Development of a variable resistance-capacitance model with time delay for urea-SCR system

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ABSTRACT

Experimental research shows that the nitric oxides (NO_x) concentration track at the outlet of selective catalytic reduction (SCR) catalyst with a transient variation of Adblue dosage has a time delay and it features a characteristic of resistance-capacitance (RC). The phenomenon brings obstacles to get the simultaneously NO_x expected to be reduced and equi-molar ammonia available to SCR reaction, which finally inhibits NO_x conversion efficiency. Generally, engine loads change frequently, which triggers a rapid changing of Adblue dosage, and it aggravates the air quality that are caused by NO_x emission and ammonia slip. In order to increase the conversion efficiency of NO_x and avoid secondary pollution, the paper gives a comprehensive analysis of the SCR system and tells readers the key factors that affect time delay and RC characteristics. Accordingly, a map of time delay is established and a solution method for time constant and proportional constant is carried out. Finally, the paper accurately describes the input-output state relation of SCR system by using "variable RC model with time delay". The model can be used for a real-time correction of Adblue dosage, which can increase the conversion efficiency of NO_x in SCR system and avoid secondary pollution forming. Obviously, the results of the work discover an avenue for the SCR control strategy.

Keywords: Diesel engine, Urea-SCR, Weight analysis, Time delay, Resistance-capacitance

1. Introduction

With the increasing demands for low engine emissions, reducing the NO_x which is produced by heavy duty diesel vehicles has become one of the major tasks. Urea-SCR (selective catalytic reduction) systems possess an outstanding performance in NO_x reduction, which can be served as a technology platform for further improvement. Therefore, urea-SCR system is regarded as the most promising approach to meet restricted emission standards [1].

Experimental researches showed that the NO_x concentration tracked at the outlet of SCR catalyst with a transient variation of Adblue dosage had certain regularity (Fig. 1). After a sudden increase of Adblue dosage, the NO_x concentration decreased at 3 seconds later, which means the time delay is encountered in SCR systems. Then, the NO_x concentration decreased slowly and it became stable gradually in 80 seconds (t_1). When the injection stopped, the NO_x concentration increased to the original level in 70 seconds (t_2).

The track of NO_x concentration features as a resistance-capacitance (RC) model, facing obstacles to obtain simultaneously NO_x expected to be reduced and equi-molar ammonia

available to SCR reaction, which finally inhibits NO_x conversion efficiency. Generally, engine loads change frequently, triggering a rapid changing of Adblue dosage, which aggravates the air quality that are caused by NO_x emission and ammonia slip.

The present research confirmed the problem which is discussed above. In order to increase the conversion efficiency of NO_x and avoid secondary pollution, a lot of work have been done [2-5]. Bonfils et al. [2] proposed a lumping method for the SCR system. Zhang et al. [5] presented a method to combine neural network model with Fuzzy Fuzzy proportional-integral-derivative (PID) to meet both NO_x emission requirements and NH_3 slip targets. However, the studies of embeddable models are relatively limited [6]. The existing embeddable models are not accurate enough to fully describe the transient response of NO_x concentration and reveal the regularity of different factors affecting SCR system characteristics [7, 8].

Based on the weight analysis of the influencing factors on SCR system, the paper presents "variable RC model with time delay" to describe the input-output state relations of SCR system comprehensively. Such model can be used for a real-time correction of Adblue dosage, thus to creating an avenue for the SCR control strategy.

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Received June 15, 2014 Accepted April 14, 2015

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Fig. 1. Outlet mole fractions of NO_x during a transient change of Adblue dosage when the engine operated in steady state.

2. Materials and Methods

Both experimental and simulation research on the time-delay and RC characteristics of SCR system were implemented in the paper, and finally the “variable RC model with time delay” was obtained.

The experimental setup is consisted of a 4.75 L diesel engine, a 12 L $\text{V}_2\text{O}_5\text{-WO}_3/\text{TiO}_2$ catalyst, an Adblue dosing system and a Tianlan designed controller. The aftertreatment system was equipped with NO_x sensors and exhaust temperature sensors at both inlet and outlet of the catalyst converter, an AVL AMA i60 emission analyzer and a Siemens ammonia analyzer at the tailpipe.

The process of urea-SCR system can be divided into atomization and decomposition process and catalytic reaction process. The paper established a 3D Adblue injection model to describe the first process, including the evaporation of droplets, secondary break-up, droplet-wall interaction and urea decomposition. A simplified 1D Adblue injection model based on the results of 3D model was also established to speed up the simulation. The urea-SCR model for the weight analysis was consisted of 1D Adblue injection model and 1D catalytic converter model (Fig. 2). After the estimation of kinetic parameters, the simulation results well agreed with the experiments.

