Reduction of SO₂ and DPM Using Heat Exchanger and Electrostatic Precipitation in Diesel Engine

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The electrostatic precipitator (ESP) with heat exchanger was developed to reduce particles and SO₂ in a diesel engines. The influence of gas cooling on the particle and SO₂ concentrations was investigated using a 400-cc diesel engine. The particle concentrations were measured with a scanning mobility particle sizer (SMPS) and a low volume air sampler.

Soluble Organic Fraction (SOF) and SO₄²⁻, which make up the composition of the particle, were also evaluated using a soxhlet extractor and an ion chromatography. SO₂ concentration was measured with a UV fluorescence SO₂ monitor. The results show that the number and the volume concentration of particles increased due to generated SOF and sulfuric acid particles by gas cooling. It was also indicated that SO₂ concentration in the gas was reduced due to absorption into condensed water. Although the particle concentration increased, ESP showed a high particle collection efficiency.

1. Introduction

Diesel engines are commonly used as main and auxiliary engines in ships due to their high heat efficiency and low CO₂ emissions. However, their exhaust gas contains air pollutants, such as sulphur oxides (SOx) and particulate matters (PM). In 2008, to overcome this problem, the International Marine Organization (IMO) adopted the MARPOL 73/78 Convention Revised Annex VI¹, which stipulates that ships should use low-sulphur fuel oil or be provided with an exhaust-gas cleaning system (EGCS) whose capability is equivalent to using low-sulphur fuel oil².

As for an EGCS intended for reducing SOx in ship exhaust gas, a wet scrubber which uses seawater as the main absorbent of SOx has begun to be practically used in Western countries³. However, this system requires large-scale facilities. As a method for improving the performance of the wet scrubber, the use of seawater electrolysis has been under study and development. This method includes producing alkaline water by electrolysis and spraying it into exhaust gas to efficiently reduce SOx⁴-⁵. It also has the effect of removing CO₂⁶) and NOx⁷) from the exhaust gas by the NaOH and chlorine gas resulting from the electrolysis.

As for the removal of diesel particulate matters (DPM) in ship exhaust gas, various techniques have been studied and developed, such as the application of a diesel particulate filter (DPF) used in automobiles⁸),⁹), as well as the use of electrostatic scrubbers¹⁰),¹¹), electrostatic cyclones¹²), barrier dischargers¹³), and electrostatic precipitators (ESP) using the Hole effect or electrohydrodynamics (EHD)¹⁴)-¹⁶).

Diesel particulate filters (DPF) can be made of a heat-insulating material⁸), ceramic⁹) or the like. Regeneration of a DPF normally employs an external heat source, while a method using plasma has also been proposed⁹). For electrostatic scrubbers, one study is aimed at improving their performance by electrically charging a spray of water and the DPM with opposite polarities¹⁰), while another study is focused on what effect will be obtained if seawater is used as the electrostatically charged water spray¹¹). Electrostatic cyclones are capable of trapping fine particles by an electrostatic precipitator (ESP) and large particles by a cyclone¹²). As for the ESP, one study is concerned with the use of barrier discharge to oxidize DPM into gas¹³). Unprecedented methods have also been proposed: One example is the Hole-type ESP in which a space with low wind velocity is created by a double-cylinder electrode structure and

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re-entrained particles are guided into this space\textsuperscript{14). Another example is the electrohydrodynamic ESP which uses corona wind to guide re-entrained particles into a field-free space\textsuperscript{15).}

Meanwhile, Nishida et al. reported that cooling the diesel exhaust gas of approximately 573 K to approximately 423 K caused an increase in the concentrations of the DPM and soluble organic fractions (SOF) due to the condensation of volatile substances. They also demonstrated that those components could be efficiently removed by a single-stage ESP\textsuperscript{16).}

This finding presented us with the idea that cooling the exhaust gas to 373 K or lower temperatures where the water condensation occurs would cause SO\textsubscript{2} in the exhaust gas to be absorbed into water droplets. Transforming SO\textsubscript{2} and SOF into particles facilitates trapping them by ESPs. Accordingly, we have investigated the effect of gas cooling coupled with the ESP with the aim of removing SO\textsubscript{2} and DPM in the exhaust gas (from ships). The exhaust gas actually used in the experiment was prepared by operating a small 400-cc diesel engine fueled with Bunker A. The concentrations of SO\textsubscript{4}\textsuperscript{2–}, SOF and Dry Soot in the condensed water and DPM were measured after the exhaust gas was cooled to approximately 293 K. A DPM collection efficiency in the ESP was also investigated.

