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ABSTRACT

In Japan there is currently a strong social demand for exhaust emissions reduction from heavy-duty diesel engines. Therefore, new Long-Term Regulation will come into effect in October 2005, setting the NO_x standard at 2.0 g/kWh and the PM standard at 0.027 g/kWh. At the same time, customers always demand exceptional fuel economy from heavy-duty commercial vehicles. A urea-based Selective Catalytic Reduction (SCR) system was developed to satisfy both these demands, and will be introduced in the fall of 2004.

The operating conditions of vehicles in Japan are different from those in the US and Europe. Basically, average vehicle speeds are significantly lower. To improve the low temperature SCR performance, an oxidation catalyst was located upstream of the SCR, and an additional oxidation catalyst was located downstream of the SCR for emergency NH₃ slip. The muffler size with all three catalysts was similar to a conventional muffler.

The fuel and urea water solution consumption of the system was studied and compared with the fuel consumption of a similar system equipped with EGR and a Diesel Particulate Filter (DPF) unit.

The properties of the urea water solution used here were not well known at the time of this project. Therefore, the specifications and corrosion characteristics of relevant materials were investigated and suitable materials were selected. Urea water solutions freeze at around -11 deg C, so a thawing system was also developed.

Vehicle tests were carried out at various conditions, during which no major problems arose.

INTRODUCTION

A large proportion of the expenses of a transportation business is for fuel. So commercial vehicles are exclusively powered by diesel engines due to their high efficiency. On the environmental side, carbon dioxide emissions (a greenhouse gas) are low, but further

reduction of oxides of nitrogen (NO_x) and particulate matter (PM) are strongly required. New long-term exhaust emissions regulations will be introduced in Japan in October of 2005. The new maximum values will be NO_x = 2.0g/kWh and PM = 0.027g/kWh, which will be the lowest values in the world at that time.

It is impossible to meet the 2005 NO_x and PM levels by engine modification only. Notably, NO_x levels have a trade-off relationship with fuel economy, so if an after-treatment system can be used, which can substantially reduce NO_x levels, good fuel economy can be maintained. Therefore we combined engine modification with SCR (Selective Catalytic Reduction), a popular de-NO_x catalyst technique, and chose urea solution as the reducing agent [1-3]. This combination made it possible to achieve the new long-term exhaust emission levels and get improved fuel economy as well, without using a DPF (Diesel Particulate Filter).

On the other hand, use of a urea-water solution with an SCR is still somewhat new, and there are many issues that must be clarified, an example of which is handling the urea-water solution. Urea-water solutions are not used in the automotive field, so the characteristics and influence of many SCR system components had to be carefully planned out. Therefore, we investigated the influence of various materials, and the fundamental characteristics and stability of the urea-water solution. Since it freezes at -11 deg C, low-temperature countermeasures were applied. Development of the optimum urea-water supply system was a major part of this study.

The other important characteristic is the overall system durability, including the catalyst. We evaluated this parameter by on-road running tests.

The vehicle which used this system and meets the new long-term emissions regulations was released to the market in the Fall of 2004, about one year before the new regulations take effect.

DEVELOPMENT OF UREA SCR SYSTEM

Figure 1 shows a schematic of the Urea-SCR system developed in this project. The main components are the

catalysts and the plumbing to add the reducing agent to the exhaust stream. The reducing agent is a 32.5% urea water solution, and it is injected into the exhaust gas stream in front of SCR catalyst. Once injected, the urea undergoes hydrolysis reactions to form ammonia (NH_3), and the NH_3 reacts with NO_x , resulting in N_2 .