Fig. 2. The structure of Urea-SCR (selective catalytic reduction) model.

3. Results and Discussion

3.1. Time Delay

3.1.1. Time delay of Adblue transportation

Due to the time delay of Adblue transportation, the change of NO_x concentration track at the inlet of SCR catalyst is obviously lagging behind the change of Adblue dosage. The state-of-the-art NO_x sensors have cross-sensitivity to NH_3 [9, 10], which can be used to measure the transportation delay (Fig. 3).

The time delay of Adblue transportation is consisted of the lag time in injection pipe and exhaust pipe. The transportation delay of Adblue in injection pipe is related to the injection pressure, dosage and temperature of adblue, and also the geometrical size of injection pipe. The transportation delay of Adblue in exhaust pipe caused by the atomization and decomposition process is influenced by exhaust mass flux and temperature, Adblue dosage and injection pressure, as well as the geometrical size of exhaust pipe. According to the simulation results of 3D Adblue injection model, the transportation delay of Adblue in exhaust pipe is 0.015 seconds (Fig. 4).

Fig. 3. The value of inlet NO_x sensor during a transient change of Adblue dosage when the engine operated in steady state.

Fig. 4. The life time of droplets.

3.1.2. Time delay of exhaust flow and catalytic reaction

The value of NO_x sensors at inlet and outlet of catalyst changes respectively corresponding to the change of Adblue dosage (Fig. 5). The lag time between two NO_x sensors is consisted of the exhaust flow delay and reaction delay.

Fig. 5. The value of NO_x sensor at inlet and outlet during a transient change of Adblue dosage when the engine operated in steady state.

Fig. 6. Calculation of time delay in the catalyst.

The exhaust flow delay is related to exhaust flow, the temperature, and the structure of catalytic converter. In catalyst, NH₃ is first absorbed at active site and then reacts with gas-phase NO_x to form N₂ and H₂O. The lag time of this process is influenced by exhaust flow and temperature, the gas composition and the size of catalyst. According to the simulation results of 1D catalytic converter model, the reaction delay in catalyst is 0.014s (Fig. 6).

3.1.3. Time-delay of different operations

It can be concluded that the transportation delay of Adblue in the injection pipe and exhaust flow delay are the main compositions of time delay of urea-SCR system. Since the physical structure of injection pipe and catalytic converter are determined, the time delay of the system is mainly associated with exhaust flow and Adblue dosage. A map of time delay at different operations can be derived from various kind of experiments (Fig. 7).

Fig. 7. The time delay of urea-SCR system.

3.2. RC Characteristics

3.2.1. The weights of influencing factors

The influencing factors of urea-SCR system discussed in this paper are consisted of four parts. The first is the properties of exhaust, which includes the gas composition, mass flux and temperature. The second refers to the relevant parameters of Adblue injection system, namely the injection quantity, Adblue temperature and the HCNO conversion efficiency. The third means the structure parameters of catalyst and the last one is the kinetic parameters of catalytic rate expressions.

The levels of influencing factors of exhaust properties were determined by the European steady Cycle (ESC) and European Transient Cycle (ETC) baseline tests (ESC baseline: 10.74 g/kW·h, ETC baseline: 10.38 g/kW·h). At the same time, the levels of factors in the second part were determined [11]. Furthermore, the levels of factors in the third part were determined on common commercial catalysts which have been widely used in the world [12]. What is more, the levels of kinetic parameters were determined on the estimation results at different exhaust temperature.

The weights of influencing factors on the NO_x conversion efficiency, time t_1 and t_2 are shown in the Table 1.

3.2.2. The key factors of urea-SCR system

The mean value of the weights was taken as an evaluation index in the paper. All the factors were evaluated and preliminary screened. A weight analysis of the screened factors was carried out and finally the critical elements were found out. The key factors are shown in the Table 2 and Table 3.

As shown in Table 2 and Table 3, the mass flux, temperature and NO_x concentration of exhaust, the catalyst volume, the activation energy of NH₃ ad/desorption and standard SCR reactions, and the Adblue dosage are the key factors of urea-SCR system.