2. Principle for Removal of SO\textsubscript{2} and SOF

Fig. 1 shows the principle for the reduction of SO\textsubscript{2} and SOF. The system consists of a heat exchanger and an electrostatic precipitator (ESP). The exhaust gas discharged at a temperature of approximately 453 K contains SO\textsubscript{2} as well as gaseous H\textsubscript{2}O and SOF. This exhaust gas is cooled to 293-303 K by the heat exchanger to condense H\textsubscript{2}O and SOF in the exhaust gas into particles. The resulting water particles absorb SO\textsubscript{2} and form a mist of sulphuric acid, which reduces SO\textsubscript{2} in the exhaust gas. The DPMs inclusive of the generated particles are trapped by the ESP. Thus, SO\textsubscript{2} and SOF are removed from the exhaust gas.

3. Experimental Setup

3.1 Experimental System

Fig. 2 shows a schematic of the experimental system. The system consists of a water-cooled 4-cycle diesel engine (DA-3100SS-IV, manufactured by Denyo Co., Ltd; cylinder volume, 400 cc; output, 5.5 kW), a heat exchanger and an ESP. In the experiment, Bunker A (ENEOS LSA fuel oil; sulphur content, 0.09 \%) was used as a test fuel oil.

The heat exchanger can cool the exhaust gas from approximately 453 K to 373-293 K using a refrigerant (city water) of approximately 293 K with a controlled flow rate (ranging from 0.5×10\textsuperscript{–3} to 6×10\textsuperscript{–3} m\textsuperscript{3}/min). The cooled gas passes through the ESP, which removes DPM. The exhaust gas requires approximately 0.35 s to pass through the heat exchanger. The wind velocity inside the ESP is approximately 1 m/s.

In practical applications of this system, seawater may possibly be used as the refrigerant, in which case a liquid-supply pump is required. After-treatments of the condensed water (which contains sulphuric acid and SOF, and therefore has a low pH level) and the DPM collected in the ESP will also be necessary. A device for preventing an excessive increase in the waste-water temperature will also be needed. Additionally, wet scrubbers currently used at

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practical levels for cleaning exhaust gas from ships require approximately 100 m³/h of seawater for a 1000-kW engine, and a technique for the after-treatment of this waste water is expected to be necessary in the future. Our technique possibly provides a means for dramatically reducing the amounts of seawater supply and waste-water treatment.

3.2 Structure of Electrical Precipitator

The ESP has a two-stage structure of precharger and collector units. Fig. 3 shows the structure of each unit. The precharger has a parallel-plate electrode structure composed of high-voltage application electrodes (110×130 mm) and grounded plate electrodes (130×150 mm) alternately arranged with a gap of 9.5 mm. The high-voltage application electrodes have sawtooth edges on their upstream and downstream sides, while the grounded electrodes have no such edges. The collector also has a parallel-plate electrode structure composed of high-voltage application electrodes (110×130 mm) and grounded plate electrodes (130×150 mm) alternately arranged with a gap of 9.5 mm. All electrodes are made of stainless steel with a thickness of 0.8 mm. The precharger is supplied with DC voltage of –4 to –8 kV to generate negative corona discharge, while the collector is supplied with DC voltage of +10 kV to form an electrostatic field.

3.3 Measurement of Concentrations of Particulate Components in Exhaust Gas

To investigate the effect of our technique, we measured the generated amount of condensed water as well as the concentrations and particle-diameter distributions of SO₂ and DPM (SOF, SO₄²⁻ and Dry Soot) in the exhaust gas.