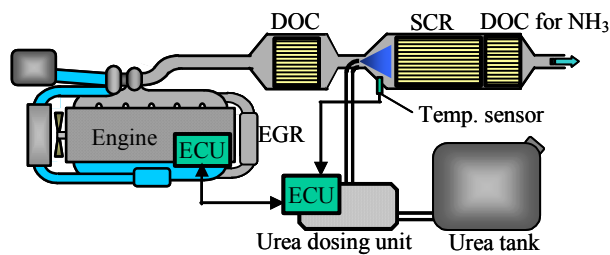


Fig. 1. Schematic of the Urea-SCR System

CATALYST DEVELOPMENT

Improvement in catalyst performance at low-temperatures

The Japanese transient cycle (JE05) adopted for the new long-term regulations has a relatively higher emphasis on low speed and low load operation, compared with the U.S. (FTP) and European (ETC) test cycles (Figure 2). As a result, the exhaust gas temperatures are generally lower than those of the European and US test cycles.

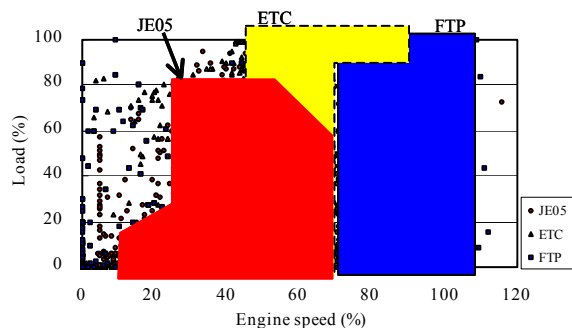
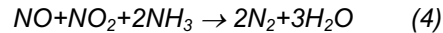
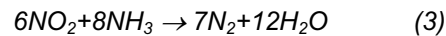
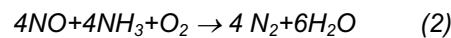
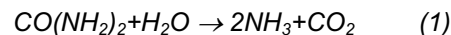


Fig. 2. Comparison of engine speed and load during JE05, ETC, FTP test cycles

It was thus decided to focus upon improving NO_x reduction reactions of the general low-temperature for the catalyst used here. The urea hydrolysis and NO_x reduction reactions are shown by reactions (1) - (4). There is a 1:1 ratio of $\text{NO}:\text{NO}_2$ in reaction (4), and since this reaction is most active, it is effective to focus here for improvement in the rate of conversion at low temperatures.



Note that the DOC (Diesel Oxidation Catalyst) was located upstream of the SCR catalyst and optimized the activity. Table 1 shows the catalyst specifications and Figure 3 shows NO_x to N_2 conversion with and without the DOC. DOC "A" (in table 1) was selected for use in this project, mainly due to its high NO to NO_2 activity. The SCR with the DOC can sharply improve low-temperature NO_x conversion during many low-speed low-load portions of the JE05 test cycle.

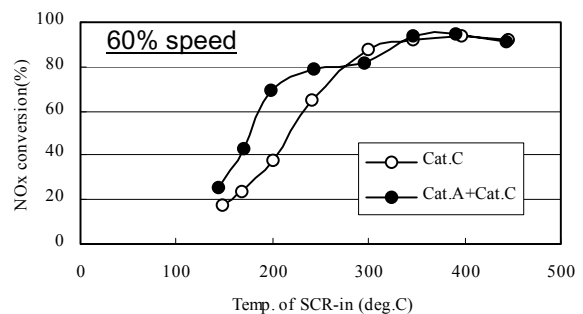


Fig. 3 Effects of DOC on NO_x conversion

Selection of an SCR catalyst

There are vanadium and zeolite type SCR catalyst systems. The vanadium type has already been adopted as the SCR system of choice for stationary engines, and it is coming into more widespread use in vehicular applications. But the vanadium type catalyst is formed by squeeze out, and is considered to be large in size. It is thus difficult to integrate this unit into a vehicle at this time. On the other hand, the zeolite type catalyst is coated on a ceramic substrate, and can easily be small in size compared with the vanadium type.

There is not a significant difference in NO_x conversion between the vanadium and zeolite type catalysts, as shown in Figure 4. Furthermore, the catalyst volume was considered, as shown in Figure 5. This plot shows a comparison of NO_x conversion with the vanadium and zeolite type catalysts against catalyst volume. The vanadium type shows a tendency for the NO_x conversion to decrease with reduction in volume, while for the zeolite type a higher conversion efficiency is indicated for a given volume.