The engine-out NO_x level directly influences the development of urea-SCR system. The space velocity and activity of catalyst are affected by the exhaust mass flux and exhaust temperature. The NO_x mass flux at the inlet is determined by the NO_x concen

Table 1. The weights of Influencing Factors

Factors	Weights		
	NO _x conversion efficiency	Time t_1	Time t_2
Adblue dosage	0.0883	0.0435	0.0195
Adblue temperature	0.0302	0.0091	0.0089
Catalyst density	0.0195	0.0200	0.0162
Catalyst volume	0.0358	0.0620	0.0401
Coverage dependency	0.0120	0.0151	0.0172
CPSI	0.0230	0.0120	0.0227
Critical surface coverage of fast SCR (CSC_fast SCR)	0.0127	0.0062	0.0359
CSC_slow SCR	0.0423	0.0203	0.0258
CSC_standard SCR	0.0150	0.0187	0.0116
Activation energy of fast SCR (E_fast SCR)	0.0336	0.0287	0.0196
E_HCNO Hydrolysis	0.0112	0.0141	0.0209
E_NH ₃ adsorption	0.0141	0.0427	0.0384
E_NH ₃ desorption	0.0386	0.0583	0.0412
E_NH ₃ oxidation	0.0120	0.0316	0.0373
E_slow SCR	0.0201	0.0315	0.0377
E_standard SCR	0.0661	0.0176	0.0247
Exhaust mass flux	0.0883	0.0512	0.0848
Exhaust temperature	0.0701	0.0448	0.0513
HCNO conversion efficiency	0.0292	0.0207	0.0130
Frequency factor of fast SCR (K_fast SCR)	0.0100	0.0182	0.0158
K_HCNO hydrolysis	0.0199	0.0302	0.0212
K_NH ₃ adsorption	0.0114	0.0183	0.0195
K_NH ₃ desorption	0.0199	0.0557	0.0110
K_NH ₃ oxidation	0.0108	0.0229	0.0307
K_slow SCR	0.0065	0.0281	0.0328
K_standard SCR	0.0146	0.0086	0.0164
Length of catalyst	0.0139	0.0198	0.0167
Max surface coverage fraction	0.0170	0.0086	0.0138
Density of active sites	0.0515	0.0534	0.0450
NO ₂ proportion	0.0126	0.0098	0.0267
NO _x concentration of exhaust	0.0442	0.0550	0.0674
Outlet pressure	0.0314	0.0174	0.0210
Specific heat of catalyst	0.0209	0.0420	0.0251
Thermal conductivity of catalyst	0.0163	0.0167	0.0290
Wall thickness	0.0083	0.0142	0.0139
Washcoat thickness	0.0284	0.0333	0.0270

CPSI: channels per square inch (in^{-2}), CSC: critical surface coverage, E: activation energy ($\text{J}\cdot\text{mol}^{-1}$), K: frequency factors ($\text{m}^4\cdot\text{mol}^{-1}\cdot\text{s}$; $\text{m}^3\cdot\text{mol}^{-1}\cdot\text{s}$; $\text{m}\cdot\text{s}^{-1}$), SCR: selective catalytic reduction.

Table 2. Key Factors of NO_x Transformation Efficiency

Rank	Factor	Weight
1	Exhaust mass flux	0.2830
2	Adblue dosage	0.1455
3	Exhaust temperature	0.1160
4	E_standard SCR	0.0876
5	NO _x concentration of exhaust	0.0805

E: activation energy ($\text{J}\cdot\text{mol}^{-1}$), SCR: selective catalytic reduction.

tration and exhaust mass flux. Therefore, the mass flux, temperature and NO_x concentration of exhaust play a very vital role on the NO_x conversion efficiency, time t_1 and t_2 .

The catalytic performance of catalyst is crucial to the urea-SCR system. The standard SCR reaction means a lot in the NO_x reduction, so its activation energy has an obvious effect on the NO_x conversion efficiency. Inside the catalyst, NH₃ takes part in the SCR reaction only after the adsorption on the active sites, which refers to the reaction of NH₃ ad/desorption, the density of active sites and the

Table 3. Key Factors of Time t_1 and t_2

Time t_1			Time t_2		
Rank	Factors	Weights	Rank	Factors	Weights
1	Catalyst volume	0.1347	1	NO _x concentration of exhaust	0.1491
2	E_NH ₃ desorption	0.1104	2	Exhaust temperature	0.1371
3	Density of active sites	0.0810	3	Exhaust mass flux	0.0951
4	NO _x concentration of exhaust	0.0774	4	E_NH ₃ desorption	0.0811
5	Exhaust mass flux	0.0758	5	Catalyst volume	0.0793
6	Exhaust temperature	0.0708			
7	Adblue dosage	0.0658	6	Density of active sites	0.0779
8	E_NH ₃ adsorption	0.0651			

E: activation energy (J·mol⁻¹).

catalyst volume have a great influence on the transient response of urea-SCR system.

