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RESEARCH ARTICLE

Investigation on the removal of NO from marine exhaust gas using the $Na_2S_2O_8$-urea redox system in seawater carrier

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Abstract The sodium persulfate ($Na_2S_2O_8$)-urea system has been proven to be an excellent scrubbing solution for the wet removal of NO. Commonly, seawater is used as a wet carrier in marine applications. To further explore the feasibility of marine denitrification using $Na_2S_2O_8$-urea system, this study proposed the $Na_2S_2O_8$-urea-seawater composite redox system for NO removal from the marine exhaust gas. The effects of seawater carrier, reaction temperature, $Na_2S_2O_8$ concentration, urea concentration, pH value, and NO concentration on NO removal were investigated. Additionally, the NO_3^- concentration in the solution was measured. Results showed that the lowest normalized NO concentration was 0.099, with the corresponding mass of NO absorbed per unit volume of solution reaching 0.108 $g \cdot L^{-1}$. The addition of seawater carrier and incremental reaction temperature, $Na_2S_2O_8$, and urea concentration promoted the NO removal performance. When the pH value increased within the range of 4–7, the NO removal performance decreased. The NO removal performance increased as the pH value further increased to 8, but decreased again when the pH value increased to 11. An increase in NO concentration was detrimental to NO removal. The Cl^-, HCO_3^-, and CO_3^{2-} in seawater could augment the total concentration of active free radicals to improve denitrification performance.

Keywords marine diesel exhaust gas, denitrification, wet scrubbing, redox, seawater carrier

1 Introduction

Marine transportation is widely regarded as a crucial mode of international trade transportation due to its large cargo capacity, high traffic efficiency, and low cost [1]. Nevertheless, marine diesel engines utilizing heavy fuel oil as the primary fuel emit significant amounts of nitrogen oxides (NO_x), seriously endangering human health and the ecological environment. In response, the International Maritime Organization (IMO) has formulated a series of related regulations for strictly limiting NO_x emissions from marine exhaust [2]. At present, numerous studies have explored NO_x emission control technologies for ships, and several of them have already been successfully applied.

Wet scrubbing technology is deemed one of the most promising technologies for controlling marine pollutant emissions due to its economic efficiency and simple operation [3]. The abundance and accessibility of seawater make it the commonly employed carrier of wet scrubbing solutions in practical marine applications. However, the main obstacle is that more than 90% of NO_x in marine exhaust comprises water-insoluble NO, resulting in low water solubility of NO_x and limiting the effectiveness of the wet scrubbing technology for NO_x removal [4]. Relevant studies have shown that seawater could convert less than 10% of NO in the flue gas [5,6]. Although hydrodynamic cavitation could effectively promote denitrification under seawater conditions, it still cannot meet the NO emission standards from marine exhaust gas [7,8]. Consequently, existing commercial marine wet scrubbers using seawater could effectively remove SO_2 but not NO. Additionally, converting NO into water-soluble NO_x is the key to achieving effective denitrification through wet scrubbing technology. Therefore, when seawater is used for denitrification, a strong oxidizing agent should be added to seawater.

In recent years, various potent oxidants, such as H_2O_2 [9,10], $KMnO_4$ [11,12], O_3 [13,14], ClO_2 [15,16], NaClO [17,18], $NaClO_2$ [19,20], $K_2S_2O_8$ [21,22], and their mixtures [23–25] have been used in wet scrubbing

