Urea Injection and Uniformity of Ammonia Distribution in SCR System of Diesel Engine

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Abstract

The kinetic mechanism of chemical reaction was used to calculate the coupling of the fluid kinetics with the urea decomposition reaction and the SCR reaction kinetics. Combined with the engine test and simulation, the distribution uniformity of the urea injection and ammonia gas was studied. Through numerical simulation on urea spray and exhaust flow in Urea-SCR system, the flow field characteristics in whole after-treatment system are gotten. By using numerical calculation in different urea injection angle and orifice sizes, the urea-crystallization and ammonia distribution have been studied and the optimal urea spray angle and nozzle size are given.

Keywords: Urea-SCR system; spray parameter; ammonia concentration distribution; uniformity.
AMS 2010 codes: 11B30.

1 Introduction

With the improvement of national emission regulations, the control of NO_X emission is becoming more and more stringent, and the after-treatment technology of diesel engine is facing new challenges [1, 2]. Only internal purification measures have not been to meet the new emission regulation requirements for NO_X emission. Selective Catalytic Reduction (SCR) catalyst is one of the most promising technologies for controlling NOx emissions by using ammonia as an active intermediate for NOx reduction [3]. Based on the national condition, Urea-SCR technology will be the main technical direction of diesel engine emission in the future, because of its high efficiency, high selectivity, high economy and sulfur tolerance [4].

The urea spray system is very important in the selective catalytic reduction technology [5–8]. The spray morphology of the urea solution and the atomization of the droplet directly affect the evaporation and decom-

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position process, and affect the concentration and distribution uniformity of the ammonia in the front of the catalyst carrier [9, 10]. The uniformity of ammonia has an important influence on the conversion rate of NOx in SCR system. If the distribution of the flow field is not uniform, such as too many local NH3, this will cause local low temperature zone and is easy to produce urea crystallization, and excess ammonia also cause NH3 leakage. While local NH_4 is too rare, it will cause the reduction of NOx conversion efficiency [11]. Therefore, the study of the urea flow field and the uniformity of ammonia distribution has a positive guiding significance for inhibiting crystallization and improving the conversion efficiency of NOx.

2 Methodology

2.1 Principle and theoretical basis of Urea-SCR

2.1.1 Working principle of Urea-SCR system

The Urea-SCR system is a device by which to control the exhaust emissions of diesel engine. Its diagram is shown in Fig. 1. The concentration of 32. 5% urea solution is injected into the exhaust pipe. Then the urea solution is decomposed into ammonia and carbon dioxide at the high temperature of the exhaust. Under the action of the catalyst, the ammonia gas reduces the NOx into the harmless nitrogen and water.
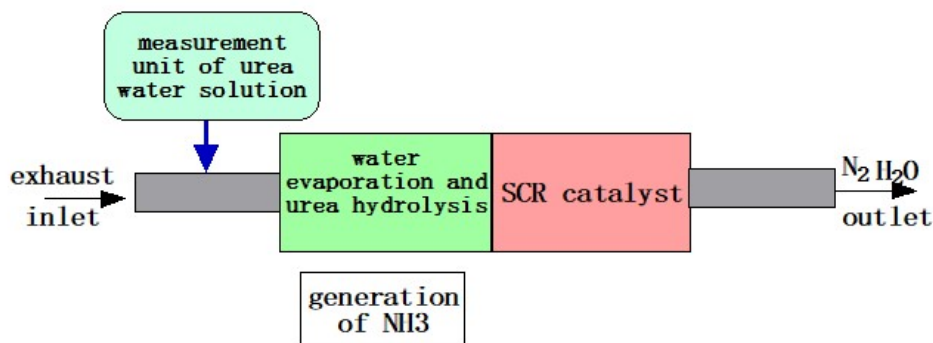

Fig. 1 Diagram of the principle of SCR system.

2.1.2 Chemical reaction

1) The formation of ammonia After the urea solution is ejected into the exhaust pipe, the urea water solution is first evaporated to precipitate the urea particles in the exhaust pipe and the catalytic converter. By the urea pyrolysis reaction, equimolar ammonia and isocyanic acid are produced [12, 13].

$$CO(NH_2)_2 \rightarrow NH3+HNCO \tag{1}$$

The further hydrolysis of isocyanic acid is:

$$HNCO +H_2O \rightarrow NH3+CO_2 \tag{2}$$

2) Catalytic reduction reaction of NOx Standard SCR reaction:

$$4NH_3+4NO+O_2 \rightarrow 4N_2+6H_2O \tag{3}$$

Rapid SCR reaction:

$$4NH_3+2NO+2NO_2 \rightarrow 4N_2+6H_2O \tag{4}$$

Equation (3) and (4) are the main reactions in the catalytic reduction reaction of NOx. NOx in diesel engine exhaust emissions is given priority to with NO, NO usually account for 85%-95% of the nitrogen content. Therefore, the chemical equation (3) is the most important reaction in the catalytic reduction reaction of the NOx, But the reaction priority of chemical equation (4) is higher than that of equation (3), so, NO_2 and a part of NO in exhaust gas can be quickly eliminated by chemical equation (4). After the reaction of chemical equation (3) and (4), most of NOx was converted to harmless N_2 and H_2O.

