Abstract: A marine Diesel engine is available to low-quality heavy oil, and also has the advantage of high efficiency. However, NO\textsubscript{x} emission of the marine Diesel engine is greater than the other internal combustion engines on the ground, such as to use automobiles and electric power plants. The NO\textsubscript{x} emission causes acid rain and photochemical smog, and it is influence directly to human health, such as lack of oxygen or respiratory disease. Especially, to keep environment protection in a harbor area, we must reduce the NO\textsubscript{x} emission urgently.

We have started to study on a SCR (Selective Catalytic Reduction) system for a 4-stroke medium speed marine Diesel engine since 2007. The SCR is a reducing technology of nitrogen oxide, NO\textsubscript{x}. A general SCR system consists of a catalyst made of titanium-vanadium and an injection nozzle to jet mist of urea water as a reducing agent. When the temperature of the exhaust gas is kept enough high, the urea is converted to ammonia, and NO\textsubscript{x} in the exhaust gas is converted to nitrogen and water by the catalysis. Also as the reducing agent, ammonia gas or ammonia water is able to use for the catalysis. In order to apply the SCR system to the marine application, it is necessary to estimate a basic performance of the SCR and to develop a control system of the reducing agent. In this paper, we show test results of several experimental studies in our project.

One of our experimental studies, to estimate the basic performance of the SCR, we have carried out several catalyst only tests without a Diesel engine. The test results are effective to design and develop a marine SCR system.

As the next step, we have constructed an experimental SCR system in our laboratory as shown in Figure 1. The system has a marine Diesel engine, and we have examined the NO\textsubscript{x} reduction rate at each load and the effects of a kind of the reducing agent, which are ammonia gas and urea water. As the results, it is confirmed that the SCR system has suitable NO\textsubscript{x} reduction performance at each load as shown in Figure 2. It is also clarified that there is no deference by the kind of reducing agent in enough high temperature of the exhaust gas.

On the other hand, we have investigated control methods with the experimental SCR system. In the control system, the reducing agent is controlled by a calculated exhaust gas flow rate and a measured NO\textsubscript{x} concentration. It is confirmed that the control system has suitable performance in our early tests.

Based on the above test results, we have designed and developed a SCR system for a marine Diesel generator on a ship. The SCR system is installed to the ship and examined on board at sea. In the actual ship, there is not enough wide space for the SCR. Therefore the distance between the injection nozzle and the catalyst of the SCR system must be short, though it is needed a long distance for the conversion to ammonia from urea generally. We developed a special injection nozzle for the system and achieved suitable NO\textsubscript{x} reduction performance as shown in Figure 3.

In conclusion, we got a lot of beneficial results to apply a SCR system to a middle-speed marine Diesel engine. In the next step, in order to develop a practical SCR system, it is necessary to develop a simple and low-cost control system and to estimate a durability performance of catalyst. Also, in order to apply the SCR system to large two-stroke Diesel engine, we need to examine the SCR system performance in detail, because the engine has too low temperature of exhaust gas.
INTRODUCTION

One of advantages of a marine Diesel engine is that the engines can use low-quality heavy oil with high efficiency. However, one of disadvantages is that NOx emission of the marine Diesel engine is greater than the other internal combustion engines on the ground, such as automobiles and electric power plants. The NOx emission causes acid rain and photochemical smog, and it influences directly to human health, such as respiratory disease. In particular, to protect air environment quality in harbor areas, we must reduce the NOx emission. Furthermore, Annex VI of MARPOL 73/78 regulations which regulates by the IMO (International Maritime Organization) took effect in May 2005. Severe requirements of NOx reduction such as Tier II and Tier III will be enforced in 2011, 2016. In the Tier III regulations, The NOx limit will be reduced 80% from current limit. Therefore, we have started to study on a SCR (Selective Catalytic Reduction) system for fore-stroke medium speed marine Diesel engine since 2007 in order to develop NOx reduction technologies [1], [2], [3], [4].

In this paper, we report the test results of several experimental studies in our project. Several experimental studies include catalyst component tests to estimate basic performance of SCR, test with the SCR system for the marine Diesel engine in our laboratory and on board test at sea with designed and developed the SCR for marine Diesel generator on a ship.

