Dynamic Characteristics of a Urea SCR System for NOx Reduction in Diesel Engine

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Abstract: This paper discusses dynamic characteristics of a urea-SCR (Selective Catalytic Reduction) system. The urea flow rate to improve NOx conversion efficiency is generally determined by parameters such as catalyst temperature and space velocity. The urea-SCR system was tested under the various engine operating conditions governing the raw NOx emission levels, space velocity, and SCR catalyst temperature. These experiments include cold-transients to determine catalyst light-off temperature and urea flow rate transients. Likewise, ammonia storage dynamics was also investigated.

The cold-transient results indicate the light-off temperature of the catalysts used in these experiments was 200-220°C. The ammonia storage and urea flow rate transients all indicate very slow dynamics (on the order of seconds), which presents control challenges for mobile applications. The results presented in this paper should provide an excellent starting point in developing a functional in-vehicle urea-SCR system.

Key words: Dynamic characteristics, Space velocity, Cold transients, Flow rate transients, Ammonia storage dynamics

1. Introduction

Diesel engines offer significant advantages over spark ignited engines in terms of peak torque production, carbon monoxide (CO) emissions, hydrocarbon (HC) emissions, carbon dioxide (CO2) emissions (known to cause the greenhouse effect) and fuel consumption. However, lean exhaust conditions render conventional automotive three way catalysts ineffective, making NOx reduction a considerable challenge. Urea SCR is a technology that has received much attention in recent years address this problem, and has shown the potential to meet the stringent regulations for NOx emissions for US 2007/2010 and Euro IV/V.(11,12)

The functionality of urea-SCR catalyst systems is defined by the selective reduction of NOx in a lean exhaust environment using ammonia, which is generated from ammonia. This makes urea-SCR well suited to use with diesel engine.
engines, which always operate significantly lean of stoichiometry. Four catalysts comprise a typical urea-SCR system, including pre-oxidation, hydrolysis, SCR, and post-oxidation catalysts\(^{(3),(4)}\).

2. Apparatus and Method

2.1 Apparatus

The research engine used for these experiments was a 103 kW turbocharged, intercooled, 2.5L VM-Motori compression-ignition, direct-injection (CIDI) engine equipped with a cooled exhaust gas recirculation (EGR) system. The engine is calibrated to meet the Euro III emissions certification level, and is representative of a modern passenger car diesel engine.

![Schematic diagram of experimental measuring apparatus](image)

A. MAF Sensor, B. VM 2.5L CIDI Engine, C. Oxidation Catalyst, D. Hydrolysis & SCR Catalyst, E. Horiba MEXA-7500 (e1: NOx, THC, CO, CO\(_2\), O\(_2\), e2: NOx, THC), F. Urea Syringe Pump, G. Air Regulator, H. Compressed Air Tank, P. Pressure Sensor, t1-t4 : Temperature Sensors

Emissions measurements were performed using a Horiba MEXA-7500 exhaust gas analyzer, with NO\(_x\) measurements available on two separate lines (for raw and post-catalyst measurements).

Figure 1 shows a schematic diagram of experimental measuring apparatus.

2.2 Calculation Method

The urea solution was created using reagent-grade urea pellets and distilled water. The urea solution concentration selected for this work was 33\% by weight, which is the eutectic solution.

The dynamic characterization of the urea-SCR system, specifically the NO\(_x\) conversion capability of the catalyst, consisted of sweeping through multiple urea flow rates at fixed engine operating points. A total of 29 engine operating points were selected, ranging from 30 to 150 ft-lb at intervals of 30 ft-lb and 1250 rpm to 2500 rpm at intervals of 250 rpm, based on common operating points for the 2.5L VM-Motori diesel engine over urban and highway driving schedules in a mid-size sport utility vehicle. The urea flow rates were selected based on the stoichiometric urea flow rate, given by Equation 1:

\[
m_{\text{urea, stoich}} = \frac{(m_{\text{NO}}/FW_{\text{NO}})(FW_{\text{NH}_3})}{(1/(U:NH_3))(1/\text{Urea})} \tag{1}
\]

where

\(m_{\text{NO}}\) : the raw NO flow rate in [g/s]  
\(FW_{\text{NO}}\) : the formula weight of NO in [g/mol]  
\(FW_{\text{NH}_3}\) : the formula weight of NH\(_3\) in [g/mol]  
\(U:NH_3\) : the NH\(_3\) produced from a unit mass of urea [%]  
\text{Urea} : the concentration of the urea solution [%]
which is calculated as a function of raw NOx emissions from the engine and the chemical and physical properties of urea, NOx, and ammonia.

Space velocity is a key consideration in catalyst design, and it has a significant impact on the NOx conversion efficiency of urea-SCR system. Space velocity is defined as the inverse of space time, which is the time an exhaust gas equivalent to one catalyst volume takes to move through the catalyst. The significance of space velocity is that it defines the time period that the exhaust gases are in contact with the catalyst.