2.1.3 Surface reaction model of catalyst

The emission of NOx in diesel engine is mainly NO and NO_2 is very few. Therefore, the standard SCR reduction reaction is mainly considered in the catalytic reduction of NO. The essence of NH_3 activation dehydrogenation is oxidized at the surface stage of the catalyst. Therefore, the oxidation of the NH_3 adsorption site or activation position determines the dehydrogenation rate and dehydrogenation of NH_3. The researchers generally believe that NH_3 selectively catalyzes the reduction of NO to satisfy the Eley Rideal mechanism. This mechanism holds that NH_3 is first adsorbed on the active surface carrier in the process of catalytic. The NH_3 adsorbed on the surface reacted with the weakly adsorbed O_2 and NO. Buzanowski, Yang and other proposed that the reaction satisfies the Arrhenius theorem and proposes the following reaction rate equation [14]:

$$\dot{r}_{NO} = k_1 c_{NO} \phi_{NH_3} \tag{5}$$

In equation (5), $\dot{r}_{NO}$ is the reaction rate and C_{NO} is the concentration of component.
k_1 is the reaction rate constant. According to the Arrhenius equation, can be obtained by the equation

$$k_1 = A \exp \left\{ \frac{-Ea}{RT} \right\} \tag{6}$$

The A and -ea in equation (6) are the frequency factor and activation energy of the reaction rate constant. ϕ_{NH_3} is adsorption rate and can be expressed by the following equation [15].

$$\phi_{NH_3} = \frac{K_{NH_3} C_{NH_3}}{1 + K_{NH_3} C_{NH_3}} \tag{7}$$

In the equation (7), K_{NH_3} is the adsorption equilibrium constant for NH3 and C_{NH_3} is the concentration of NH_3. The equation (6) and (7) were substituted into (5), and the reaction rate constant was obtained.

$$\dot{r}_{NO} = A \exp \left\{ \frac{-Ea}{RT} \right\} \frac{K_{NH_3} C_{NH_3} C_{NO}}{1 + K_{NH_3} C_{NH_3}} \tag{8}$$

2.1.4 NH_3 distribution uniformity

The uniformity of NH_3 distribution before catalytic converter has important influence on NOx conversion efficiency and catalyst life. The low concentration of NH_3 will lead to the low conversion efficiency of NOx, and too much NH_3 will lead to crystallization and NH_3 leakage. The uneven distribution of NH_3 for a long time will also lead to heterogeneous aging of catalyst, which will affect the life of catalyst and the performance of SCR system. There are many kinds of uniformity evaluation [16]. In the uniformity evaluation of NH_3 distribution, there are two main kinds of relative standard deviation CV and homogeneity index r. Wang et al. [17] applied relative standard deviation CV to optimize the ammonia distribution uniformity of marine engine. According to the literature [16], the relative standard deviation CV has low stability, while the uniformity index r has better stability. The uniformity index is an evaluation standard for velocity uniformity of flow field established by Herman Weltens and others in the early 90s. Based on the definition of statistical deviation, the uniformity index definition is proposed [18]. The following equation (9) shows that the equation can comprehensively reflect the distribution of NH3 concentration in the whole flow section, which has strong comparability and applicability.

The evaluation index of homogeneity in this study is uniformity index r.

$$\gamma = 1 - \frac{1}{2n} \sum_{i=1}^{n} \frac{\sqrt{(ci - \bar{c})^2}}{\bar{c}} \tag{9}$$

In equation (9), n is the number of measurement points of cross section, and ci is the concentration of NH_3 at the point i. $\bar{c}$ is the average concentration of NH_3 on the whole section. The r is larger and it's value is close to 1, indicating the more uniform distribution on the selected cross section.

2.2 Establishment of numerical simulation model for Urea-SCR system

The three-dimensional numerical calculation model includes the drawing of the geometric model, the division of the grid, the establishment of the calculation model and the setting of the calculation boundary. This study is based on the original processing system for geometric modeling and three-dimensional numerical calculation.

2.2.1 The establishment of geometric models and the division of computational grids

(a) original model.   (b) simplified model.
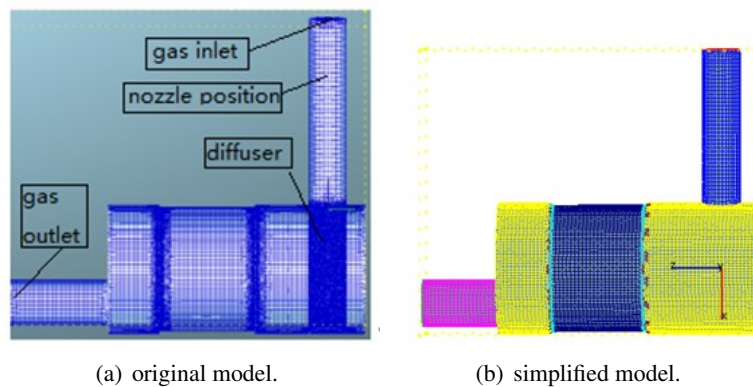
Fig. 2 Catalytic converter

The research object is the post-processing system of 6114 diesel engine, and its SCR catalytic converter is a reversing structure. First, the three-dimensional solid diagram is drawn according to the object, and the 3D solid diagram is derived in the STL format. In order to ensure the quality of the grid and the convergence of the calculation, the small hole part of the diffusion device needs to be refined. As shown in Fig. 2 (a), the number of grids in the whole model is close to 2 million, and the calculation time will cost several times. According to the [19] 58 pages of the literature, the calculation of diffuser and the calculation of no diffuser have little effect on the conversion efficiency. So the variable parameter study of the system can be simplified as a non-diffuser structure to calculate. The simplified calculation model grid, as shown in Fig. 2 (b), is 260 thousand.