OUTLINE OF SELECTIVE CATALYTIC REDUCTION

A general SCR system

A SCR is one of the most prospective reducing technologies for NOx. It has been already practically used in a field of automobiles, such as buses and trucks. NOx in exhaust gas reacts with NH3 to N2 and H2O on a catalyst as follows:

\[
4\text{NO} + 4\text{NH}_3 + \text{O}_2 \rightarrow 4\text{N}_2 + 6\text{H}_2\text{O} \quad (1)
\]

\[
2\text{NO}_2 + 4\text{NH}_3 + \text{O}_2 \rightarrow 3\text{N}_2 + 6\text{H}_2\text{O} \quad (2)
\]

The main reaction is Equation 1 because almost of NOx in exhaust gas of fore-stroke medium speed Diesel engines is NO. NH3 as a reducing agent is injected into exhaust gas. A reducing agent is ammonia gas or ammonia solution or urea solution. In many cases a reducing agent uses urea solution.

A general SCR system consists of a catalyst and an injection nozzle to jet mist of urea solution as shown in Figure 1.

Catalyst used SCR system

In this study, the catalyst (TiO2 and V2O5 based), which has been already practical used in a field, is used. Exhaust gas of marine Diesel engine contains SOx from sulfur component in fuel. Therefore, a catalyst will be deteriorated and NOx removal performance will be decreased. The used catalyst has resistance to deteriorate by SOx and can activate the performance at low exhaust gas temperature.

Injection nozzle

A SCR system has an injection nozzle before catalyst. An injection nozzle is to inject and form a jet mist of a reducing agent in exhaust gas. It is important for forming a jet mist. To achieve it, two-fluid type injector is used in this study. The two-fluid type injector forms a jet mist of a reducing agent by compressed air.

Controller

In addition, it is necessary to control amount of injected reducing agent, because amount of NOx emission will change with an operation state of engine. If not optimize amount of a reducing agent injected, NOx will not react or excessive ammonia gas will release from a SCR system, ammonia slip.

Type of a reducing agent

NOx reduction achieves by reacting with NH3. NH3 is produced ammonia gas, ammonia solution or urea solution. Ammonia solution easily produces a NH3 by evaporating. But ammonia solution is difficult to handle to causticity nature and low boiling temperature.
On the other hand, urea solution converts to ammonia when temperature of the exhaust gas is kept high enough as follow:

$$\text{NH}_2\text{CO-NH}_2 \rightarrow \text{NH}_3 + \text{HNCO} \quad (3)$$

$$\text{HNCO} + \text{H}_2\text{O} \rightarrow \text{NH}_3 + \text{CO}_2 \quad (4)$$

After evaporation of urea solution, urea thermally decomposes into ammonia and isocyanic acid (HNCO) as Equation 3. Then isocyanic hydrolyzes ammonia and carbon dioxide. As a result 1mol of urea produces 2mol ammonia and 1mol of carbon dioxide. Urea solution is easy to handle unlike ammonia solution. In this study, urea solution is used as a reducing agent. But urea may form a crystal and block up an injector and pipe. Therefore, in on board test at sea, we prepared three injection nozzles.

**Performance of NOx removal**

In this study, NOx removal performance describes as NOx conversion rate as follows:

$$\text{NOx conversion rate} \,(\%) = \left( 1 - \frac{\text{NOx concentration after the catalyst [ppm]}}{\text{NOx concentration before the catalyst with injection air [ppm]}} \right) \times 100 \quad (5)$$

Strictly, since water in exhaust gas will be increased by injected urea solution as a reducing agent. NOx concentration decreases. But it is negligible change. Because flow rate of injection urea solution is vanishingly small, compared with that of exhaust gas.

**CATALYST COMPONENT TEST**

**Outline of catalyst component test**

To apply a SCR system for a ship, it is necessary to estimate NOx removal performance. A catalyst data is insufficiency for estimating the performance, as marine Diesel engine exhaust gas have low temperature and different components such as SOx in comparison of field used engines. Therefore, to estimate its performance, we have studied the factor that exhaust components had an influence on NOx removal performance individually with the micro-reactor.