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systems for NO removal from flue gas, and the results demonstrated their effectiveness in NO removal. The presence of Cl^-, CO_3^{2-}, and HCO_3^- ions in seawater can impact the NO removal efficiency to some extent [26]. Seawater also has a natural acid-base buffering capacity, which can effectively stabilize pH levels during the scrubbing to prevent sharp declines due to acid generation [27]. Han et al. [28] and Lu et al. [29] investigated the NO removal using the $NaClO_2$ seawater solution and found that seawater with $NaClO_2$ had a significantly enhanced breakthrough time than freshwater with $NaClO_2$. The buffering ability of seawater could effectively suppress the acidic decomposition of $NaClO_2$ into ClO_2 and improve the utilization of $NaClO_2$ in the solution. The Cl^-, HCO_3^- and CO_3^- ions in seawater could react with the oxidants to form multi-active inorganic free radicals (e.g., $\cdot Cl$, $\cdot CO_3^-$, and $\cdot Cl_2^-$), which might also play an important role in denitrification [30,31]. Some oxidants with high costs, high toxicity, and poor stability were unsuitable for NO removal from the marine exhaust due to the special environmental conditions and space constraints aboard ships. Sodium persulfate ($Na_2S_2O_8$), as an environmentally friendly potent oxidizing agent has the advantages of good stability, low cost, strong oxidation, non-toxicity, etc. $Na_2S_2O_8$ has been widely applied for soil remediation and the treatment of toxic organic gases [32]. For denitrification, Xi et al. [33] investigated the wet oxidative removal of NO using $Na_2S_2O_8$ in a bubble column reactor. The results showed that the 0.15 mol·L^{-1} $Na_2S_2O_8$ solution at 80 °C completely absorbed the NO in simulated exhaust gas from marine diesel engine. The final products were SO_4^{2-} and NO_3^-. The gas-phase resistance and liquid-phase resistance controlled the mass transfer process of NO absorption using the $Na_2S_2O_8$ solution. The increase in the gas-phase volume fraction could reduce the gas-phase resistance, and the increase in the concentration of oxidant, solution temperature, and pH value could reduce the liquid-phase resistance, thus improving the mass transfer performance [6]. It was inferred that $Na_2S_2O_8$ was a viable oxidant for removing NO from marine diesel exhaust using wet scrubbing, and the seawater carrier could promote the NO absorption in the $Na_2S_2O_8$-seawater solution.

Nevertheless, the oxidation process in wet scrubbing produces large amounts of NO_3^- as the final product. The excessive release of NO_3^- into the environment disrupts the nitrogen cycle, causing eutrophication, which can severely affect aquatic ecosystems [34]. IMO stipulates that the NO_3^- concentration in the washing waste liquid must be less than 60 mg·L^{-1} or the amount of NO_3^- corresponding to the removal of 12% NO_x from the exhaust gas. Urea is a cost-effective and strong alkaline reducing agent, which is frequently used for removing SO_2 and NO from marine exhaust [35]. However, the studies on wet denitrification using urea are still in the exploratory stage, and the addition of oxidants could improve the denitrification efficiency of urea [36]. $KMnO_4$ [37,38], H_2O_2 [39,40], NaClO [37] and $NaClO_2$ [41] are the commonly used oxidants, with $KMnO_4$ having the best denitrification performance. The denitrification efficiency could reach 96.3% under the conditions of $KMnO_4$ concentration of 5 mmol·L^{-1}, reaction temperature of 70 °C, and urea concentration of 9% [37]. Meanwhile, NO_x was found to convert primarily to N_2, significantly reducing the concentration of NO_3^- and NO_2^- in the solution [42]. It was inferred that urea as a reducing agent could reduce high-valence nitrogen to N_2, effectively inhibiting NO_3^- formation [43]. Xi et al. [44] studied NO removal from marine exhaust using $Na_2S_2O_8$ as the oxidant in the $Na_2S_2O_8$-urea solution. The scrubbing solution with 0.1 mol·L^{-1} $Na_2S_2O_8$ and 4.0 mol·L^{-1} urea had a NO_3^- concentration of 14.93 mg·L^{-1} at 80 °C, while the scrubbing solution without urea had a NO_3^- concentration of 194.54 mg·L^{-1}. When synergizing SO_2 and NO, the NO_3^- concentration was 8.56 mg·L^{-1} at 70 °C with 0.2 mol·L^{-1} $Na_2S_2O_8$ and 2 mol·L^{-1} urea [45]. The addition of urea effectively reduced the NO_3^- emission in the wet scrubbing system. The $Na_2S_2O_8$-urea solution system showed excellent application prospects for NO removal from marine exhaust. However, the application and popularization of $Na_2S_2O_8$-urea system on ships are hindered since the denitrification performance of the $Na_2S_2O_8$-urea system in seawater remains underexplored, and the specific denitrification mechanisms are not yet fully understood. Therefore, studying the denitrification process of $Na_2S_2O_8$-urea redox system in seawater is imperative.