2.2.2 Calculation model and boundary setting

In this study, the simulation of exhaust flow in the post-processing system is based on the basic conservation law of conservation of mass, conservation of momentum and conservation of energy to solve the mean transport equation.

The urea jet takes air-assisted ejection, that is, AdBlue and compressed air are ejected at the same time from the nozzle. In the standard condition, the consumption of compressed air is relatively small relative to the discharge flow, so the injection of the urea solution is considered only in the simulated spray, and the auxiliary air is ignored. The simulation of evaporation, pyrolysis and hydrolysis after spray of urea solution is a complicated process. The SCR- model [20] is used in this study. First, the droplets of the spray are heated and evaporated by the tail gas. Because the boiling temperature of water is lower than urea's, at first pure water droplets are complete evaporated. Then Urea $(CO(NH_2)_2)$ is pyrolyzed into NH_3 and isocyanate (HCNO), such as reaction

equation (10). The specific process of evaporation and pyrolysis is shown in Fig. 3. So, the evaporation rate of water and the pyrolysis rate of urea can be described independently, and the evaporation rate can be calculated by Ambrazon-Sirignano evaporation model.

The flow characteristics of the catalyst are simulated by the porous medium model, and the pressure drop calculation is calculated by using the Forchheimer model (AVL porous boundary model).
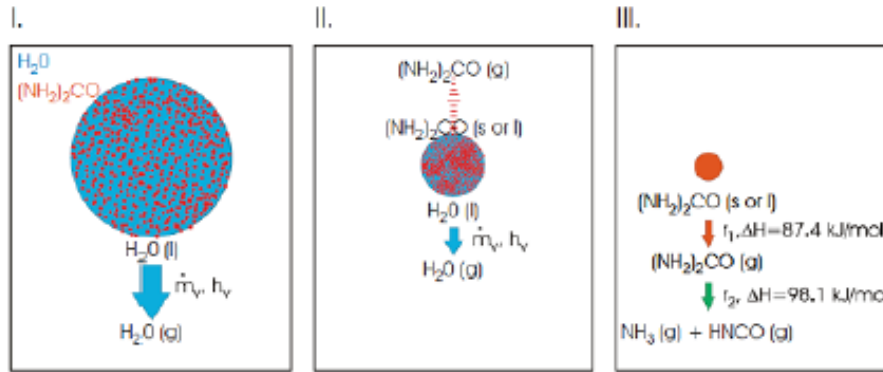
$$(NH_2)_2CO\,(liquid)\ NH_3\,(gas) + HNCO\ (gas) \tag{10}$$

Fig. 3 Evaporation of droplet and pyrolysis of urea.

A100, B100 and C100 of thirteen working conditions are selected to simulate. The simulation optimization of injection parameters is carried out by B100 working conditions. A, B and C represent the high and low speed in the calculation conditions, and the numerical values of specific rotational speed, exhaust temperature, tail gas mass flow, urea injection and NOx concentration under different working conditions is shown in Table 1. The boundary in the calculation is input to the actual experimental value, and the ejection urea is 32. 5% saturated urea aqueous solution. The inlet boundary condition adopts the method of given mass flow and temperature. In the turbulence parameter, the turbulence velocity takes 10% of the inlet velocity, and the characteristic length is 10/% of the inlet diameter. The outlet is set to the static pressure boundary condition and the pressure is 0.101 MPa. The mass fraction of the imported components is calculated based on the volume concentration measured by engine bench test.

Table 1 Boundary condition of typical working condition

point	Rotation (r/min)	Exhaust temperature(° C)	Gas mass (kg/h)	urea injection (ml/h)	NO_X concentration (10^{e-6})
A100	1349	477	811.77	3300	1660
B100	1672	440	1190.86	3400	1230
C100	1994	412	1430.11	3200	950

2.2.3 Verification of model

Fig. 4 shows the calculation results and the experimental results of NO_x conversion rate in three different working conditions. As shown in Fig. 6, the NO_x conversion rate of the three-dimensional numerical simulation is compared with the NO_x conversion rate obtained by the experiment. Under the three conditions, the NO_x conversion rate is slightly lower than the test results. This is mainly due to the existence of the diffusion hole in the test device, and the existence of the diffusion hole here is helpful to the better mixing of the urea spray and the tail gas, and the catalyst can be improved. The distribution in the front of the carrier is homogeneous,

