Research Article

NO_x Removal from Simulated Marine Exhaust Gas by Wet Scrubbing Using NaClO Solution

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The experiments were performed in a lab-scale countercurrent spraying reactor to study the NO_x removal from simulated gas stream by cyclic scrubbing using NaClO solution. The effects of NaClO concentration, initial solution pH, coexisting gases (5% CO₂ and 13% O₂), NO_x concentration, SO₂ concentration, and absorbent temperature on NO_x removal efficiency were investigated in regard to marine exhaust gas. When NaClO concentration was higher than 0.05 M and initial solution pH was below 8, NO_x removal efficiency was relatively stable and it was higher than 60%. The coexisting CO₂ (5%) had little effect on NO_x removal efficiency, but the outlet CO₂ concentration decreased slowly with the initial pH increasing from 6 to 8. A complete removal of SO₂ and NO could be achieved simultaneously at 293 K, initial pH of 6, and NaClO concentration of 0.05 M, while the outlet NO₂ concentration increased slightly with the increase of inlet SO₂ concentration. NO_x removal efficiency increased slightly with the increase of absorbent temperature. The relevant reaction mechanisms for the oxidation and absorption of NO with NaClO were also discussed. The results indicated that it was of great potential for NO_x removal from marine exhaust gas by wet scrubbing using NaClO solution.

1. Introduction

The exhaust gas emitted from marine diesel engines contains a large number of atmospheric pollutants, such as sulphur oxides (SO_x), nitrogen oxides (NO_x), and particulate matters (PMs), which has caused serious damage to the ecological environment [1]. A direct way to reduce SO_x emission is to adopt low sulphur fuel oil (LSFO), but this will increase the transportation cost greatly. An alternative way is to install an exhaust gas cleaning system (EGCS) on board to achieve the abatement of SO_x emission equivalently. But it is very difficult for SO_x scrubbers to remove NO_x effectively at the same time. Generally, the methods for reducing marine NO_x emission can be divided into combustion control and postcombustion control techniques. The combustion control techniques include exhaust gas recirculation (EGR) [2], fuel-water emulsion (FWE) [3], and direct water injection (DWI) [4]. They aim at reducing the formation of NO_x during the combustion process, but it will result in the decrease of total heat efficiency. At present, the typical postcombustion control

approach for ocean-going ships is selective catalytic reduction (SCR), which can remove NO_x with an efficiency of 80–95%. As a relatively mature denitrification technology, SCR has been extensively applied in power plants and mobile vehicles. But there are still some problems that limit the application of SCR in marine industry. It requires large additional space and high investment/operation cost. The ash content or sulfate salts may result in the inactivation of SCR catalyst. A complex control system is also required to reduce the ammonia slip [5–7]. Therefore, ship owners around the world are still seeking a better way to reduce NO_x emission. It is of great importance to develop an efficient denitrification technology that can cater for the needs of ocean-going ships.

During the past decades, wet scrubbing technique becomes more and more attractive for the simultaneous removal of SO_x, NO_x, and other pollutants. As to NO_x removal, a preoxidation process is required to oxidize NO into NO₂ or other nitrogen oxides of higher values. That is due to the low solubility ($1.93 \times 10^{-3} \text{ mol} \cdot \text{L}^{-1} \cdot \text{atm}^{-1}$ at 25°C) of NO in water. The nonthermal plasma (NTP) can be used

to oxidize NO effectively, and then an absorption process is followed for NO₂ absorption. However, this method requires a high energy consumption [8, 9]. Similarly, ozone oxidation method encounters the same limitation [10–12]. Another feasible approach is to add the oxidants into the absorbent. For this purpose, various oxidants such as hydrogen peroxide (H₂O₂) [13–15], potassium permanganate (KMnO₄) [16], sodium chlorite (NaClO₂) [17, 18], calcium hypochlorite (Ca(ClO)₂) [19], and sodium hypochlorite (NaClO) [20–24] have been investigated to enhance the NO removal efficiency of the scrubbing solution. Compared with other oxidants, NaClO has some distinct advantages, such as low cost, easy availability, strong oxidative ability, easy to storage, good stability, and low toxicity. So it is attractive for researchers to investigate the simultaneous removal of NO and SO₂ by wet scrubbing using NaClO solution [25–28]. A previous study implied that NO could be effectively removed by wet scrubbing using NaClO solution in a cyclic mode, and the utilization of NaClO oxidant in solution was extremely high [29]. But the effect of the operating parameters (such as NaClO concentration, solution pH, absorbent temperature, NO, and SO₂ concentrations) on NO removal efficiency by cyclic scrubbing using NaClO solution had not been investigated. In this paper, marine exhaust gas was chosen as the treatment objective. Though the components of marine exhaust gas might vary with the engine load and fuel type, the typical compositions of marine exhaust gas contained ~13% O₂ and ~5% CO₂. The effect of coexisting gases on NO_x removal efficiency had also been studied preliminarily, and the relevant reaction mechanism was discussed.