As mentioned above, the zeolite type catalyst was chosen for the present system, in consideration of the vehicle mounting situation and integration circumstances.

Table1 Urea-SCR catalyst specifications

		DOC		SCR				NH3-SLP
		A	B	C	D	E	F	G
Diameter	mm(inch)	266.7(10.5)	←	241.3(9.5)	266.7(10.5)	←	←	←
Length	mm(inch)	152.4(6.0)	101.6(4.0)	254(10)	152.4(6.0)	←	←	101.6(4.0)
Volume	L	8.5	5.7	11.6	8.5	←	←	5.7
Cell Density	cell/inch ²	400	←	300	400	←	←	←
Composition		Pt-type	←	Vanadium-type	Zeolite-type I	Zeolite-type II	Zeolite-type III	Pt-type

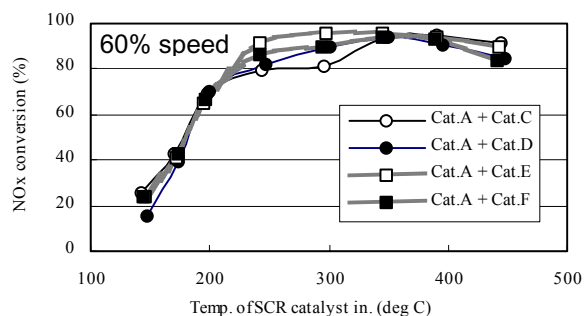


Fig. 4 Influence of catalyst on NOx conversion

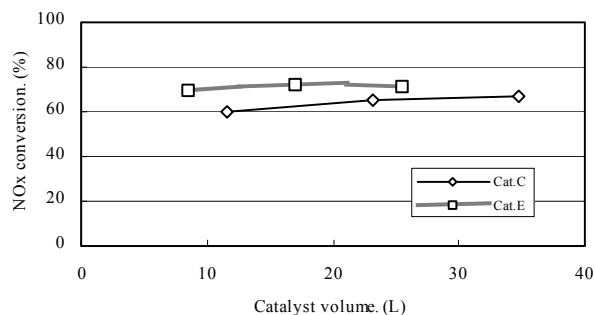
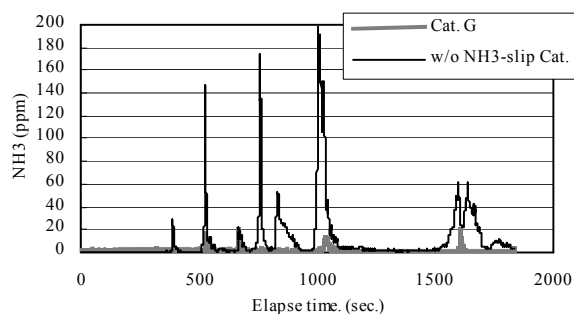


Fig. 5 Influence of catalyst volume on NOx conversion (JE05)

Prevention of NH₃ slip

If more-than-sufficient NH₃ reducing agent is supplied to the SCR for the amount of NOx present, the excess NH₃ (which could not react with NOx) will slip. In general, an equivalent amount of urea solution is injected for the NOx present, but the downstream DOC is included for such cases of emergency NH₃ slip.

The oxidation performance of the NH₃ slip catalyst is shown in Figure 6. By use of this catalyst, even when there is NH₃ slip, the tailpipe levels can be reduced sharply.

Fig. 6 Performance of NH₃-Slip catalyst (JE05)

DEVELOPMENT OF UREA WATER-INJECTION EQUIPMENT

A Urea-SCR system has to inject the solution at the proper location and time for optimum NOx reduction, to minimize NH₃ slip, and to efficiently utilize the urea solution. Figure 7 shows the urea solution addition equipment, made by Bosch.