In this study, the $Na_2S_2O_8$-urea-seawater composite redox system was utilized to remove NO from marine diesel engine exhaust in a bubbling reactor. Thereinto, seawater, $Na_2S_2O_8$, and urea served as the solution carrier, oxidant, and reducing agent, respectively. The effects of various parameters, such as seawater carrier, reaction temperature, $Na_2S_2O_8$ concentration, urea concentration, initial NO concentration, and solution pH, on NO removal performance were systematically investigated. Additionally, for the $Na_2S_2O_8$-urea-seawater composite redox system, the corresponding NO_3^- concentration after the reaction was measured, and the denitrification mechanism was elucidated. Consequently, this study could provide further theoretical and technical support for the potential engineering application of $Na_2S_2O_8$-urea-seawater composite redox system.

2 Experimental

2.1 Experimental materials

The standard gases were purchased from Shandong

Deyang Special Gas Co., Ltd., China, which included 99.999% N_2 and 1% NO (with N_2 as the balance gas). $Na_2S_2O_8$ was supplied by China Aladdin Industrial Corporation. Urea and magnesium chloride hexahydrate ($MgCl_2 \cdot 6H_2O$) were provided by Shanghai Macklin Biochemical CO., Ltd., China, KCl was obtained from Tianjin Kermel Chemical Reagent Co., Ltd., China, $CaCl_2$, $NaHCO_3$, NaCl, and Na_2SO_4 were purchased from Sinopharm Chemical Reagent Co., Ltd., HCl and NaOH used to adjust the pH of the solution were purchased from Sinopharm Chemical Reagent Co., Ltd., China. The primary components of the simulated seawater prepared according to the American ASTM1141 standard are shown in Table 1 [46].

2.2 Experimental system

The experimental system for NO removal using the $Na_2S_2O_8$-urea-seawater composite redox system is depicted in Fig. 1. The experimental system mainly included three parts: the simulated gas distribution system, the bubbling reactor absorption system, and the flue gas analysis and tail gas treatment system. The simulated gas distribution system included gas cylinders, mass flow meters, pressure-reducing valves, and gas mixers. The simulated mixed gas consisted of N_2 and NO, which were supplied by gas cylinders. The total flow rate was maintained at 1.0 $L \cdot min^{-1}$, and the gas flow was accurately controlled using mass flow meters. Gas components were mixed in the gas mixer according to a predetermined ratio. The bubbling reactor absorption system primarily encompassed the plexiglass bubbling reactor and the constant temperature water bath. The inner diameter and height of the bubbling reactor were 60 and 500 mm, respectively. The gas distributor of the bubbling reactor facilitated the uniform distribution of mixed gas flow, and its diameter, height, and diameter of the inner hole were 20 mm, 30 mm, and 10 μm, respectively. The upper end of the bubbling reactor was equipped with a feeding hole, a mixed gas inlet, and a mixed gas outlet. The solids and simulated seawater could be introduced into the reactor through the feeding hole, from which the bubbling solution could be taken out. The temperature of bubbling solution and reactor is regulated by the constant temperature water bath. The flue gas analysis and tail gas treatment system included a gas dryer, gas analyzer (ECOM-EN2, ECOM GmbH, Germany), and tail gas treatment device. The gas analyzer was used to measure the NO concentration of the reaction system. To prevent corrosion or damage to the gas analyzer, granular anhydrous $CaCl_2$ was utilized to dry the mixed gas before entering the gas analyzer. The exhaust gas was emptied after being treated with $KMnO_4$/NaOH solution and activated carbon. The NO_3^- concentration in the liquid phase was determined by ion chromatography (ICS900, Dionex Corporation, USA). The pH value of the solution was measured by the pH meter (PHS-3E, Shanghai Yoke Instrument Co., Ltd., China).

Table 1 The concentrations of the primary components in simulated seawater according to the American ASTM 1141 standard

Component	Concentration/($g \cdot L^{-1}$)
NaCl	24.530
Na_2SO_4	4.090
KCl	0.695
$MgCl_2 \cdot 6H_2O$	11.110
$CaCl_2$	1.160
$NaHCO_3$	0.201

2.3 Experimental method

For the experiments, 1.0 L of simulated seawater was precisely measured and poured into the bubbling reactor. Subsequently, the bubbling reactor was placed in the constant temperature water bath to preheat the mixed