2. Experimental Section

2.1. Materials. As mentioned in [11], exhaust gas scrubbers are designed in accordance with the maximum power and exhaust amount of target engine for the practical application in marine industry. The exhaust gas compositions are also measured at maximum load of engine. In this study, the concern is mainly focused on the NO_x removal efficiency, and the concentrations of gas components of a typical marine slow-speed 2-stroke diesel engine are considered as the reference. The fuel type is heavy fuel oil with 2.4% sulphur content. Thus, the concentrations of O₂, CO₂, SO₂, and NO_x are ~13%, ~5%, ~600 ppm, and ~1000 ppm, respectively. Here the simulated exhaust gas was prepared by blending various kinds of synthetic gases. Five kinds of gases, N₂ (99.999%), O₂ (99.995%), CO₂ (99.999%), NO (10.04% NO with N₂ as the balance gas), and SO₂ (10.1% SO₂ with N₂ as the balance gas) (Dalian Date Gas Co., Ltd), were used to make the simulated flue gas. As NO accounted for more than 95% of NO_x in marine exhaust gas, only NO span gas was used to prepare the NO_x components in simulated gas stream.

The NaClO solutions were prepared using the commercial NaClO solution (5% available chlorine, Shanghai Aladdin Bio-Chem Technology Co., Ltd) and the deionized water. The volume of NaClO solution was 1 L for each test. The pH value was adjusted by adding 0.5 M H₂SO₄ solution and determined using an acidimeter (Mettler-Toledo International Trading Co., Ltd).

FIGURE 1: Schematic diagram of the experimental setup.

2.2. Experimental Apparatus. A schematic diagram of the experimental apparatus is shown in Figure 1. It consists of a gas distributing system, a gas-liquid countercurrent scrubbing reactor, and a gas analyzer.

N₂, O₂, CO₂, NO, and SO₂ were provided from separate air bottles and metered through mass flow controllers (MFC, Beijing Sevenstar Electronics Co., Ltd). The simulated flue gas was obtained from the feed gases by blending with an on-line mixer, and then it was introduced into the spraying column. The height and inner diameter of the column were 25 cm and 5 cm, respectively. A spraying nozzle (BI/4TT-SS+TG-SS0.4, Spraying System Co., Ltd) was located at the top of the column. The size of liquid droplet sprayed from the nozzle was in the range of 80–100 μm. The flow rate of the simulated flue gas was fixed at 1.25 L/min. The calculated residence time of flue gas in the column was ~23 s.

When the initial gas concentrations were adjusted to the required level, the simulated flue gas was introduced into the scrubber from the bottom of the column. The NaClO absorbent was sprayed from top to bottom. A peristaltic pump was used to pump the scrubbing solution cyclically. The flow rate of the scrubbing solution was ~0.27 L/min. Each run of the test was 20 min. The outlet concentrations of flue gas were measured at an interval of 10 s. The solution temperatures were adjusted by the constant water bath (F34-MA, Julabo Labortechnik GmbH) and measured with a mercury thermometer. A MRU MGA-5 gas analyzer was used to determinate the gas concentrations of O₂, CO₂, NO, and NO₂ in flue gas.

2.3. Data Process. The gas concentrations measured by the bypass are taken as the inlet concentrations. The average concentrations within 20 min measured by the gas outlet are considered as the outlet concentrations. The removal efficiencies of NO_x and SO₂ are calculated by the following equation:

$$\eta = \frac{C_{\text{in}} - C_{\text{out}}}{C_{\text{in}}} \times 100\% \quad (1)$$

in which η is the removal efficiency and C_{in} and C_{out} are the inlet and outlet concentrations, respectively. Here NO_x refers to the mixture of NO and NO₂ in flue gas.