An air assist type nozzle was used to inject the urea solution. This type of injector has good atomization and dispersion of the urea solution and resists plugging. Furthermore, an electric heater is built into the injector body and piping to prevent freezing, and for thawing frozen solution. Further details are provided in a later section.

The urea water-injection equipment has ECU and it took on the task of urea solution injection control. Input variables included activity temperature, adsorption and desorption of NH₃, NOx discharge level, etc. With this arrangement, system optimization was carried out. Results indicated high levels of NOx conversion without NH₃ slip. (Figure 8)

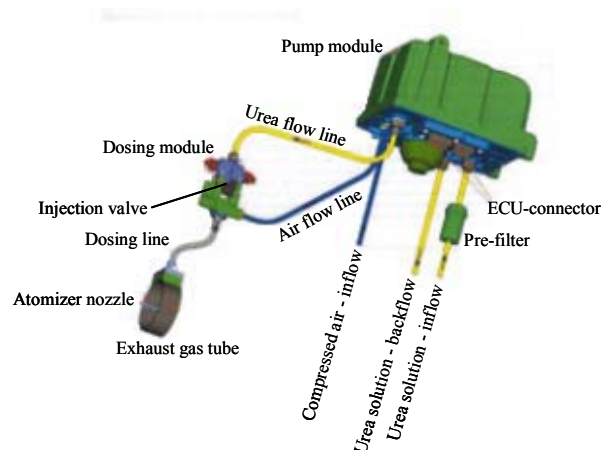
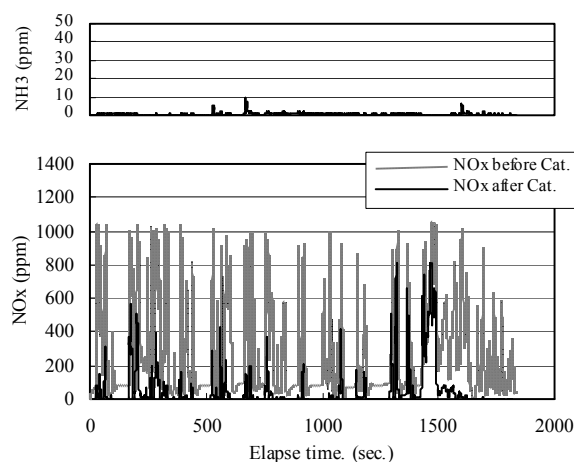


Fig. 7 Schematic of urea metering unit

Fig. 8 NOx and maximum NH₃ concentration on JE05 mode.

ENGINE DEVELOPMENT FOR OPTIMIZATION WITH UREA SCR

Engine development was carried out on the assumption that NOx can be reduced with a urea-SCR system. The fuel-injection pressure was increased to reduce PM emissions. For the 13L engine used here, we adopted a new miniaturized unit injector. Then the fuel injection system and combustion system were re-

optimized, which allowed us to achieve PM levels below the new long-term regulated value - without a DPF. Furthermore, a cooled EGR system was used to reduce NO_x emissions in the low exhaust temperature region, in which the catalyst does not perform well. Table 2 shows the specifications of the two engine types.

Table 2 Engine specifications

Engine Type	DI, L6, TCI, 4-Stroke	DI, L6, TCI, 4-Stroke
Displacement	13.074 L	9.203 L
Bore × Stroke	φ 136mm×150mm	φ 125mm×125mm
Compression ratio	17.5	16.5
Injection system	Unit Injector	Common rail
Maximum power	280kW/1800rpm	250kW/2200rpm
Maximum torque	1648Nm/1400rpm	1446Nm/1400rpm
Maximum Inj. Press.	200MPa	160MPa
EGR	Cooled EGR	Cooled EGR

EXHAUST-GAS MEASUREMENTS TO DETERMINE COMPLIANCE WITH NEW LONG-TERM REGULATIONS

Test results of the 13L engine for the JE05 cycle are shown in Figure 9. Values are shown comparing emissions levels for the engine equipped with the Urea-SCR system to an engine without the Urea-SCR system. The results show that NO_x and PM were reduced sharply and did not exceed the new long-term regulated values.