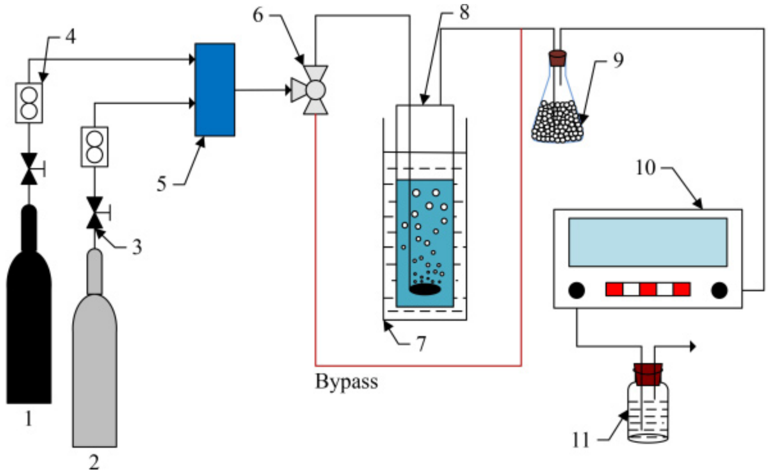

Fig. 1 Schematic diagram of the experimental system (1: Nitrogen gas cylinder; 2: NO gas cylinder; 3: pressure-reducing valves; 4: mass flow controllers; 5: gas mixer; 6: three-way valve; 7: constant temperature water bath; 8: bubbling reactor; 9: gas drier; 10: gas analyzer; 11: tail gas absorber).

solution to the designated temperature. The gas flow was switched to the bypass before each experiment to obtain the initial NO concentration. When the NO concentration reached the target value and remained stable, the initial NO concentration was measured using the gas analyzer. The required amounts of $Na_2S_2O_8$ and urea solids were weighed, then injected into the bubbling reactor through the feeding hole, and dissolved completely in 1.0 L of simulated seawater. Subsequently, the pH value of the solution was adjusted to the required value. The mixed gas was then switched to the main route and entered the bubbling reactor by turning the valve. The reaction time was immediately recorded. After passing through the gas dryer, the mixed gas was analyzed using the gas analyzer. The gas analyzer readings were recorded every 5 min to monitor the outlet NO concentration. Each experiment lasted 90 min, and the total gas flow rate was maintained at 1.0 $L \cdot min^{-1}$. The same experiment was repeated three times, and the obtained data was averaged before further analysis and calculation.

The normalized NO concentration, defined as the ratio of the outlet NO concentration to the inlet NO concentration, is expressed as shown in Eq. (1):

$$\text{Normalized NO concentration} = \frac{C_{\text{out}}}{C_{\text{in}}}, \tag{1}$$

where C_{in} and C_{out} represent the inlet and outlet concentration (ppm) of NO, respectively.

The mass of NO absorbed per unit volume of scrubbing solution could be calculated by Eq. (2):

$$q = \int_0^t 30 \times Q \times (-C_{\text{out}}) / (22.4 \times V)\,dt, \tag{2}$$

where Q represents the total gas flow rate ($mL \cdot min^{-1}$); V represents the volume (L) of scrubbing liquid, t represents the reaction time (min).

3 Results and discussion

3.1 Effect of seawater carrier

Since seawater was generally used as a wet carrier in marine applications, the influence of seawater carrier on NO removal from marine exhaust gas using the $Na_2S_2O_8$-urea composite redox system was investigated and the results were illustrated in Fig. 2. The NO concentration initially decreased and eventually stabilized as the experiment progressed. The normalized NO concentration of $Na_2S_2O_8$-urea-seawater composite redox system was 0.154, which was lower than that of $Na_2S_2O_8$-urea-deionized water solution (0.197). This indicated that the seawater carrier significantly enhanced the NO removal performance of the $Na_2S_2O_8$-urea composite system.