FIGURE 2: Effect of initial solution pH on NO_x removal efficiency (gas flow is 1.25 L/min, inlet NO_x concentration is 1000 ppm, NaClO concentration is 0.0125–0.1 M, and solution temperature is 293 K).

3. Results and Discussion

3.1. Effect of Initial pH and NaClO Concentration. The active components in NaClO solution were available chlorine, which mainly includes HClO, ClO^- , and Cl_2 . The compositions of NaClO solution depended greatly on the solution pH. Firstly, it was necessary to investigate the effect of initial solution pH on the NO_x removal efficiency. Figure 2 showed that NO_x removal efficiency was very low when the solution pH was higher than 10. That was because little HClO existed in the solution when solution pH was higher than 10, but HClO was considered to be the main component in NaClO solution to oxidize NO [22]. It demonstrated that NaClO solution without optimizing the initial pH was not suitable for removing NO_x . With the decrease of initial pH from 10 to 8, NO_x removal efficiency increased quickly, which was due to the increase of fractional composition of HClO in solution. HClO oxidized NO into NO_2 , N_2O_3 , N_2O_4 , and NO_3^- in chain reactions of (2)–(9) [30, 31]. The possible reaction pathways were summarized as shown in Figure 3.

FIGURE 3: Reaction pathways for NO_x removal by NaClO solution.

As shown in Figure 2, with the initial pH decreasing from 8 to 7, a slight drop of NO_x removal efficiency appeared. On further reducing the initial pH, the changes of NO_x removal efficiency depended on NaClO concentration. When initial NaClO concentration was below 0.05 M, NO_x removal efficiency continued to decrease with initial pH decreasing from 7 to 6. This was because the oxidation power of ClO^- was stronger than that of HClO in neutral or weak acidic medium. As shown in Figure 4, HClO concentration increased while ClO^- concentration decreased with solution pH decreasing from 7 to 6. However, when NaClO concentration was higher than 0.05 M, the oxidation power of active chlorine was high enough to oxidize NO effectively at pH 6 and 7 [32]. At the moment, the effect of the change of active chlorine concentration was not obvious enough. The result indicated that a high NaClO concentration might be appropriate for practical application for it was easy to obtain a high and stable NO_x removal efficiency in a relatively wide range of solution pH.

Figure 5 showed the effect of initial solution pH on NO_x concentration in flue gas. When NaClO concentration was higher than 0.05 M and solution pH was in the range of 4–8, outlet NO concentration was lower than 124 ppm. It suggested that the majority of NO had been oxidized into NO_2 by NaClO. When NaClO concentration was 0.05 M, outlet NO concentration decreased sharply to 35 ppm with initial pH decreasing from 10 to 8. At the same time, NO_2 concentration in outlet gas increased from 14 ppm to 353 ppm. On further decreasing the initial pH down to 6, a complete removal of NO could be achieved with 0.05 M NaClO while outlet NO_2 concentration reached about 400 ppm. With the initial pH decreasing from 6 to 4, NO in outlet gas increased to 124 ppm while NO_2 in outlet gas decreased to 270 ppm. NO_x removal efficiency changed little in the range of pH 4–8. The results implied that, to a certain extent, NO_x removal efficiency for wet scrubbing using NaClO solution was mainly limited to the absorption of NO_2 rather than the oxidation of NO.

3.2. Effect of Coexisting CO_2 . During wet scrubbing process, CO_2 in flue gas would react with absorbent thus influencing

FIGURE 4: Equilibrium concentrations of active chlorine species in NaClO solution as a function of solution pH.

the oxidation and absorption of targeted pollutants. As an acidic oxide, CO_2 was sensitive to the solution pH. The effect of coexisting CO_2 on NO_x removal efficiency was investigated, and results were shown in Figure 6. It can be seen that, with initial solution pH increasing from 4 to 8, NO_x removal efficiency was relatively stable. A complete removal of NO had been achieved and outlet NO_2 concentration was ~ 350 ppm. The coexistence of CO_2 had not affected the NO_x removal efficiency so much. But the average CO_2 concentration in outlet gas changed obviously with initial solution pH. When initial solution pH was below 6, the average CO_2 concentration kept stable at 5%. However, it began to decrease quickly with initial pH increasing from 6 to 8.