The condensed water was sampled from the drain pipe in the heat exchanger after the exhaust gas was cooled to 393-293 K. The sampled condensed water was passed through a filter, and the SO₄²⁻ concentration in the filtered water was measured with an ion chromatograph. By a preliminary experiment, it was previously confirmed that the filter adsorbs approximately 5 % of SO₄²⁻ when the condensed water is passed through it. The pH of the condensed water was measured with a glass-electrode hydrogen-ion concentration indicator (TPX-999i, Toko Chemical Laboratories).

The DPM concentration in the exhaust gas was measured by drawing a portion of the exhaust gas and passing it through a Teflon-coated glass-fiber filter to sample the DPM. The difference between the filter masses measured before and after the DPM sampling was substituted in equation (1) to calculate the mass concentration \( C_{DPM} \) [mg/m³]. The temperature of the sampling tube, filter holder and diluter was controlled by the tape heater so that it would be equal to the temperature of the drawn gas, in order to prevent the natural cooling of the sampling tube which may possibly cause condensation inside the tube and change the particle-diameter distribution or the ratio of components.

\[
C_{DPM} = \frac{(M_{DPM} - M_f)}{(Q_L \cdot t)} \quad (1)
\]

In equation (1), \( M_f \) and \( M_{DPM} \) are the filter masses [mg] before and after the DPM sampling, respectively. \( Q_L \) is the drawing rate \([16.7 \times 10^{-3} \text{ m}^3/\text{min}]\) and \( t \) is the drawing time [5 min]. The filter mass was measured before and after the DPM sampling by drying the filter at 323 K for two hours in a thermostatic oven and subsequently weighing it.

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From the filter thus sampled, the concentrations of the SOF, sulphate ion, bound water and Dry Soot in the exhaust gas were analyzed. The analyzing procedure for each component was as follows:

After the DPM concentration ($C_{DPM}$) was measured, the SOF concentration $C_{SOF}$ [mg/m$^3$] was calculated by subjecting the filter to a Soxhlet extraction with dichloromethane as the solvent and substituting into equation (2) the difference between the filter masses measured before and after the extraction. The filter was further subjected to ultrasonic washing to extract sulphate ion into ultrapure water. The extracted liquid was analyzed with an ion chromatograph to determine the sulphate ion concentration $C_{SO_4}$ [mg/m$^3$]. The bound-water concentration of the sulphuric acid $C_{BW}$ [mg/m$^3$] was assumed to be 1.3 times the sulphate ion concentration$^{17}$. Finally, the Dry Soot concentration $C_{DS}$ [mg/m$^3$] was calculated by equation (3).

$$C_{SOF} = \frac{(M_{DPM} - M_{ext})}{(Q_L \cdot t)}$$  \hspace{1cm} (2)

$$C_{DS} = C_{PM} - C_{SOF} - C_{SO_4} - C_{BW}$$  \hspace{1cm} (3)

$M_{ext}$ in equation (2) is the filter mass [mg] after the extraction of oil components.

To measure the SO$_2$ concentration and particle-diameter distribution, a portion of the exhaust gas was drawn from the duct and diluted by the diluter at the same temperature as the exhaust gas. The SO$_2$ concentration was measured with a UV fluorescence SO$_2$ analyzer (APSA-370, HORIBA). The particle-diameter distribution was measured with a scanning mobility particle sizer (3936NL76, TSI Incorporated). The measured values were substituted into equations (4) and (5) to calculate the SO$_2$ removal rate $\eta_{SO_2}$ and collection efficiency $\eta_P$.

$$\eta_{SO_2} = \{1 - (C_{SO_2D} / C_{SO_2U})\} \times 100$$  \hspace{1cm} (4)

$$\eta_P = \{1 - (N_{PD} / N_{PU})\} \times 100$$  \hspace{1cm} (5)

In these equations, $C_{SO_2U}$ and $C_{SO_2D}$ are the SO$_2$ concentrations [mg/m$^3$] on the upstream and downstream sides of the apparatus, respectively. $N_{PU}$ and $N_{PD}$ are the number concentrations of the DPM [part/m$^3$] on the upstream and downstream sides of the apparatus, respectively.