and the three-dimensional numerical simulation is simplified. Therefore, the conversion rate of NO_x is slightly reduced, but the basic trend is consistent, and the error is less than 5%. It shows that the calculation model can well simulate urea injection and internal physical-chemical process of SCR catalysts, and the selective catalysis established is verified. The effectiveness of the reduction system model is instructive in the optimization of urea injection parameters and the design of SCR catalyst.
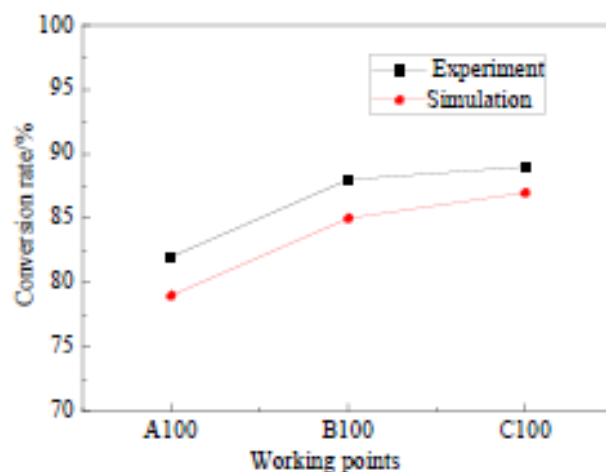
Fig. 4 Comparison of catalytic conversion rate between calculation and test.

3 The calculation results and analysis

The nozzle of the post-processing system in this study, as shown in Fig. 5, has four diameter 0.5 mm holes in the circumferential distribution. The influence of different spray directions on the spray flow field and the distribution uniformity of NH_3 was studied, and the injection position was 481mm from the axis of the catalytic converter. The research of spray parameters mainly focuses on the analysis and research of variable parameters for B100 working condition.


Fig. 5 Urea nozzle.

3.1 Calculation analysis of different injection angles

In urea spray parameters, the choice of the spray cone angle is critical. In this study, three dimensional numerical analysis is carried out for different injection angles. The injection angle of the original nozzle is 179

degrees. When the spray cone angle is too large, the droplets with large diameter are affected by the gas flow velocity, which can easily hit the tube wall and form the crystallization. The injection Angle is gradually reduced and the 3-D numerical analysis is carried out for different injection angles.
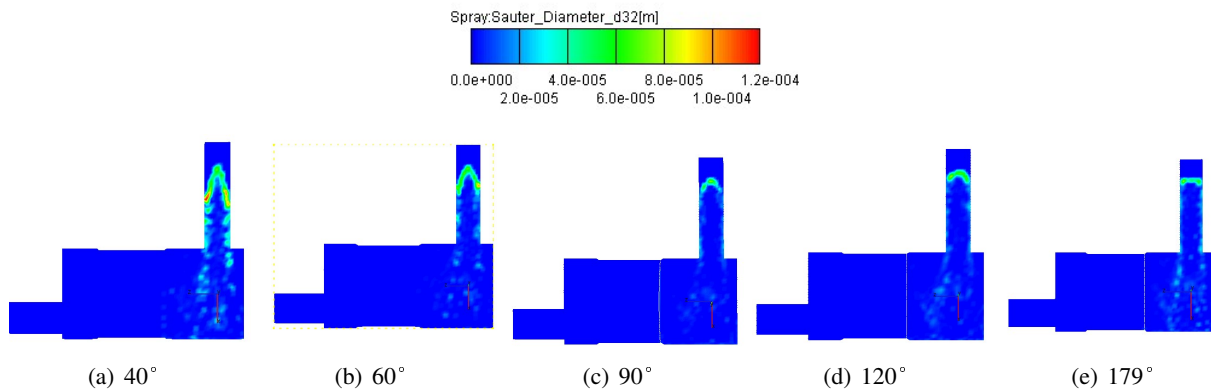
Fig. 6 Particle distribution at different injection angles (Y section)

3.1.1 Spray distribution of urea

Fig. 6 is the particle distribution map of the Y section at different angles of injection. It is shown from the diagram that the Sauter diameter of the urea particle is larger when it is sprayed, and the excessive spray cone angle makes the urea particle hit the wall. Part of them bounce back after the wall collision, and some of the urea particles attached on the wall which need to absorb the heat in process of evaporation and pyrolysis of urea hydrolysis that will cause the decrease.of local temperature. As shown in Fig. 7, the temperature distribution map (A, B, C, D) shows that it is easy to form crystallization. With the decrease of spray cone angle from 179 degrees, urea evaporated, pyrolysis and hydrolyzed under heating and airflow blowing, and its Sauter diameter decreased. But when the spray cone angle descends to 40 degrees, we can see from Fig. 6 that under the same ruler, the Sauter diameter of urea particles is larger. This is mainly because the 40 degree spray cone angle is too small, when the urea solution is injected into the exhaust pipe of the exhaust gas, the larger gas flow velocity will blow some drops with the flow in the cylinder, reducing the time of the urea pyrolysis and hydrolysis, resulting in the low average concentration of NH_3 and easy to cause ammonia leakage.

3.1.2 Concentration distribution and wall film deposition of NH_3

The uniformity of NH_3 distribution in the Usea-SCR system has an important effect on the leakage of ammonia and the conversion rate of NOx. If the NH_3 distribution is not uniform, the NH_3 is too much in the local region lead to NH_3 leakage, and the conversion efficiency of NOx is too low in some other NH_3 rarefied regions. The study of the influence of NH_3 uniformity on NH_3 leakage and NOx conversion efficiency in a foreign institution concluded that with the improvement of the uniformity index, the conversion efficiency of NOx increased and the leakage of NH_3 decreased rapidly. In addition, the uneven distribution of urea droplets will cause low local temperature, high concentration of urea and easy to crystallize, which will plug the exhaust pipe and cause the engine power decline [21].