The change of outlet CO_2 concentration during the cyclic scrubbing process was shown in Figure 7. When initial pH was in the range of 4–6, CO_2 concentration decreased to 4.4–4.5% at the start of the scrubbing process. Then it recovered to the initial level due to the hydrolysis equilibrium between CO_2 and absorbent solution. The hydrolysis reactions of CO_2 were described in (10) and (11). Furthermore, a certain amount of H^+ might be produced during the hydrolysis process, which would affect the chlorine hydrolysis equilibrium reactions as shown in (12) and (13). Since HCO_3^- and CO_3^{2-} had buffering ability to some extent, the hydrolysis of CO_2 would not influence the solution pH obviously when initial pH was in the range of 4–6. But with initial pH increasing from 6 to 8, the absorption of CO_2 might reduce the solution alkalinity, resulting in the decrease of the solution pH. Thus it was necessary to keep the solution pH at 6 in order to reduce the consumption of solution alkalinity when NaClO solution was adopted to remove NO_x from flue gas.

When initial pH was in the range of 7–8, CO_2 concentration decreased largely at the very beginning of the cyclic scrubbing process. It suggested that much more CO_2 had been absorbed by the scrubbing solution due to the weak alkaline medium. With the proceeding of the scrubbing process, CO_2 concentration began to increase slowly. Although no evidence showed that CO_2 would react with NaClO directly, the hydrolyzation and absorption of CO_2 would increase the consumption of solution alkalinity obviously. It meant that extra alkaline solution was required to maintain the solution pH during cyclic scrubbing process, which would largely increase the operation complexity and cost at the same time. Thus initial pH of 6 might be appropriate for practical application.

Figure 8 showed the change of NaClO solution pH after scrubbing for 20 min. With the proceeding of cyclic scrubbing process, the solution pH would decrease slowly due to the absorption of NO_x and CO_2 . It was worth noting that the solution pH increased from 4 to 4.63 for NaClO solution with initial pH 4. That was because active chlorine species in NaClO solution were mainly HClO and Cl_2 at pH 4, as shown in Figure 4. During the scrubbing process, Cl_2 would be purged out easily from the solution and reacted with NO effectively in gas phase as (14) and (15) [25]. The decrease of Cl_2 in NaClO solution would lead to a left shift of the hydrolysis equilibrium of active chlorine as shown in (16), resulting in the increase of solution pH for NaClO solution with initial pH 4.

It was possible that excessive Cl_2 escaped from NaClO solution might result in secondary pollution. In addition, acidic mist formed in the flue gas might result in the corrosion of operation system. In view of this, NaClO solution with pH 6 was appropriate for NO_x removal in cyclic scrubbing mode.

3.3. Effect of Coexisting O_2 . For marine diesel engines, there was typical $\sim 13\%$ O_2 in exhaust gas. O_2 could partially oxidize NO under certain conditions, so it was necessary to investigate the effect of coexisting O_2 on NO_x removal efficiency. In the experiments, only NO standard gas was used to prepare NO_x in the initial simulated flue gas. The introduction of O_2 has oxidized a little of NO into NO_2 in the gas mixer. As shown in Figure 9, the initial NO_2 concentration in inlet gas increased almost linearly with the increase of NO concentration. When inlet NO_x concentration was 1000 ppm, the initial NO_2 concentration reached 112 ppm.

With the NO_x increasing from 250 to 700 ppm, the outlet NO concentration decreased gradually to 0. When inlet

FIGURE 5: The change of (a) NO concentrations and (b) NO₂ concentrations in flue gas with initial pH of NaClO solution (gas flow is 1.25 L/min, inlet NO_x concentration is 1000 ppm, NaClO concentration is in the range of 0.0125–0.1 M, and solution temperature is 293 K; solid points refer to inlet NO_x concentrations and open points refer to outlet NO_x concentrations).

FIGURE 6: The change of NO_x removal efficiency and averaged CO₂ concentration in flue gas with initial pH (gas flow is 1.25 L/min, inlet NO_x concentration is 1000 ppm, initial CO₂ concentration is 5%, NaClO concentration is 0.05 M, and solution temperature is 293 K).