4. Result and Discussion

4.1 Water Condensation By Gas Cooling

To confirm the occurrence of water condensation due to the cooling of the exhaust gas, we measured the amount of condensed water obtained from the drain pipe of the heat exchanger. Fig. 4 shows the gas-temperature characteristic of the amount of condensed water in the heat exchanger. The graph shows the amount of condensed water collected per 25 minutes. No formation of condensed water was confirmed when the cooling temperature was 323 K or higher. When the gas temperature became lower than 323 K, the condensed water suddenly began to be formed, which amounted to 210 cm$^3$ at 303 K, and 420 cm$^3$ at 293 K. This result evidently proves that cooling the exhaust gas causes water condensation. A measurement with the pH analyzer demonstrated that the condensed water had a strong acidity of pH=2.

When the exhaust gas was cooled to 303 K, the amount of SO$_4^{2-}$ in the condensed water was approximately 5.5 mg/m$^3$ per 1 m$^3$ of the exhaust gas (in terms of the volume at a gas temperature of 297 K). It should be noted that this value includes the SO$_3$ concentration converted into the SO$_4^{2-}$ concentration. The actual concentration should be approximately 5 % higher, because, as already noted, approximately 5 % of SO$_4^{2-}$ is adsorbed by the filter when the condensed water is passed through the filter. The likely reason why SO$_4^{2-}$ was detected is that SO$_2$ in the exhaust gas was absorbed by the condensed water and reacted with H$_2$O to form a mist of sulphuric acid. SOF was also detected in the condensed water, although we did not measure its accurate mass. This is most likely because the gaseous SOF was condensed due to the cooling and caught into the condensed water.

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4.2 Influence on Particle Concentration

Considering that the water condensation could also affect the particle concentration, we measured the particle-diameter distribution and mass concentration. Both measurements were performed at the location of duct 2 in Fig. 2. Fig. 5 shows the influence of the gas temperature on the particle-diameter distribution. At any temperature, the particle-diameter distribution peaked at approximately 70 nm. The particle-diameter distribution barely changed within a temperature range from 393 K to 323 K. By contrast, within the range from 313 K to 293 K, the particle concentration increased with decreasing gas temperature. This is most likely due to the previously described phenomena, i.e. the formation of the mist of sulphuric acid by water condensation and the transformation of gaseous SOF into particles.

Fig. 6 shows the relationship between the gas temperature and DPM mass concentration. The mass concentration was 26.8 mg/m$^3$ at a gas temperature of 433 K, which increased with decreasing temperature, reaching 39.2 mg/m$^3$ at 313 K. From these results, it is evident that cooling the exhaust gas has the effect of transforming gaseous matters into particles.
4.3 Influences on SO₂ Concentration and DPM Components

The water condensation may cause absorption of SO₂ in the exhaust gas. Accordingly, we measured the relationship between the SO₂ concentration and the gas temperature. Fig. 7 shows the result. The SO₂ concentration was 29.5 mg/m³ at a gas temperature of 453 K, which decreased with decreasing gas temperature, to be eventually stabilized at 21.3 mg/m³ within a range from 313 K to 293 K. In terms of the SO₂ removal rate, the value was stabilized at approximately 28% within the range from 313 K to 293 K.

Fig. 8 shows the relationship between the concentrations of the DPM components and the gas temperature. The concentrations are expressed by the mass contained in 1 m³ of the exhaust gas (in terms of the volume at a temperature of 297 K). The SO₄²⁻ concentration was 5.5 mg/m³ at a gas temperature of 433 K and barely changed with decreasing temperature. The results described thus far suggest that cooling the exhaust gas causes the condensation of water and the absorption of SO₂ in the gas into the condensed water which is eventually discharged. However, one question remains: According to Fig. 7, the decrease in the SO₂ concentration is 29.5–21.3=8.2 mg/m³, which corresponds to 12.3 mg/m³ of the SO₄²⁻ concentration and is greater than the approximate value of 5.5 mg/m³ mentioned in section 4.1 as the amount of SO₄²⁻ found in the condensed water when the cooling temperature of the exhaust gas was 303 K. This discrepancy calls for further study in the future.