The Adblue dosage which determines the concentration of NH₃ and HCNO at the inlet has a great effect on the NO_x conversion efficiency and time t_1 but little effect on time t_2 .

3.3. The Description of Urea-SCR System

Based on the comprehensive analysis of urea-SCR system, the paper presents “variable RC model with time delay” to describe the characteristic of the system. The transfer function is calculated by Eq. (1):

$$w(s) = \frac{T_{ratio}}{RCs + 1} \cdot e^{-\tau s} = \frac{T_{ratio}}{T_{RC}s + 1} \cdot e^{-\tau s} \quad (1)$$

Where, R is the value of resistance, C is the value of capacitance, T_{RC} is the time constant, T_{ratio} is the proportional constant, and τ is the time delay.

The parameter τ is derived from the interpolation of time delay map that is shown in Fig. 7. According to the results of weight analysis, the time constant and proportional constant are calculated by Eq. (2):

$$T = \prod_i^N (A_i \cdot x_i^2 + B_i \cdot x_i + C_i) \quad (2)$$

Where, N is the number of key factors, x is the weight of key factors, A, B and C are state parameters.

As shown in Fig. 8, different T_{RC} is used according to the trends

Fig. 8. The calculation process of T_{RC} and T_{ratio} .

Fig. 9. Comparison between experiment and simulation results after the estimation.

of outlet NO_x concentration.

After the estimation of state parameters by using genetic algorithm, the simulation results well agreed with the experiment (Fig. 9) and the error less than 0.5%. The control strategies of Adblue dosage developed based on this model will have a better performance in keeping high NO_x conversion efficiency and shortening the time t_1 and t_2 , which can help to reduce NO_x emission and avoid NH_3 slip.

3.4. Model-based Control of Urea-SCR System

A model-based control strategy of urea-SCR system is presented in the paper (Fig. 10). The prediction of engine-out NO_x is based on the NO_x map that is derived from the universal performance curves. These steady-state values are corrected according to the

change rate of speed and torque. What's more, the maximal NO_x conversion efficiency (the NH_3 slip is keeping at 10 ppm) at different SV and exhaust temperature are taken into consideration in the calculation of basic Adblue dosage. Also, the Adblue dosage must be modified during the sudden changes of engine operating conditions. The variable RC model is used for the prediction of system-out NO_x . According to the NO_x target and prediction, the fuzzy controller gives the emendation of Adblue dosage. At last, a correction is made by the rapid increasing controller for preventing a greatly increase of Adblue dosage.

The model-based control strategy was verified by ETC and ESC test and the results were lower than Euro V emission standard limit $2.0 \text{ g/kw}\cdot\text{h}$ (Fig. 11). Furthermore, as the key part in the SCR control, this model can be easily used in other strategies.

Fig. 10. Block scheme of model-based control strategy.

Fig. 11. (a) ESC and (b) ETC test results (The NO_x specific emission of ESC and ETC were $1.64 \text{ g/kW}\cdot\text{h}$ and $1.62 \text{ g/kW}\cdot\text{h}$. The peak NH_3 slip of ESC and ETC were 2.7×10^{-6} and 7.5×10^{-6}). ESC: European steady cycle, ETC: European transient cycle.

4. Conclusions

The paper gives a comprehensive analysis of time delay of urea-SCR system, and the results show that the transportation delay of Adblue in injection pipe and exhaust flow delay are the main composition. A weight analysis of influencing factors is carried out and the result shows that the mass flux, temperature and NO_x concentration of exhaust, the catalyst volume, the activation energy of NH_3 ad/desorption and standard SCR reactions, and the Adblue dosage are the key factors of urea-SCR system.

Accordingly, a map of time delay is established and a solution method for time constant and proportional constant is carried out. Finally, the paper presents “variable RC model with time delay” to describe the input-output state relation of SCR system accurately and to reveal the regularity of different factors affecting SCR system characteristics. Moreover, this model can be used for a real-time correction of Adblue dosage, which can increase the conversion efficiency of NO_x and avoid secondary pollution forming.

Acknowledgments

The research was supported by the National Natural Science Foundation of China (No. 51379165) and the Natural Science Foundation of Educational Department of Hubei government (No. 20520005). And the author appreciates the support of Key Laboratory of High Performance Ship Technology of Ministry of Education (Wuhan University of Technology).

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