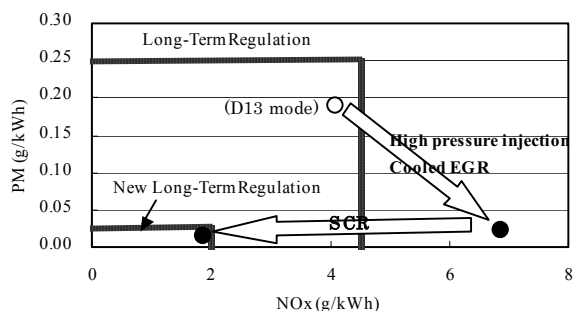


Fig. 9 JE05 exhaust emissions test results

IMPROVEMENT OF FUEL ECONOMY BY SCR SYSTEM APPLICATION

There are two approaches to achieving the new long term emissions regulations [4]. One is to reduce PM by in-cylinder combustion improvement and reduce NO_x with a Urea-SCR catalyst. The other is to reduce NO_x by high levels of EGR and reduce PM by using a DPF. Fuel economy may be modeled and compared when using these two approaches to determine which is more promising. Generally, fuel economy is related to NO_x emissions levels. Figure 10 shows fuel economy as a function of NO_x level at 60% speed and 60% load, with the NO_x level varied by injection timing and EGR rate. It

can be seen that there is a strong dependence of fuel consumption on injection timing and EGR rate.

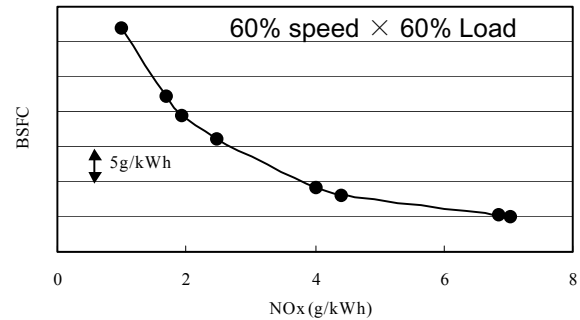


Fig. 10 NO_x Fuel Consumption Trade-off

When comparing the fuel economy of a vehicle with an engine using high levels of EGR and a DPF, we need to account for a fuel consumption penalty of about 3% due to active regeneration of the DPF (for some driving conditions).

For the Urea-SCR approach, one must account for the urea solution's cost, after determining the consumption and price. The consumption changes with driving conditions, but it is at most about 5% of fuel consumption. If the urea solution price is half the fuel price, the cost penalty is 2.5% of fuel cost.

As mentioned above, we simulated the fuel economy of vehicles utilizing the two above approaches to achieve the new long-term regulations. Conditions were 90km/h constant speed, which is a typical running condition for heavy-duty commercial vehicles in Japan. In this case, DPF does not need active regeneration, so the resulting fuel consumption penalty is not observed. The simulation results are shown Figure 11. For the Urea-SCR system, the fuel consumption is 6.5% better than the high EGR + DPF case. Accounting for the urea solution's cost, it is 4% better. For the city driving mode, which has more low speed conditions, urea consumption is lower, and active regeneration is needed for the DPF, so a greater fuel economy advantage appears for the Urea-SCR system.

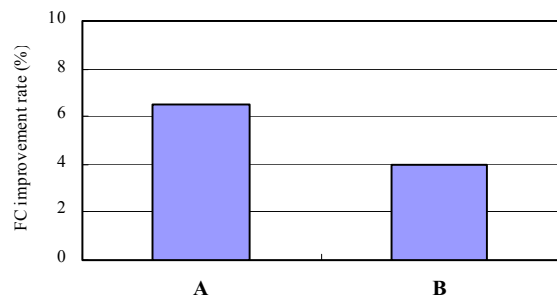


Fig. 11 Fuel consumption Improvement for the Urea-SCR system compared to high EGR+DPF system at 90km/h speed

A: Not including urea solution cost
B: Including urea solution cost

UREA WATER SOLUTION

CHARACTERISTICS OF UREA WATER SOLUTION

The SCR system uses ammonia (NH_3) as the actual NO_x reducing agent. NH_3 , however, requires high-pressure gaseous storage and handling. This makes ammonia unsuitable for vehicular use. Urea is easier to handle and safer to store and use. Urea is a white odorless crystal, is used for cosmetics (moisturizer) and medical supplies. Additional information is available on the product safety data sheet.