Fig. 7 Relationship between SO₂ Concentration and Gas Temperature

The SOF concentration was 3.3 mg/m³ at 433 K, which increased with decreasing gas temperature, reaching 14.4 mg/m³ at 303 K. This result demonstrates that SOF which is in the form of gas when the exhaust gas has a high temperature turns into particles as the exhaust gas is cooled. SOF in the form of gas cannot be trapped by the ESP in the subsequent stage; the transformation of SOF into particles facilitates its trapping. The particles can also be removed by the heat exchanger, as already explained in 4.1.

The Dry Soot concentration was 10.8 mg/m³ at 433 K and barely changed with decreasing gas temperature.

4.4 Effect of Collection by ESP

As described thus far, gaseous matters can be transformed into particles by cooling the exhaust gas. Those particles (DPM) must be eventually removed. Accordingly, we investigated the DPM collection capability of the ESP. Fig. 9 shows how the voltage applied to the precharger influences the particle-diameter characteristic of the collection efficiency in the ESP. The voltage applied to the collector was constantly maintained at DC +10 kV. The gas temperature on the downstream side of the heat exchanger was set at 293 K. The collection efficiency was calculated from the particle concentrations on the upstream and downstream sides of the ESP. The particle concentrations were measured with the scanning mobility particle sizer (SMPS). Regardless of the voltage applied to

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the precharger, the collection efficiency for particles of approximately 200 nm or smaller in diameter was nearly constant. By contrast, the collection efficiency for particles larger than 200 nm was slightly lower. A probable cause of this is the re-entrainment\textsuperscript{18}. The collection efficiency at –4.0 kV was no higher than 92 %, whereas it was improved to a maximum of 99 % when the voltage was from –7.0 to –8.0 kV. This result demonstrates that the particulate matters in the exhaust gas can be collected by the ESP.

Fig. 10 shows the influence of the gas temperature on the collection efficiency in the entire system including the heat exchanger and the ESP. The voltages applied to the precharger and the collector were DC –8 kV and DC +10 kV, respectively. The collection efficiency was calculated from the particle concentrations on the upstream side of the heat exchanger and the downstream side of the ESP. The particle concentrations were measured with the SMPS. When the gas temperature was 293 K, the collection efficiency for the particle diameter of 30 nm was slightly lower than the values obtained for the other diameters, which were approximately 90 %. At the gas temperatures from 393 to 313 K, the collection efficiency ranged from 81 to 93 %, which was shifted downward to 70-90 % at 293 K. This is due to the increase in the particle concentration which resulted from the water condensation as the exhaust gas was cooled. In any case, the collection efficiency was not less than approximately 80 %. Thus, high levels of collection efficiency were achieved.

5. Conclusion

With the aim of removing SO\textsubscript{2} and DPM in exhaust gas emitted from ships, we have studied the effects of the gas cooling coupled with the electrostatic collection of particulate matters, and obtained the following results:

(1) Cooling the exhaust gas has the effect of increasing the number concentration and mass concentration of DPM. This is due to the condensation of gaseous water and SOF.

(2) A 28 % reduction of SO\textsubscript{2} was achieved by cooling the exhaust gas. This is due to the absorption of SO\textsubscript{2} into the condensed water.

(3) The condensed water contains SO\textsubscript{4}\textsuperscript{2–} and SOF.

(4) High levels of collection efficiency reaching approximately 80 % or higher were achieved with the electrostatic precipitator.

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We are planning to further continue this research in various other aspects, such as the SO$_2$ removal capability of the ESP and its improvement.

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