**Experimental apparatus and methods**

The experimental apparatus and methods used in this experiment are discussed below. The schematic diagram of the micro-reactor is shown in Figure 2. The Micro-reactor is able to carry out experiment with simulated exhaust gas. The simulated exhaust gas is composed of prepared gases in cylinders and compressed dry air. Gas components are N₂, CO₂, CO, NO and SO₂, and the flow rates of the prepared gases and compressed air are controlled by mass-flow controllers. The simulated exhaust gas is heated in the evaporator and pre-heater, in which urea solution as a reducing agent is injected and evaporates. Heated simulated exhaust gas mixed with a reducing agent is led to the catalyst set in the electric furnace, where the NOx is converted. The temperature in the electric furnace can be set arbitrary within the range 200-450 deg C.

Urea solution is used as a reducing agent. Urea is changed into ammonia in evaporator and pre-heater, foregoing Equations 3 and 4, and this ammonia reacts with NOx on the surface of the catalyst in the electric furnace, and N₂ and H₂O are produced, foregoing Equations 1 and 2. Urea solution is supplied equivalently to remove NOx. The concentration of H₂O at 10 % consists of the sum of the prepared gases and urea solution.

In this study, components of simulated exhaust gas are fixed as Table 1. The composition of the simulated exhaust gas led into the micro-reactor is confirmed by the exhaust gas analyzer, HORIBA MEXA-1600DEGR.

**Table 1. Components of simulated exhaust gas**

<table>
<thead>
<tr>
<th>NO</th>
<th>CO</th>
<th>CO₂</th>
<th>H₂O</th>
<th>O</th>
<th>N</th>
</tr>
</thead>
<tbody>
<tr>
<td>[ppm]</td>
<td>[ppm]</td>
<td>[%]</td>
<td>[%]</td>
<td>[%]</td>
<td>[-]</td>
</tr>
<tr>
<td>1500</td>
<td>400</td>
<td>10</td>
<td>10</td>
<td>13</td>
<td>Balance</td>
</tr>
</tbody>
</table>

**Figure 2. Schematic diagram of micro-reactor**
Result and discussion of catalyst component test

The experimental results are shown in Figure 3 and 4. We used three types sample A and B each for the experiment. Sample A and B each were separated into three types by temperature. In the Figure 3 and 4, it is clear that the catalyst should be selected to use temperature.

We applied foregoing NOx conversion rate to equation generally used in catalytic chemistry as follows:

\[ \eta = 100 \cdot \left[ 1 - \exp \left( -\frac{Ka}{SV} \cdot Ap \right) \right] \]  \hspace{1cm} (6)

where \( \eta \) [%] is NOx conversion rate. \( Ka \) is the first order reaction constant. \( SV \) [h\(^{-1}\)] is defined as the exhaust gas volume flow rate at normal state divided by the volume of the catalyst. \( Ap \) [m\(^{-1}\)] is defined as total surface area divided by the volume of the catalyst.

From the Equation 6, it is clear that there is \( Ka \) value which makes a correlation \( SV \) value and NOx conversion rate in the condition of constant temperature. Equation 6 is arranged as follows:

\[ -\ln \left( 1 - \frac{\eta}{100} \right) = Ka \cdot \frac{Ap}{SV} \]  \hspace{1cm} (7)

Under Equation 7, NOx conversion rate of sample B is shown in Figure 5. Figure 5 shows the variation in \(-\ln(1-\eta/100)\) as a function of \( Ap/SV \). Therefore, \( Ka \) value is as a gradient in Figure 5. The measurement point is approximated by straight lines. From the Figure 5, it is confirmed that \( Ka \) value is constant not relational of \( SV \) value and it is possible to calculation \( Ka \) value from NOx conversion rate.

Secondly, consider the relation between \( Ka \) value and temperature of a catalyst with Arrhenius equation as follows:

\[ Ka = A \cdot \exp(-B/T) \]  \hspace{1cm} (8)

where \( A \) and \( B \) are constant value. Equation 8 is arranged as follows:

\[ \ln(Ka) = \ln(A) - B/T \]  \hspace{1cm} (9)

Under Equation 9, \( Ka \) value calculated by Equation 7 is shown in Figure 6 and 7. Figure 6 shows that it possible to apply calculated \( Ka \) value to Arrhenius equation. Table 2 shows linear approximate experimental equations of \( Ka \) value.