Urea, which is in crystalline form, dissolves in water and is used at a concentration of 32.5%. The main characteristics of the 32.5% urea water solution are shown in Table 3. Its specific gravity is about 1.1, it is a colorless and odorless liquid. Its freezing point is -11 deg C.

Table 3 Properties of 32.5% urea solution

Concentration (wt%)	32.5
Density at 20°C	1.090
Freezing point (°C)	-11
pH	8.8
Color	colorless
Odor	odorless

Considering typical vehicular operating conditions, a low freezing point was desired. The minimum freezing temperature is achieved at the 32.5% urea concentration. Figure 12 shows the relation between urea concentration and freezing point. The 32.5% concentration results in the lowest freezing point of -11 deg C. Stronger or weaker solutions have a higher freezing point.

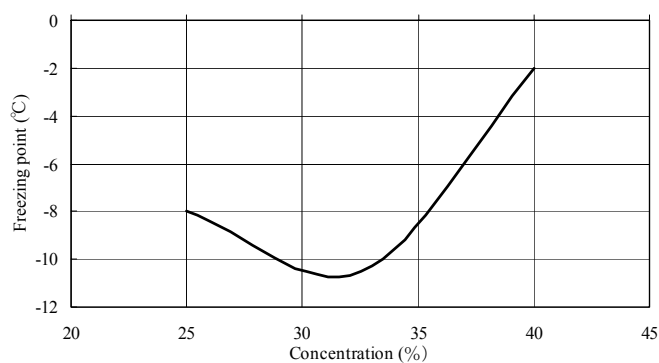


Fig. 12 Influence of concentration on freezing point

From this point we will always assume a 32.5% urea concentration in this work. For other work using different concentrations, one should remember that the NO_x conversion rate, and NH_3 slip amount may be different. Since the solution is injected through relatively small tubing, plugging due to impurities must be avoided.

Japan and Europe are establishing standards for urea water solutions on the basis of "Refined Urea" and ion exchange water. Automotive Standard JASO E502 was established in Japan, and this will become a JIS

standard in the near future. Similarly, the German federal standard DIN70070 was enacted by the European commission. Work is being done to create an ISO standard based on the JASO and DIN standards.

Table 4 Highlights of the JASO standards

Characteristics	Unit	Limits	
		Min	Max
Concentration	weight-%	31.8	33.3
Density at 20°C	g/cm ³	1.087	1.092
Refractive index at 20°C	—	1.3817	1.384
Alkalinity as NH_3	%	—	0.2
Carbonate as CO_2	%	—	0.2
Biuret	%	—	0.3
Aldehyde	mg/kg	—	10
Insolubles	mg/kg	—	20
Phosphate(PO_4)	mg/kg	—	0.5
Calcium	mg/kg	—	0.5
Iron	mg/kg	—	0.5
Copper	mg/kg	—	0.2
Zinc	mg/kg	—	0.2
Chromium	mg/kg	—	0.2
Nickel	mg/kg	—	0.2
Magnesium	mg/kg	—	0.5
Sodium	mg/kg	—	0.5
Potassium	mg/kg	—	0.5

STABILITY OF UREA WATER SOLUTION

Urea water solutions may be left in storage for long periods during the distribution process and when the vehicle is not regularly operated. We investigated the long-term stability of urea water solution in terms of the solution's pH, density, and sediment formation.