NO_x concentration was higher than 700 ppm, NO could be removed completely. As expected, the outlet NO₂ concentration increased almost linearly with the increase of inlet NO_x concentration. It indicated that NO_x removal efficiency depended to a great extent on the absorption of NO₂ during the scrubbing process.

Figure 10 presented the change of NO_x removal efficiency and outlet O₂ concentration with the initial NO_x

FIGURE 7: The change of CO₂ concentration during the cyclic scrubbing duration (gas flow is 1.25 L/min, inlet NO concentration is 1000 ppm, initial CO₂ concentration is 5%, NaClO concentration is 0.05 M, and solution temperature is 293 K).

concentration. Since O₂ concentration was relatively high, it changed little during the scrubbing process. With the increase of NO_x concentration from 250 ppm to 700 ppm, NO_x removal efficiency increased from 47% to 74%. It could

FIGURE 8: NaClO solution pH after scrubbing for 20 min (gas flow is 1.25 L/min, inlet NO_x concentration is 1000 ppm, initial CO_2 concentration is 5%, NaClO concentration is 0.05 M, and solution temperature is 293 K).

FIGURE 9: The change of NO_x concentration with the initial NO_x concentration (gas flow is 1.25 L/min, inlet O_2 concentration is 13%, inlet NO_x concentration is in the range of 250–1000 ppm, NaClO concentration is 0.05 M, initial solution pH is 6, and solution temperature is 293 K; solid points refer to inlet NO_x concentrations and open points refer to outlet NO_x concentrations).

be ascribed to the improvement of the mass transfer at the gas-liquid interface. When O_2 was present in flue gas, NO_x removal efficiency was a little higher than that without O_2 in flue gas. The existence of O_2 improved the NO_x removal efficiency in way of partly oxidizing NO in initial flue gas.

3.4. Effect of NO_x Concentration. The effects of NO_x concentration on NO_x removal are investigated, and the results are shown in Figure 11. It can be seen that, with initial NO

FIGURE 10: The change of NO_x removal efficiency and outlet O_2 concentration with the initial NO_x concentration (conditions: gas flow: 1.25 L/min; inlet O_2 concentration: 13%; inlet NO_x concentration: 250–1000 ppm; NaClO concentration: 0.05 M; initial solution pH: 6; solution temperature: 293 K).

concentration increasing from 250 ppm to 500 ppm, NO_x removal efficiency increased from 43% to ~63%. NO could be removed completely when inlet NO concentration was higher than 500 ppm. Outlet NO_2 concentration increased quickly with the increase of inlet NO concentration, resulting in a relatively stable NO_x removal efficiency.

3.5. Simultaneous Removal of NO_x and SO_2 . Marine diesel engines usually burn heavy fuel oil (HFO) in order to save the operating cost. At present, the mass concentration of sulphur (S) content in HFO was about 2.5% at average. The combustion of HFO fuel would produce a large amount of SO_2 in exhaust gas. Experiments were conducted to investigate the effect of coexisting SO_2 on NO_x removal efficiency, and the results are shown in Figures 12 and 13.

Figure 12 depicted the change of NO_x removal efficiency and NO_x concentration with inlet NO_x concentration. A complete removal of SO_2 and NO had been achieved simultaneously. Due to its high solubility, SO_2 could be absorbed effectively by scrubbing solution. Then SO_2 was removed through the hydrolysis reaction as (18) and (19). The hydrolysis products of SO_3^{2-} would be oxidized by active chlorine species into SO_4^{2-} quickly. Thus the removal of SO_2 would consume the solution alkalinity and oxidants at the same time. The result demonstrated that NaClO solution could be used to remove NO and SO_2 simultaneously from marine exhaust gas. However, outlet NO_2 concentration increased gradually with the increase of inlet NO_x concentration.

FIGURE 11: The change of (a) NO_x removal efficiency and (b) NO_x concentrations with the initial NO_x concentration (gas flow is 1.25 L/min, inlet NO_x concentration is 250–1250 ppm, NaClO concentration is 0.05 M, initial solution pH is 6, and solution temperature is 293 K; solid points refer to inlet NO_x concentrations and open points refer to outlet NO_x concentrations).