It follows from foregoing discussion that \( Ka \) value is uniquely decided by temperature of a catalyst. \( Ka \) value decided NOx conversion rate as Equation 6. Therefore NOx removal performance will be calculated by temperature of a catalyst and a catalyst shape. In design a SCR system, \( Ka \) value will become the useful information. We designed the SCR for marine Diesel generator on a ship as will hereinafter be described in detail.
THE SCR SYSTEM FOR ENGINE

Outline of the SCR system for engine

As the next step, we have constructed the experimental SCR system with the marine Diesel engine in our laboratory. In this chapter, we report the result of NOx removal performance at each load and the effects of the reducing agent type which is urea solution (40%) or ammonia gas.

Experimental facility

The experimental SCR system is shown in Figure 8. The catalyst is sample A for low temperature. Exhaust gas components are measured after sampling at upstream and downstream the catalyst by the exhaust gas analyzers, HORIBA MEXA-1600DEGR and HORIBA MEXA-6000FT. SV value of the experimental SCR system is approximately 5000 [h^{-1}]. This experimental facility has a fore-stroke medium speed marine Diesel engine. The engine specification is listed in Table 3. Operation data of the engine and the experimental SCR system such as engine load, temperatures measured and recorded.

Control method of the experimental SCR

Amount of injected reducing agent is controlled by SCR controller and PC in Figure 8.

Controller decides amount of injected reducing agent by measurement NOx concentration and the flow rate of exhaust gas. Used NOx concentration is measured with foregoing the gas analyzer. Generally, flow rate of exhaust gas measurement uses carbon balance method. To calculate flow rate of exhaust gas by carbon balance method, it is necessary to measure accurately several data which is fuel consumption, fuel property and CO₂ and CO concentration of exhaust gas. But it is not
appropriate to measure accurately several data for a ship. Therefore, we used a flow rate of exhaust gas calculated by the engine speed, the charge air pressure and the charge air temperature. Our used method is simple and has accuracy to control amount of an injected reducing agent. Amount of an injected reducing agent is decided from foregoing Equation 1, 3 and 4.

Result and discussion of the SCR system for engine

Figure 9 and 10 show the experimental SCR system for NOx reducing performance with ammonia gas and urea solution. Measurement data are separated by the ship characteristic load percentage. These Figures show that NOx appropriately converted with each reducing agent which is ammonia gas or urea solution in each load. The temperature of exhaust gas in each load are listed the Table 4.

Figure 11 and 12 show the NOx conversion rate as a function of equivalence ratio with ammonia gas and urea solution. The equivalence ratio is defined the ratio of actual urea flow rate and the ideal urea flow rate for 100 % of NOx reduction. In figure 11 and 12, each reducing agent which ammonia gas or urea solution, NOx conversion rate is directly proportional to equivalence ratio.

From these results, it is confirmed that there is no deference by the type of a reducing agent in enough high temperature of the exhaust gas and the SCR system has good performance. On the other hand, experiments carried out in variable temperature of a catalyst in the micro-reactor, in the next step, to compare NOx removal performance by the Diesel engine with that by micro-reactor, we will carry out the experiment of the reducing temperature of exhaust gas and calculate Ka value.

Table 4. Temperature of exhaust gas

<table>
<thead>
<tr>
<th>Load [%]</th>
<th>25</th>
<th>50</th>
<th>75</th>
<th>100</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temp. [deg C]</td>
<td>350</td>
<td>400</td>
<td>390</td>
<td>400</td>
</tr>
</tbody>
</table>

Figures:

Figure 9. The experimental SCR system NOx reducing performance with ammonia gas

Figure 10. The experimental SCR system NOx reducing performance with urea solution

Figure 11. The experimental SCR system NOx reducing performance with ammonia gas

Figure 12. The experimental SCR system NOx reducing performance with urea solution
DEVELOPMENT OF SCR SYSTEM FOR FIELD TEST

Outline of development the SCR for field test

In order to confirm the technical problems for NOx reduction and to find their solutions for developments of practical technology in the marine application, we have developed a SCR system for field tests on a ship. The SCR system is carried into a cement carrier named "Pacific Seagull" as shown in Figure 13. The ship has a prime Diesel engine and three fore-stroke Diesel engine generators. The SCR system is set to an exhaust pipe of No. 3 Diesel engine generator. Since the No.3 Diesel engine generator runs and stops optionally in navigations, the navigations are not affected even if the SCR system has any troubles. Table 3 lists specifications of the Diesel engine generator.