As a design consideration, a balance must be reached between time for catalytic activity and excessive by-product formation and/or heat transfer. Space velocity is defined mathematically in Equation 2.

$$\text{SV} = \left(\frac{m_{\text{exhaust}}}{\rho_{\text{exhaust}}}\right)(1/V_{\text{cat}}) \tag{2}$$

where

- $\text{SV}$: space velocity in [1/hr]
- $m_{\text{exhaust}}$: the exhaust mass flow rate in [g/hr]
- $\rho_{\text{exhaust}}$: the exhaust gas density in [g/L]
- $V_{\text{cat}}$: the catalyst volume in [L]

A map of NOx conversion efficiency as a function of space velocity and catalyst temperature was generated based on the static urea mapping results presented previously, and it is displayed in Figure 2.

The figure shows the trends in NOx conversion efficiency for varying catalyst temperature and space velocity. As expected, NOx conversion efficiency increases greatly with increasing catalyst temperature.

Fig. 2 NOx conversion efficiency map as a function of space velocity and catalyst temperature

The other effects of varying space velocity is also displayed, and NOx conversion efficiency is adversely affected by increasing space velocity above the threshold of 40,000-50,000 1/hr. This was anticipated because of the decreased time available for the catalytic reduction of NOx over the catalyst as space velocity increased.

3. Experiment Methods and Results

3.1 Transient urea flow rate

As engine operating points change rapidly under normal driving conditions for mobile applications, rapid changes in urea flow rate are also required to maintain high NOx conversion efficiency and minimize ammonia slip. As such, the urea flow rate dynamics were investigated by applying step changes in urea flow rate at a fixed engine operating point. The eight engine operating points considered represented varying NOx emissions, space velocity, and catalyst
temperature as outlined in the previous section. Step changes in urea were applied in two fashions: a step from the stoichiometric urea flow rate to the maximum urea flow rate at the operating point as established in the static testing and vice versa.

The results are presented in the form of NO\textsubscript{x} conversion efficiency on the main y-axis and urea flow rate of the secondary y-axis. Common trend in results are presented for the urea flow rate transient experiments on Figure 3 through Figure 6. The results presented in the figures generally indicate slow dynamics for the urea flow rate transients, regardless of operating point or urea step direction. However, there are some variations in the results to note.

Figure 3 represents a case where a very long delay is encountered after the urea flow step increase before the NO\textsubscript{x} conversion efficiency returns to steady-state. This is likely due to the low catalyst temperature and NO\textsubscript{x} flow rate at this operating point (1250 rpm, 30 ft-lb), as well as the poor steady-state NO\textsubscript{x} conversion efficiency. Figure 4 represents a case where a shorter delay is realized in returning the steady-state NO\textsubscript{x} conversion upon the step increase in urea flow rate. This can be attributed to the high NO\textsubscript{x} flow rate, catalyst temperature, and steady-state conversion efficiency of the operating point (2250 rpm, 150 ft-lb), which are all beneficial effects for faster transients in this case. Figure 5 represents a long delay time for a case of a step change from high to low urea flow. This can be explained by the low NO\textsubscript{x} flow rate at the operating point (1750 rpm, 90 ft-lb), and the associated time to consume the stored ammonia more than the stoichiometric urea flow rate (\(\alpha > 1\)) used initially. Finally, Figure 6 is a typical short delay time for a step change from high to low urea flow. The high catalyst temperature and NO\textsubscript{x} flow rate for this operating point (2250 rpm, 150 ft-lb) maximize NO\textsubscript{x} conversion efficiency and minimize the time to consume stored ammonia from the initially high urea flow rate, reducing the delay time.

![Fig. 3 Transient urea flow rate at 1250 rpm, 30 ft lb (Stoichiometric to high flow)](image)

![Fig. 4 Transient urea flow rate at 2250 rpm, 150 ft (Stoichiometric to high flow)](image)
conversion efficiency while limiting ammonia slip. For these experiments, a high urea flowrate was selected for each fixed engine operating point, and the catalyst was saturated with ammonia over the course of several minutes. The urea flowrate was then zeroed, and the response of the NO$_x$ conversion efficiency was recorded. Results are presented in Figures 7 through 9.

The figures are the set of data collected for the urea-SCR ammonia storage dynamics experiments. Figure 7 represents a case where a delay time is encountered to consume the available ammonia. This is largely due to the relatively low NO$_x$ flow rate at the operating point (2500 rpm, 60 ft-lb), despite the high steady-state NO$_x$ conversion efficiency. Figure 8 is the case where the delay time is longer to consume the stored ammonia. At this operating point (2000 rpm, 120 ft-lb), the NO$_x$ flow rate is fairly high, but the steady-state NO$_x$ conversion efficiency at this operating point is lower, which is the likely explanation for the delay time.