Fig. 8 is shown the NH_3 concentration distribution of the entrance section of SCR carrier at different injection angles. When the injection angle is 40 degrees, the crystallization of the urea particles is less in the wall of exhaust pipe. Therefore, from Fig. 8, the concentration of the inlet NH3 at 40 injection angle is higher than that of the injection angle of 179, 120 and 90 degrees, but the distribution uniformity is poor, and the wall has a poor uniformity. Excessive concentration of NH_3 exists, which is very likely to cause ammonia leakage and cause secondary pollution to the environment.

Combined with Fig. 6, Fig. 7 and Fig. 8, we can see that when the injection angle is 60 degrees, the

(a) 40°   (b) 60°   (c) 90°

(d) 120°   (e) 179°
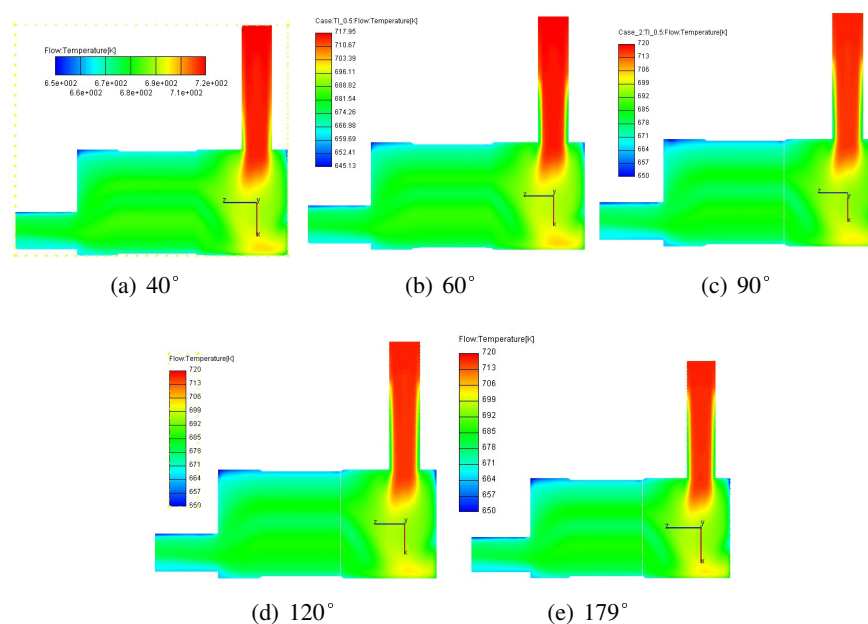
Fig. 7 Temperature distribution at different injection angles (Y section).

(a) 40°   (b) 60°   (c) 90°   (d) 120°   (e) 179°
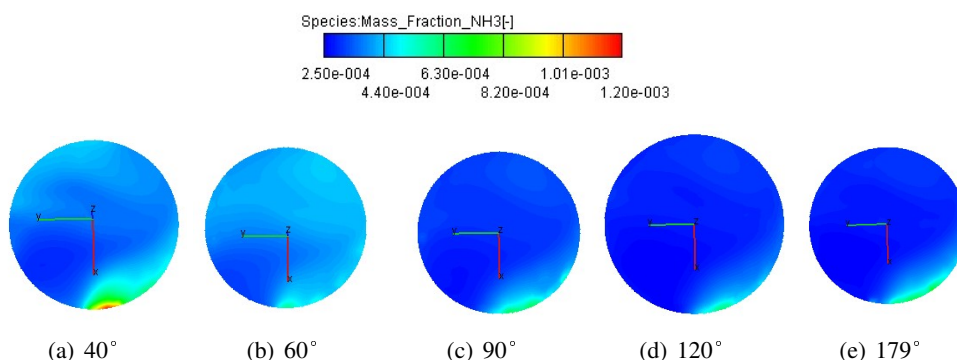
Fig. 8 Mass-fraction of NH_3 in Inlet cross section of carrier at different injection angle

concentration of the inlet section of the carrier is high. And it can be seen from Fig. 9 that the concentration distribution is more uniform. This is mainly because the urea is ejected by the nozzle, and most of the particles enter the tube to flow rapidly with the airflow, and a small number of particles hit the wall to form a small amount of crystallization.

Fig. 9 shows the comparison of the distribution uniformity of NH3 in inlet section of catalytic carrier at different injection angles. The uniformity of the NH_3 distribution of the carrier entrance section will directly affect the reducibility of the NH_3, thus affecting the catalytic conversion efficiency of the NOx. It can be seen from the Fig. 9 that the uniformity of the entry entry section with the injection angle of 60 degrees is the best. When the injection angle is too large or too small, it will affect its evenness. Such as the injection angle is 40 degrees, the uniformity of its entry section is the lowest.