FIGURE 12: The change of (a) NO_x removal efficiency and (b) NO_x concentration with the inlet NO_x concentration (gas flow is 1.25 L/min, inlet SO₂ concentration is 600 ppm, inlet NO_x concentration is 250–1250 ppm, NaClO concentration is 0.05 M, initial solution pH is 6, and solution temperature is 293 K; solid points refer to inlet NO_x concentrations and open points refer to outlet NO_x concentrations).

Figure 13 exhibited the effect of SO₂ concentration on NO_x removal efficiency and NO_x concentration in flue gas. The result showed that SO₂ together with NO was completely removed. But with the increase of inlet SO₂ concentration, NO_x removal efficiency exhibited a slightly downside trend. This phenomenon could be ascribed to the competition reactions between NO_x and SO₂. Some oxidants in the absorbent would be consumed through the hydrolyzation

and absorption of SO₂. With inlet SO₂ concentration increasing from 200 ppm to 600 ppm, the pH of the scrubbed NaClO solution decreased from 5.25 to 4.52. The decrease of solution pH was negative for the absorption of NO₂ through (5) and (6).

3.6. Effect of Absorbent Temperature. The reaction temperature could greatly influence the diffusion, dissolution, and

FIGURE 13: The change of (a) NO_x removal efficiency and (b) NO_x concentration with the initial SO_2 concentration (gas flow is 1.25 L/min, inlet SO_2 concentration is 200–600 ppm, inlet NO_x concentration is 1000 ppm, NaClO concentration is 0.05 M, initial solution pH is 6, and solution temperature is 293 K; solid points refer to inlet NO_x concentrations and open points refer to outlet NO_x concentrations).

FIGURE 14: The change of (a) NO_x removal efficiency and (b) NO_x concentration with the absorbent temperature (gas flow is 1.25 L/min, inlet NO_x concentration is 1000 ppm, NaClO concentration is 0.05 M, initial solution pH is 6, and solution temperature is 293–333 K; solid points refer to inlet NO_x concentrations and open points refer to outlet NO_x concentrations).

reaction characteristics of molecules or ions in liquid phase [19]. The effect of absorbent temperature on NO_x removal was investigated, and the results were shown in Figure 14. Generally, the absorbent temperature was kept below 333 K in industrial application in order to reduce the supply of make-up water. So the absorbent temperature was chosen to be in the range of 293–333 K in the experiments. Figure 14 showed that NO_x removal efficiency increased gradually with

the increase of absorbent temperature. The highest NO_x removal efficiency of 69% was achieved at 333 K. As NO could be removed by 100% easily, the increase of NO_x removal efficiency resulted from the improvement of NO_2 absorption. This could be explained by the Arrhenius equation of the reaction rate constant. The increase of temperature could enhance the mass transfer of NO_2 to the absorbent, thus improving the absorption of NO_2 accordingly.

4. Conclusion

NO_x removal by wet scrubbing using NaClO solution was studied based on a spraying reactor in a cyclic mode. The results showed that when NaClO concentration was higher than 0.05 M and initial solution pH was below 8, NO_x removal efficiency was relatively stable, which was higher than 60%. The coexisting CO_2 (5%) had little effect on NO_x removal efficiency, but the solution pH began to decrease with the proceeding of cyclic scrubbing process when initial pH was higher than 6. The coexisting O_2 (13%) could oxidize NO partially in the gas mixer, resulting in a little improvement of NO_x removal efficiency. When initial NO_x concentration was higher than 500 ppm, NO could be removed completely while outlet NO_2 concentration increased almost linearly with the increase of initial NO_x concentration, resulting in a relative stable NO_x removal efficiency. A complete removal of SO_2 and NO could be achieved simultaneously at 293 K, initial NaClO solution pH 6, and 0.05 M NaClO concentration. With the increase of inlet SO_2 concentration, the outlet NO_2 concentration increased slightly due to the decrease of solution pH. NO_x removal efficiency increased with the increase of absorbent temperature. The relevant reaction mechanism for the removal of NO_x and SO_2 by wet scrubbing using NaClO solution was also discussed. The results demonstrated that it was of great potential for NaClO solution to remove NO_x from marine exhaust gas.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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