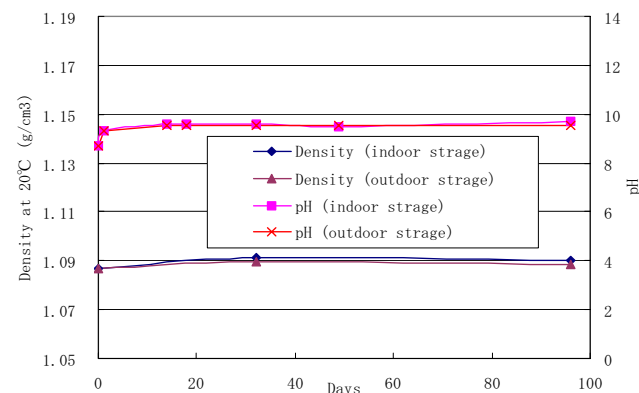


Fig. 13 Urea water stability in storage

Figure 13 shows analysis results for a urea water solution left outdoors for about 14 weeks (96 days). The pH and density were measured. In spite of the sunlight, the concentration was unchanged, and there was also no sediment formation.

INFLUENCE OF UREA WATER SOLUTION ON WETTED MATERIALS

In order to investigate the influence of urea water solution on wetted materials in the Urea-SCR system, such as the tank and piping, soak testing of metal, resin, and rubber materials was performed. A sample of test material was placed in the urea water solution and another in distilled water for comparison. From here, tests were performed with immersion at 80 deg C test temperature for a maximum 1,000hr. We describe here the test results of 80 deg C x 120hr, which were the initial material selection conditions.

(1) Tests of Metallic Materials

The amount of corrosion was measured for stainless steel, aluminum, cast iron, steel (galvanized), brass, and copper. These materials may be used for storage tank or piping. The test results are shown in Figure 14.

Although notable corrosion was found on the copper and brass, others were unchanged. Therefore the stainless steel was adopted for the storage tank.

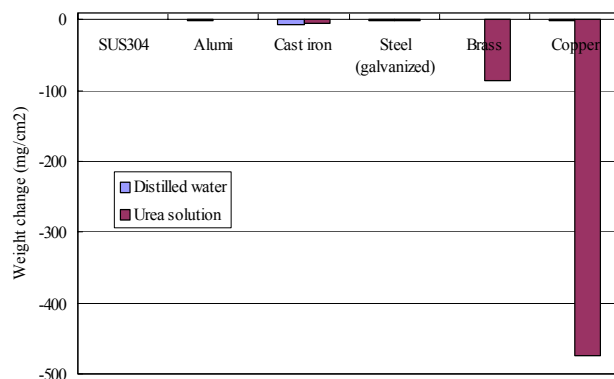


Fig. 14 Corrosion of metal samples

(2) Tests of Resin Materials

Volume, Dimensional and weight changes were measured for PFA, PA, PVC, and PVC+PTFE. Volumetric change of these test results are shown in Figure 15. PFA and PA12 were adopted for piping, since these samples exhibited little volumetric, dimensional or weight changes compared with the other resin materials.

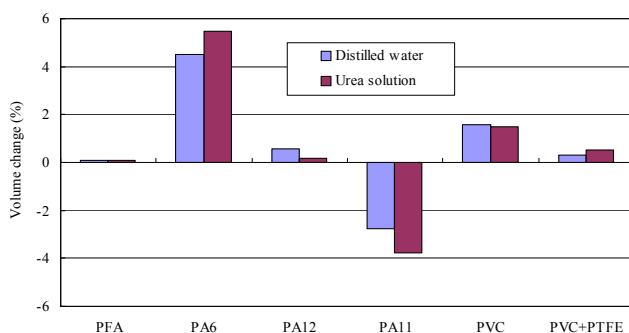


Fig. 15 Volumetric change of plastics

(3) Tests of Rubber Materials

The change in volume and hardness were measured for EPDM, NBR, and NBR+PVC. Figure 16 shows volumetric change of these samples. It can be seen that

EPDM shows the least volume and hardness change compared with the other materials. EPDM was therefore used for seals in the Urea-SCR system.