Fig. 10 shows the mass of the wall film at different injection angles. The mass of the wall film increases with the increase of reaction time. The mass of the wall film is different for different injection angles. When the injection angle is large, the growth rate of the wall film is the fastest. The main reason is when the injection angle is large, most particles are sprayed directly to the wall, then a part of them bounced back after impingement and another part of them stick on the wall to cause crystallization. While injection angle is too small, as the urea
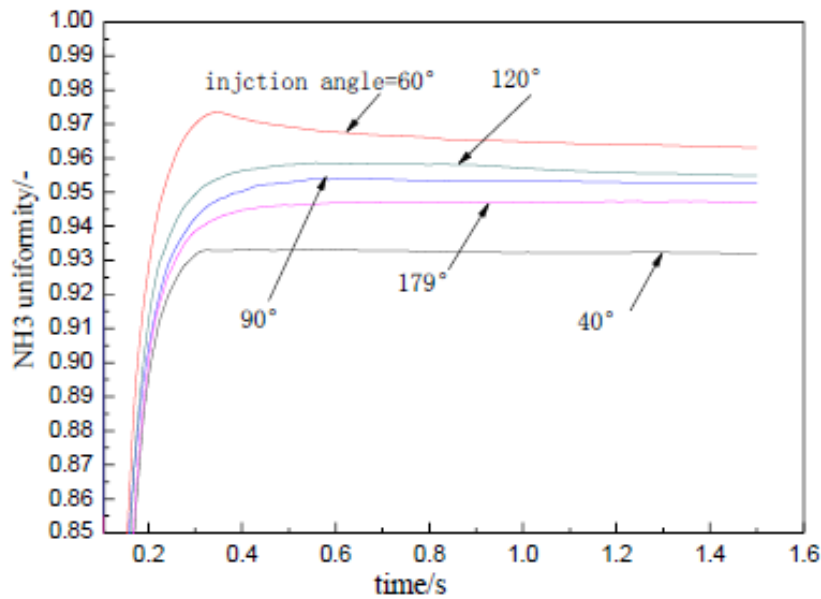
Fig. 9 Distribution uniformity of NH$_3$ in the inlet section of catalytic carrier.
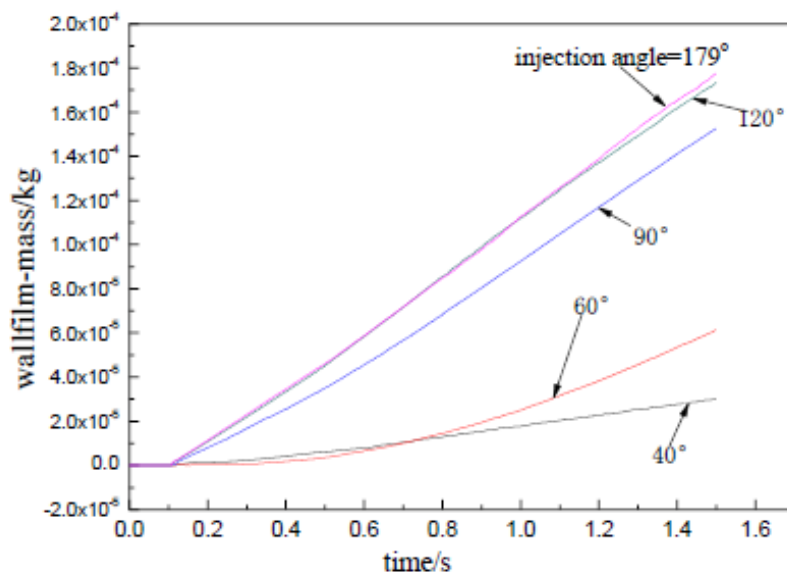

Fig. 10 Comparison of wallfilm mass at different injection angles.

particles flow forward directly with the exhaust and the wall-impingement particles are few, so the growth speed of the wall film with the small injection angle (40 degree angle) is the slowest.

In order to evaluate the injection angle, it is necessary to integrate many characteristics. Through the analysis and research, it can be found that the injection angle of 60 degrees is the optimal injection angle. At this angle, the wall film mass is low and the distribution of the NH$_3$ in the carrier entrance section is well distributed, which helps to reduce the crystallization of the urea in the tube wall and improve the conversion rate of NO$_X$.

3.2   Calculation and analysis of different nozzle diameters

3.2.1   Urea spray distribution and NH$_3$ concentration distribution in different nozzle diameters

The difference in the size of the urea jet has an effect on the spray of urea and the distribution of NH$_3$ concentration, so the spray distribution and NH$_3$ concentration distribution of different orifice diameters are studied.

Fig. 11 is the particle distribution map of Y section under different nozzle diameter. From the diagram, we find that the sauter diameter of the urea particle is larger when the nozzle is enlarged. Fig. 12 is the mass comparison of the wall film. It can be seen from the drawing that the increase or decrease of the jet holes will increase the wall film mass. This is mainly due to the better atomization effect and the increase of the evaporation and hydrolysis of urea while the nozzle hole is small. While the nozzle is small, the atomization is good and can enhance the evaporation of the urea, hydrolysis, and so on. But under the same injection pressure, smaller nozzle causes of urea injection speed increase and urea particles more easily impinge on the wall. And enlarging the nozzle aperture, the atomization quality is reduced, so that the evaporation and pyrolysis of particles are affected, so the mass of the wall film increases rapidly.