Design-flowchart and performance prediction of SCR system

Figure 14 shows a design flowchart of the SCR system. As design conditions of the SCR system, values of flow rate, temperature and NOx concentration of exhaust gas are needed. However, some data could not be obtained due to the early design stage, so we estimated the SCR performance using estimated values.

SV value is set to 11000 h⁻¹ at rated power. And the number and length of the catalysts are decided with considerations of the carried space in the engine room and pressure loss of the catalysts.

Table 5. The engine specification

<table>
<thead>
<tr>
<th>Engine</th>
<th>Generator on ship</th>
</tr>
</thead>
<tbody>
<tr>
<td>Engine type</td>
<td>4-stroke</td>
</tr>
<tr>
<td>Bore-Stroke</td>
<td>165mm - 210mm</td>
</tr>
<tr>
<td>Num. of Cylinders</td>
<td>6</td>
</tr>
<tr>
<td>Rated Power</td>
<td>353 kW</td>
</tr>
<tr>
<td>Rated Engine speed</td>
<td>1200 rpm</td>
</tr>
<tr>
<td>Fuel</td>
<td>heavy oil (A)</td>
</tr>
</tbody>
</table>

Table 6. Specifications and Predicted Performance of SCR

<table>
<thead>
<tr>
<th>Catalyst Type</th>
<th>Sample A for low temp.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Size</td>
<td>150 x 150 x 260mm</td>
</tr>
<tr>
<td>Number</td>
<td>30</td>
</tr>
<tr>
<td>Cell (Mesh Num.)</td>
<td>45 (per 1 edg)</td>
</tr>
<tr>
<td>Space Velocity</td>
<td>11000 h⁻¹ at rated power</td>
</tr>
<tr>
<td>NOx conversion rate</td>
<td>79.5~79.8 % (at 80% equivalent ratio)</td>
</tr>
<tr>
<td>Slip-Ammonia</td>
<td>less than 6 ppm</td>
</tr>
<tr>
<td>Reducing Agent</td>
<td>40% Urea Water</td>
</tr>
<tr>
<td>Urea Flow Rate</td>
<td>60~120 mL/min</td>
</tr>
</tbody>
</table>

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As the result of calculation with experimental equations proposed from foregoing catalyst component tests in our laboratory, it is confirmed that the SCR has enough NOx removal performance and few slip-ammonia. Table 6 lists specifications and the predicted performance of the SCR.

Figure 15 shows the installation of the SCR to the Diesel engine generator in the ship. A urea injection nozzle and a catalyst case are located into the straight part of the exhaust pipe at upper side of the generator.

Generally, it must keep the distance between the nozzle and the catalysts as long as possible to secure enough time for urea to be changed into ammonia. However, in the case of the ship, both the nozzle and the catalysts should be located in the straight apart of 2 m, and the located space is limited.

Figure 16 shows a detailed plan of the SCR. The distance between the nozzle and the catalysts is only 840 mm approximately. It is too shorter than that of previous SCR system. Therefore, we have to improve the structure of the nozzles and a urea flow control system with repeating of operations.

**Operating conditions and measurement SCR**

The Load conditions of the Diesel engine generator cannot be set optionally for onboard tests. The output at the general navigation and anchored condition are about 180 to 200 kW that means load rate of 50 to 60 %. In the operating condition, the exhaust gas temperature is about 330 to 360 deg C.

Handy-type analyzers, testo 350-XL, measure the NOx concentration of upstream and downstream of the SCR, though the analyzers cannot be used continuously.

**Development of urea injection nozzles and NOx removal performance**

After the installation of the SCR system to the ship, we did several onboard tests. From the results, we confirm that a type of the urea injection nozzles affects strongly NOx removal performance. Therefore, we develop and improve different types of the nozzles, and use them in the field test.

