


Figure 4. Reaction Rate Constants for Hydrolysis of HNCO for Catalysts

As shown in Figure 5, urea sediment can be divided into urea crystallization and urea stone according to the process of formation. Urea crystallization is due to the loss of water in urea solution which leads to over saturation and production of urea, which is the

product of process of physical reaction. With the increase of temperature, urea crystallization can continue to be decomposed. Urea stones were caused by the by-product caused by side effects of urea decomposition process, which belongs to chemical reaction products. Urea stones need a higher temperature to decompose. Because the quality of urea droplets is much larger than that of gas, the remain in the airflow stagnation zone can be formed crystallization, and if it cannot be decomposed completely in time, it will be the core of continuous growth. Because the crystallization cannot be completely decomposed, and ultimately urea stones were formed. Urea stones is accumulated to a certain extent, it is possible to plug the urea flow channel, and therefore the analysis of sediment generation need to study the process of urea decomposition carefully.

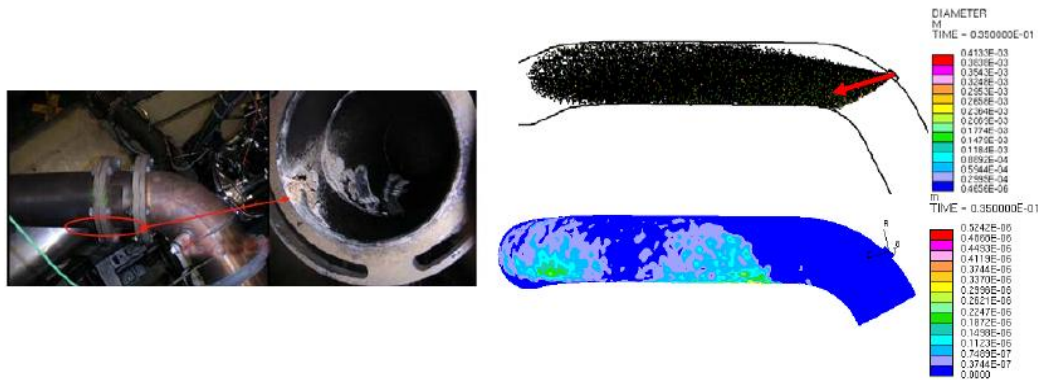


Figure 5. Urea Sediment and its Analysis

The Ford Motor Co studied the urea by experiment, the results showed that the sediment was produced by a period of time after a period of time under 300°C, and the main component of sediment was cyanuric acid and urea. But the sediment was not formed at 350°C [11].

Navistar company of the United States also carried out the study, the results show that in the low exhaust temperature, low ambient temperature, relatively large urea solution injection volume and relatively small exhaust flow, the sediment is the most easy to produce. Under these conditions, 25-65% injected urea solution is converted to the sediment. When the temperature is raised to 350°C, the amount of the injected urea solution converted is less than 1% [12].

Weichai Power Technology Center Yu Jiao's research [13] shows that in SCR system, urea decomposition deposition is an important factor that affects the efficiency of the catalytic converter. And urea decomposition and injection quantity and exhaust temperature have great relationship. The spray nozzle position of urea deposition is due to the nozzle flange design is not reasonable. In the position urea deposition was produced by the presence of a exhaust flow vortex. Urea deposition in the exhaust pipe is that urea injection angle is not reasonable. Urea solution is injected into the wall, and the wall temperature is reduced, so the result is deposition of urea in the wall. Urea deposition in SCR catalyst inlet is caused by no completely evaporation of the injected particles or the excessive amount of injection.

Some researchers [14] have studied and achieved some results by optimizing the arrangement of SCR system to improve the conversion efficiency of NO_x at low temperature and reduce the sediment generation, but it also needs further research to be able to effectively prevent the generation of sediment.

These deposits may clog the nozzle of urea and the catalyst carrier. It also may affect the activity of catalyst and increase the consumption of urea. These repercussions bring about the difficulty to control the SCR system. Thus the cryogenic sediment of urea is a hot issue of SCR field in the current [15-16].

It is common practice to install mixer before the SCR catalyst for promoting the evaporation, decomposition of the urea and the mixture of exhaust. Numerical simulation and experiment show that mixer is helpful to reducing the generation of sediment and improving conversion efficiency of SCR [17-19].

3.3. Catalysts

Catalyst has three basic parts: the coating, carrier and package [20]. The study of catalyst focuses on the choice of chemical reaction coating, internal carriers of catalyst and the design of muffler.

Coating what consists of water coating and activity components of catalytic is the core of the catalyst [21]. Urea-SCR catalyst bases on vanadium-based catalyst ($\text{TOi}_2\text{-V}_2\text{O}_5$) and zeolite catalyst. The advantages of vanadium-based catalyst are high conversion efficiency, high and wide temperature window, good resistance to sulfur and low cost. But the volatilization of vanadium will lead to inactivate when the temperature is higher than $600 \sim 800$ °C. The temperature from heavy engine to the surface of catalyst cannot reach such a high level, so it is applied to the domestic heavy engine. By comparing conversion efficiency of NO_x with Fe, Cu zeolite catalyst and alumina-based catalyst, we can know that Cu zeolite catalyst have a good low temperature performance. Fe zeolite catalyst have a good high temperature performance, while the performance of alumina-based catalysts is somewhere in between. However, the composite zeolite catalyst of copper and iron can integrate the all advantages [22-24].