(A) 0.5 mm   (B) 0.4 mm   (C) 0.6 mm
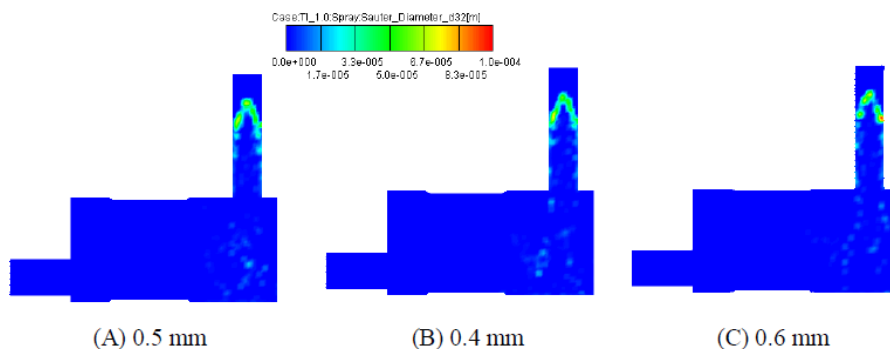
Fig. 11 Particle distribution with different nozzle diameters (Y cross section).
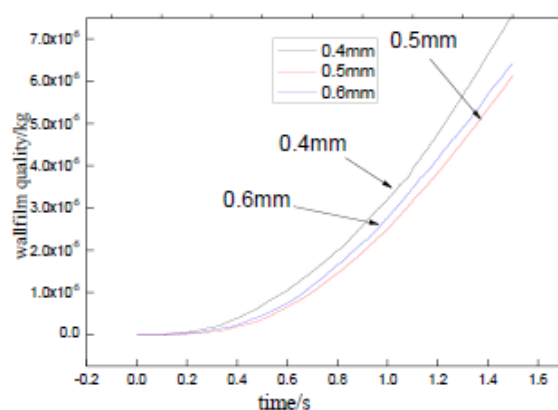


Fig. 12 Wall film mass comparison diagram under different nozzle diameter.

Fig. 13 is shown the comparison of the material fraction of NH$_3$ in the entrance section of catalytic carrier under different nozzle diameter. Fig. 14 is shown the comparison the distribution uniformity of NH$_3$ in the
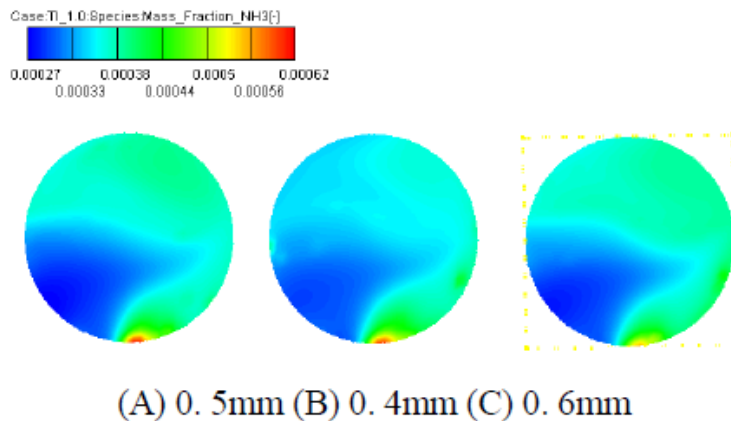
Fig. 13 Mass-fraction of NH$_3$ at different entry diameter.

entrance section of catalyst carrier under different nozzle diameter. From Fig. 13, it can be seen that the difference of the material fraction is small and the NH$_3$ distribution concentration in the 0.6 mm nozzle hole is slightly lower. From Fig. 14, it can be found that, in three cases, the uniformity of NH$_3$ distribution is not very different. The uniformity of NH3 distribution in 0. 6mm nozzle hole is slightly lower than that of the other two cases. Therefore, it can be concluded that the best diameter of urea nozzle is 0.5 mm.
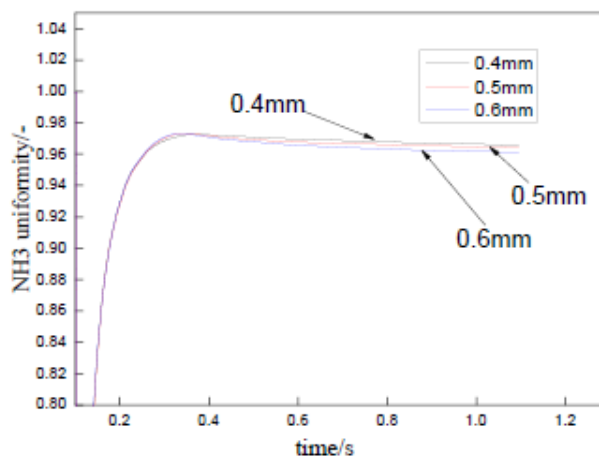

Fig. 14 Uniformity of NH$_3$ in inlet section of catalytic carrier under different nozzle diameters.

Through calculation and comparison with different urea spray angle and spray hole size, it is concluded that the 60 degree injection angle and the injection hole of 0. 5mm are the best. The chemical reaction of the B100 condition is calculated, and the catalytic conversion rate of NOx is increased from 85% to 91%, which is increased nearly 6%.