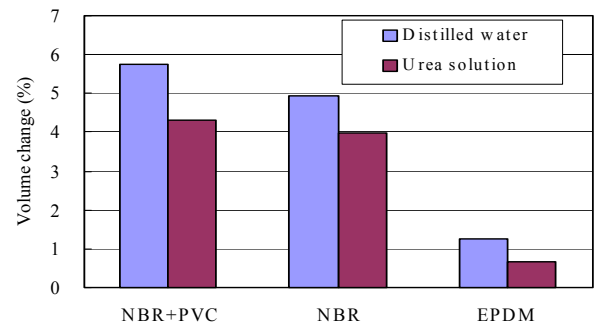


Fig. 16 Volumetric change of rubber materials

UREA WATER SUPPLY SYSTEM

It is convenient for a truck driver to fill the urea water solution simultaneously with a fuel. About 80% of the domestic medium and heavy-duty truck drivers use own fuel tank or truck station, as shown in Table 5. So, for drivers who use own fuel tank, they can supply urea water solution for their own tank too. There are seven main in-tank bases in Japan, which are nearly ready to supply urea water solution to vehicles. The wholesale supply organization to about 80 percent of the roughly 1,000 truck stations nationwide is also ready to begin supplying urea water solution. Further, the supply organization to all NISSAN DIESEL dealers nationwide is also ready to begin supplying urea water solution. A container for storage of emergency backup urea water solution was also developed. When our trucks equipped with the Urea-SCR system were released to the market, adequate supply infrastructure was already attained.

Table 5 Share of fuel filling stations (Heavy-duty truck users)

	number	share
Truck station	≈1,000	20~25%
Freight own tanks	≈10,000	50~60%
Gas station	≈44,000	20~25%

The driver can monitor a urea water solution indicator in the instrument panel, which tells at a glance the quantity remaining in the tank (Figure 17).



Fig. 17 Urea solution level indicator

Vehicle	Type	Heavy Duty Truck
	Gross vehicle weight	24,840kg
	Vehicle weight	8,730kg
Engine	Type	DI , TCI
	Numbers of cylinder	6
	Bore×Stroke	φ 136×150mm
	Displacement	13.074L
	Rated power	272kW/1900rpm

CONCLUSION

A Urea-SCR system has been applied to heavy-duty commercial vehicles, for the purpose of achieving a drastic reduction of emissions.

(1) A small zeolite type SCR catalyst was developed, which achieved high levels of NO_x reduction.

(2) A DOC unit was installed upstream of the SCR catalyst, which improved the low-temperature NO_x conversion rate.

(3) A NH₃ oxidization catalyst was installed downstream of the SCR catalyst; this prevented NH₃ slip.

(4) The engine was developed on the assumption that NO_x emissions can be greatly reduced by the Urea-SCR system, thus PM can be reduced without a DPF.

(5) The above catalysts and the improved engine were combined, and with further optimization of urea addition control, the vehicle could achieve the new 2005 long term emissions regulations.

(6) By the specifications corresponding to the new long term regulation, the Urea-SCR system's fuel consumption improvement is estimated to be 6.5% compared with a similar unit using the high-EGR + DPF technique for high-speed operation. Even if we take the consumption of urea solution into consideration, there is still a 4% advantage.

(7) Important properties and characteristics of the urea water solution used for an automotive urea SCR catalyst have been clarified.

(8) The effect of the urea-water solution on wetted system materials was studied, and optimum materials were selected for the Urea-SCR system components.

(9) Low ambient temperature freeze prevention and thawing systems for the urea storage and injection systems were developed.

(10) Several test vehicles using Urea-SCR systems have amassed a combined 700,000km distance, and to date there have been no major troubles nor a decline in the NO_x conversion rate.

Many vehicles equipped with this Urea-SCR systems will arrive on the market in the Fall of 2004, about one year before the new long-term emissions regulations take effect. We plan to continually make improvements to these emissions control systems and the vehicles, as many customers are relying on us.

ACKNOWLEDGMENTS

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