The conventional coating application of SCR catalyst also includes catalyst that is used to remove ammonia. This catalyst is named AMOX (Amm on Oxidation catalyst). And it is also be used to remove NH_3 that is remained after the main reaction of SCR. Alternative technology is that we installs the after-oxidation catalytic converter after the SCR catalyst. But there are problems with these solutions. Not only may oxidise these solutions from NH_3 into NO_x as to increase pollution and system cost, but also cause waste of urea injection to a certain degree. So we place the before-oxidation catalysts before the SCR catalyst as the pretreatment device of engine exhaust generally.

Carrier is the skeleton part of SCR system, AMOX and coating are coated on the carrier. Most of carrier is a whole which has cylindrical cellular channel. Its basic material can be divided into two categories: metal and ceramic based. Metal-based carrier is small, and it can be made into various shapes to make the design of catalytic converters easy. But the cost of it is higher. Ceramic carrier is mainly composes of silicon carbide and cordierite. Cordierite is mainly used in heavy engine. The advantages of cordierite are high temperature resistance and high mechanical strength. Its disadvantages are the poor corrosion resistance and small thermal conductivity. In addition, there is a poisoning effect on the noble metal catalyst because cordierite contains a large of SiO_2 (55% or more). SiC mainly is applied to small-displacement engines. It has better performance than cordierite [19]. The advantages of SiC are high porosity and strong thermal diffusion ability. The disadvantages are high cost of preparation and the need of adhesive for a bigger volume. Its wall thickness and porosity of the carrier are affected by the accuracy of machining, durability of catalysts and influence of packaging process.

Package for SCR catalyst that is used to connect the carrier with the exhaust pipe is the outer packing of carrier. It can be divided into packages with silencer and without silencer according to the different functions as the outer packing of carrier. It also can be divided into boxes and barrels type package depending on the arrangement of whole vehicle. Box package's advantages are remarkable noise-reducing effectiveness, lower airspeed and higher efficiency of catalytic reaction. But there is a certain degree of difficulty on vehicle layout because of the large volume. It is suitable for large vehicles and trucks. Barrel type package is convenient for layout because the volume of post-processor close to the muffler of original vehicle. Its performance is slightly worse than the box-type post-processor due to the lack of structural design of muffler and the high airspeed of catalytic.

So the barrel type package is mainly suitable for small space vehicles and city buses. The most important parameters of diesel SCR catalytic converter package are the uniformity of flow field and entire backpressure of catalytic converters.

3.4. Control Unit

The control unit of Urea-SCR collects signals from the operation condition of the engine ECU and the sensor of SCR system. It also treats with these signals. And it can calculate the demand for urea according to the strategy of urea injection. At the same time, it takes the signal to the corresponding execution unit and controls the urea injection. In addition, it has the function of fault inspection and diagnosis. Progress in the control unit is mainly reflected in two aspects: software and hardware. The key of hardware is the further study of the sensor, and the key of software is the control strategy of urea injection.

The control strategy of urea injection can be divided into open-loop control and closed-loop control from the structure. The main difference is whether it uses signal of NH_3 sensor (or NO_x sensor) which is loaded in downstream of SCR for feedback control. SCR open-loop control strategy system doesn't have the ability to correct automatically or compensate. SCR closed-loop control strategy can improve the accuracy of the control and stability, and the response time is shortened. However, there is still a lack of sensor sensitivity and the cost issues, owing to the cross-sensitivity of NO_x sensor and the immature technology of NH_3 sensor [25-27]. Current solutions include stimulus-response method and adding post-oxidation catalytic converter.

The urea injection control strategies of SCR system can be divided into mapping based on pulse spectrum [28] and model based on calculations from the controller design [29]. The control strategy of model based on calculations is complexity, and it has few applications. The structure design ideas of open-loop urea injection control strategies based on the pulse spectrum is simple, and its development difficulty is low. So there is more widely application in engineering.

SCR system requires fine control of urea to meet the strict emissions standards. However, feedback control and the optimization of control strategy are very difficult because the SCR system involves many aspects.

4. Conclusions

Follow the author information by two blank lines before main text. For the present, with the increasingly strict emission standards, the optimization of fuel economy and system design costs will become an important constraint condition to the control of diesel engine emission. In this condition, the main research of urea storage device focuses on alternatives of urea and storage problem of urea solution under the low temperature. The research direction of urea supply system is that we need improve the low temperature properties of SCR technology and the efficiency of the decomposition of urea. Catalyst is the core of SCR technology research, and it trends for reducing the installation dimension and cost of the system under the premise of meeting emission standards. In addition, it will develop a new type of catalyst and reductant. The main research direction of control unit is that we need develop new sensors and more precise control strategies.

In recent years, China increases investment in science and technology for environmental protection. It makes great progress in the overall level of science and technology, and it reaches or approaches international advanced level in some areas. Research Center for Eco-Environmental Sciences, Tsinghua University, the PLA Academy of Military Transportation, China Environmental Science Research Institute and many other domestic research institutes all study for purification technology of diesel exhaust. Dalian University is studying the closed-loop control strategy of SCR based on pulse spectrum. Experiments show that this strategy can improve the catalytic efficiency

of SCR, and it also can reduce the difficulty of the SCR system control. So it will achieve more precise feed control. This SCR can open a new direction of diesel engine exhaust gas purification.

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