Figure 17 shows structures of the urea injection nozzles used in the field test. These nozzles consist of duplicated stainless tubes. The urea flows in the inner tube, and the air flows the circular space between the outer tube and the inner tube. Type A as shown in Figure 17 (a) flows the urea-jet with the same direction of the exhaust gas, such as...
previous nozzles. Type B as shown in Figure 17 (b) flows the urea-jet with the right angle direction of the exhaust gas, because the space in the exhaust pipe around the nozzle is used effectively for converting from urea to ammonia. Also two kinds of Type B are prepared for the test. One has 4 holes, and another has 6 holes for the jet. Type C as shown in Figure 17 (c) is a revised type of Type B in order to keep stable the jet from plural holes. It is confirmed that Type C makes suitable water mists without exhaust gas flow.

Figure 18 and Figure 19 show examples of the test results of NOx concentration and the NOx conversion rate as a function of the equivalence ratio. And the NOx conversion rate is defined the ratio of removed NOx concentration and the NOx concentration before the catalysis.

From these results, the NOx removal performance using Type A and Type B (4 holes) are lower than that of other nozzles. It is considered that Type A cannot be evaporated urea mists satisfactorily in the exhaust pipe. Also, Type B (4 holes) has low assembling accuracy, and it cannot make suitable mists. In the case of Type B (6 holes), it is confirmed to deteriorate after 13 hours operation, though it has good performance in the early stage. The reason is that the clearance of the inner and outer tube has not kept suitable in the operation.

On the other hand, Type C has good performance both in the early stage and after 30 hours operations. However, it is confirmed that a lot of solid products, cyanuric acid, are accumulated in the exhaust pipe after 50 hours operations. It is considered that partial temperature drops with collisions between the urea mists and the inner wall of the exhaust pipe cause the trouble. More improvements of the nozzle and a control method for urea flow are needed in order to keep the stable performance in long-time operations.

**Operation of urea-flow control system**

Also we have developed a urea flow control system for the field test as shown in Figure 20. The urea water flows from a urea tank to the nozzle through a pump. A flow rate of the urea is controlled using an inverter that is set to A/C induction motor of the pump. A programmable logic controller, PLC, controls the inverter and several electromagnetic valves for air, urea and water.

A sequential control program of the PLC has been developed in the ship with the SCR operations. The SCR is programmed to start when the following 3 conditions are satisfied, (i) the exhaust gas temperature is higher than the set temperature, 300
(i) the temperature is kept in a set time, 3 minutes continuously, and (ii) a digital signal of L/O pressure of the Diesel engine generator has output voltage, that means the engine is running.

The urea flow rate is decided by collecting data of the exhaust gas temperature only. In the calculation of the urea flow rate, NOx concentration and the load of the Diesel engine generator are not used.

When the exhaust gas temperature is lower than a set temperature, 300 deg C, the PLC is programmed to start a sequence routine to stop the SCR operation. The set temperature should be somewhat higher than the temperature in an idling operation. In the sequence routine, the valves are switched from the urea to the water automatically. The nozzle is washed in a set time, 1 minute. And the valve of water and air are closed sequentially.

A ship engineer stops the engine manually after 5 minutes from starting the idling, because the PLC does not communicate to the engine control system.

Figure 21 shows an onboard test result of the urea flow control system.

In this chapter, development the SCR for field test and its results are described. Then it is confirmed that our SCR system shows suitable performance in the early stage of the field test. Also, the field test of total 100 hours brings us a lot of knowledge, such as the importance of urea injection nozzle. In the next step of our on board study, we will improve the SCR system to operate longer time, and find more technical problems.

CONCLUSIONS

In this paper, we show test results of several experimental studies in our project. From the result of catalyst component tests, it is show that NOx removal performance can be estimated. Secondly, it is clear that NOx in exhaust gas of the medium speed marine Diesel engine are reduced with the SCR system. Accordingly, NOx reduction is achieved with the designed and developed the SCR system on ship at sea.

In conclusion, we have got a lot of beneficial results to apply a SCR system to a medium speed marine Diesel engine. The results of this paper contribute to developments of practical-used marine SCR system. We will study and develop a SCR system for marine Diesel engine in future.
ACKNOWLEDGMENT

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