3.3 Experimental research

This research takes the post-processing system of vehicle 6114 diesel engine as the research object, and the engine post treatment test bench is shown in Fig. 15. Through numerical simulation, we get the best injection parameters. The injection angle and the size of the jet hole are improved and it is verified by experiments. As is shown in Fig. 16, the efficiency of the NOx conversion is obviously improved and the increase rate is from

2.53% (working point 2) to 5. 75% (working point 6).
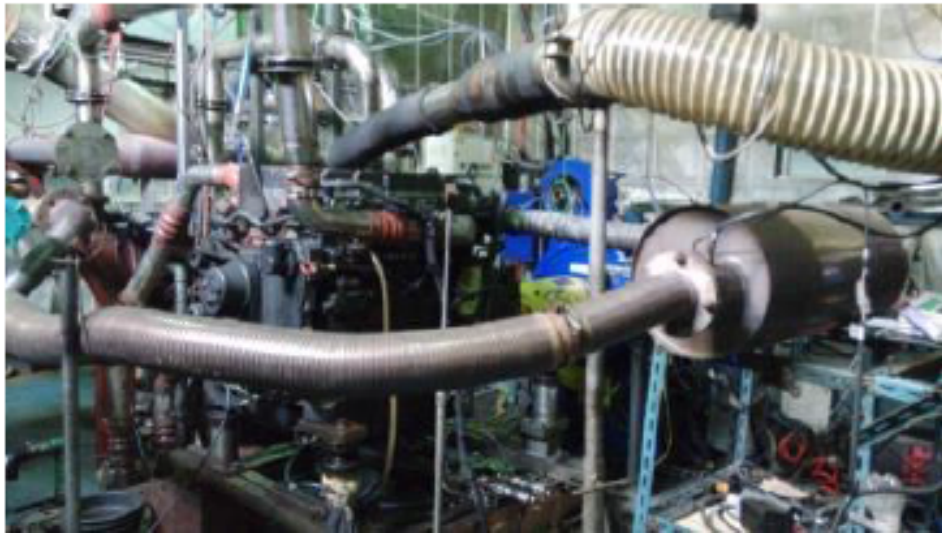
Fig. 15 Engine post treatment test bench.
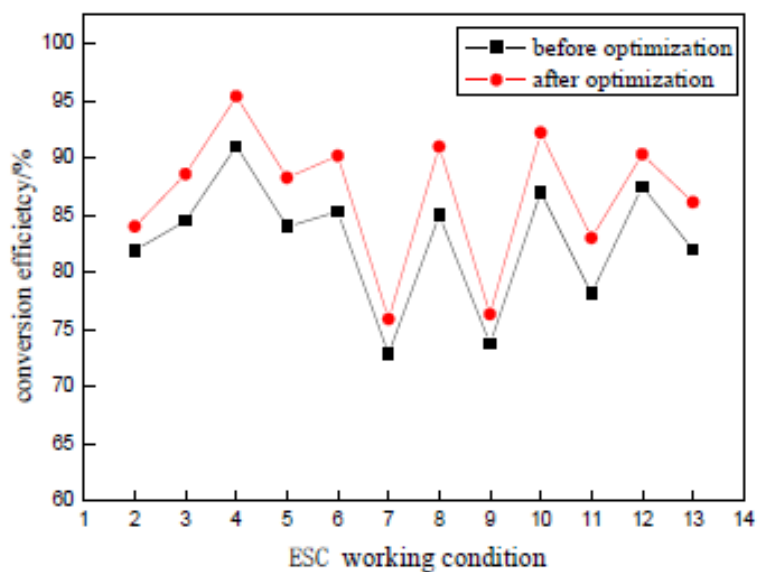
Fig. 16 Comparison of NOx conversion efficiency.

4 Conclusion

Through the three-dimensional numerical simulation of the Urea-SCR system, the urea injection and the uniformity of the flow of tail gas in the Urea-SCR system are studied. The distribution of the urea particles, the distribution of ammonia gas and the distribution of the wall film are studied and predicted in different working conditions. The area of the urea crystallization is also predicted. The following conclusions are drawn:

(1) The study was done about the variable parameters of urea injection. The effect of the jet cone angle and

the diameter of the nozzle on the uniformity of the NH$_3$ distribution is obtained. The large or too small injection angle will affect the distribution of the urea particles and the velocity of the pyrolysis reaction, and will affect the concentration and distribution uniformity of NH$_3$ in the carrier inlet. There is little influence on the distribution uniformity of urea while the nozzle is with different pore sizes, but it has great influence on the mass of the wall film. Under the same injection pressure, the smaller nozzle makes the rate of urea jet increasing and the particles are more easily impacted on the wall, so the wall film mass increases rapidly. If the diameter of the nozzle is too large and the atomization quality is reduced, so the evaporation and hydrolysis of the particles are affected and it also causes the rapid growth of the wall film mass.

(2) The over large spray angle causes a large number of droplet to hit the wall, and the evaporative heat absorption leads to further reduction of temperature and easy formation of crystallization. The over small injection angle is greatly influenced by the exhaust flow of urea particles, so that the urea particles do not have enough time for pyrolysis and hydrolysis, which can cause the leakage of NH$_3$, thus affecting the conversion efficiency of NOx.

(3) Through calculation and test verification, it was concluded that the NOx conversion efficiency improved greatly with the new injection parameters.

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