3.2 Ammonia storage dynamics

An important control strategy consideration for urea-SCR systems for mobile applications is the phenomena of ammonia storage dynamics. Urea-SCR catalysts have the ability to store unused ammonia from the SCR process for later use. Implicit accounting for ammonia storage dynamics is necessary for mobile applications, where transient operation occurs frequently, to maintain high NO$_x$ storage capacity.
from an engine occur after the engine (and exhaust system) have been cold-soaked (remained dormant for several hours). This occurs for two reasons: higher raw emissions during the first minutes of operation (primarily for spark-ignited engines) and insufficient exhaust and catalyst temperature for catalytic activity. Many emissions certification driving schedules feature a cold-soak portion, so several cold transient experiments have been performed using the urea-SCR system to better understand factors such as light-off time and temperature and the impact of the engine operating point on these factors. The cold transient experiments were performed at 8 engine operating points of varying NOx emissions. SCR catalyst space velocity, and equilibrium(warm) catalyst temperature. The experimental procedure consisted of soaking (cooling) the engine until the SCR catalyst and engine coolant reached room temperature, then activating the engine and urea dosing system at the given operating points and recording the NOx response. Three cases are presented here: a case where catalyst light-off, defined by 50% NOx conversion efficiency, is never reached during the time data is recorded, a case where catalyst light-off occurs after a relatively lengthy time period, and a case where catalyst light-off occurs in a relatively short period of time. These results are presented in Figure 10 through Figure 12. In addition, the results, including light-off time and temperature, for the cold transient experiments are summarized in Table 1.

3.3 Cold transients

A significant source of the emissions
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![Fig. 10 Urea SCR cold transient at 1250rpm, 30 ft lb](image)

![Fig. 11 Urea SCR cold transient at 2500 rpm, 90 ft lb](image)

**Table 1 Cold transient light-off results**

<table>
<thead>
<tr>
<th>Operating Point</th>
<th>50% Light-Off Time (sec)</th>
<th>50% Light-Off Temperature (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1250rpm, 30 ft-lb</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>1250rpm, 120 ft-lb</td>
<td>82.3</td>
<td>218.3</td>
</tr>
<tr>
<td>1750rpm, 90 ft-lb</td>
<td>59.1</td>
<td>220.5</td>
</tr>
<tr>
<td>2000rpm, 90 ft-lb</td>
<td>80.0</td>
<td>215.5</td>
</tr>
<tr>
<td>2000rpm, 120 ft-lb</td>
<td>44.3</td>
<td>194.1</td>
</tr>
<tr>
<td>2250rpm, 150 ft-lb</td>
<td>46.4</td>
<td>234.4</td>
</tr>
<tr>
<td>2500rpm, 60 ft-lb</td>
<td>85.5</td>
<td>226.1</td>
</tr>
<tr>
<td>2500rpm, 90 ft-lb</td>
<td>62.1</td>
<td>227.9</td>
</tr>
</tbody>
</table>

Several trends can be extracted for the figures and table presented above. The light-off temperature of the catalyst is approximately 220°C, and is fairly insensitive to the engine operating conditions, NOx flow rate, etc. The light-off time, though, is very sensitive to engine operating conditions. Figure 10 is represents low-speed, low-load operating points. Due to the low NOx emissions and low equilibrium temperature (warm) of the catalyst at this operating point (1250 rpm, 30 ft-lb), there is a very long associated light-off time. In fact, light-off was not reached for this operating point in the 10 minutes of data collected.

Figure 11 is the mid-load operating points featuring medium catalyst temperature and NOx flow rate. The catalyst reaches light-off at this operating point (2500 rpm, 90 ft-lb) in the time data was recorded, but only after a somewhat lengthy period of time. The main factor limiting higher NOx conversion in a shorter time period is the relatively low (compared to high-load operating points) equilibrium (warm) catalyst temperature.

![Fig. 12 Urea SCR cold transient at 2250 rpm, 150 ft lb](image)
Figure 12 is the high-load engine operating conditions. The SCR catalyst reaches light-off quickly for this operating point (2250 rpm, 150 ft-lb) due to the more immediate increase in exhaust temperature encountered from the high-load operation and the subsequent faster increase in catalyst temperature. Regardless of operating point, the SCR catalyst generally offers no NO\textsubscript{x} conversion for the first 10-20 seconds after engine activation.

4. Conclusion

The NO\textsubscript{x} conversion efficiency of the catalysts depends sensitively on the engine operating condition, as well as the space velocity and catalyst temperature. NO\textsubscript{x} concentration varied slowly with the ammonia storage and urea flow rate transients (on the order of seconds), which suggest the possibility for mobile applications. The raw NO\textsubscript{x} flow rate and catalyst temperature greatly influenced the response time. The cold-transient results show the light-off temperature of the catalysts used in these experiments was 200-220°C and the light-off time was greatly influenced by engine operating conditions.

References


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He received the B.S., M.S., and Ph. D. degrees from Korea Maritime University in 1985, 1991 and 1999, respectively. He joined and worked at Research & Development Division for Hyundai Motor Company from Feb. 1992 to Feb. 2000. He was an Academic visitor for CAR (Center for Automotive Research) of Ohio State University USA in 2004. He is currently an associate professor at Mokpo Maritime University, Division of Marine Engineering in Mokpo, South Korea.

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