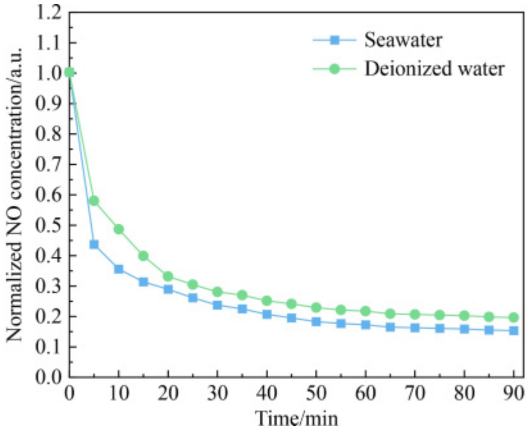

Fig. 2 Variation of NO concentration with time under different solution carriers during NO removal using the $Na_2S_2O_8$-urea-seawater composite redox system (experimental conditions: reaction temperature = 70 °C, $Na_2S_2O_8$ concentration = 0.1 $mol \cdot L^{-1}$, urea concentration = 0.5 $mol \cdot L^{-1}$, NO concentration = 1000 ppm, pH = 7).

$Na_2S_2O_8$ could generate the sulfate radical ($\cdot SO_4^-$), and some of the $\cdot SO_4^-$ were converted to the hydroxyl radical ($\cdot OH$) in water through reactions (3) and (4). The Cl^-, HCO_3^-, and CO_3^{2-} ions in seawater increased the content of active free radicals in the $Na_2S_2O_8$-urea-seawater composite redox system, strengthening the NO removal [30]. Moreover, the alkaline buffer capacity of seawater could promote the treatment of the acidic gas NO_x. Therefore, the seawater carrier could increase the active free radical concentration in the $N_2S_2O_8$-urea-seawater composite redox system and provide an alkaline buffer system to promote NO removal.

$$S_2O_8^{2-} \rightarrow 2 \cdot SO_4^- \tag{3}$$

$$\cdot SO_4^- + H_2O \rightarrow SO_4^{2-} + \cdot OH + H^+ \tag{4}$$

3.2 Effect of reaction temperature

The variation of NO concentration with time under different temperatures and the mass of NO absorbed per unit volume of $Na_2S_2O_8$-urea-seawater composite redox system were illustrated in Fig. 3. The reaction temperature significantly impacted NO removal using the $Na_2S_2O_8$-urea-seawater composite redox system. The normalized NO concentration decreased from 0.867 to 0.143 as the reaction temperature rose from 20 to 70 °C. The equilibrium NO concentration gradually decreased with the increase in the reaction temperature. Correspondingly, as shown in Fig. 3(b), the mass of NO absorbed per unit volume of solution increased from 0.012 to 0.095 $g \cdot L^{-1}$ when the reaction temperature increased from 20 to 70 °C. This indicated that higher reaction temperature favored the NO removal using the $Na_2S_2O_8$-urea-seawater composite redox system.

The activation degree of $Na_2S_2O_8$ was limited when the reaction temperature was 20 °C, and $S_2O_8^{2-}$ was the primary oxidizing substance. The oxidizing ability of

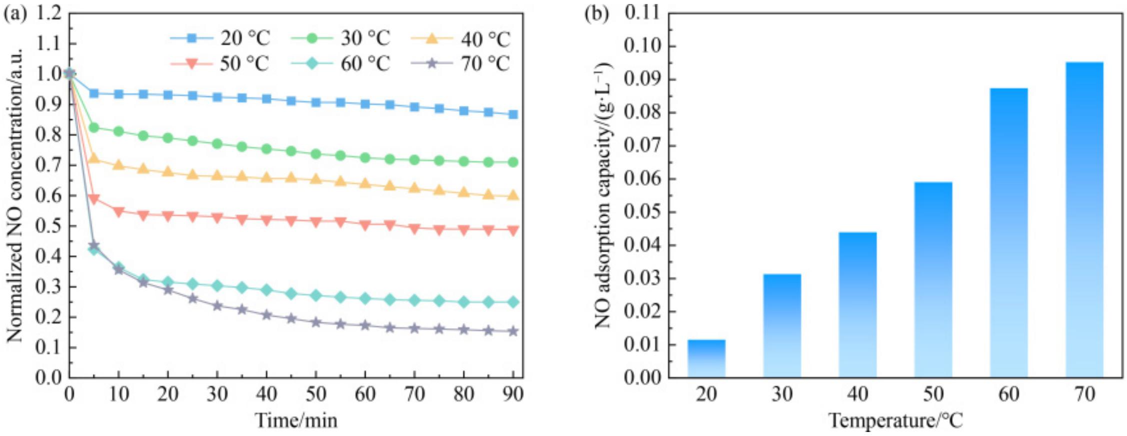

Fig. 3 (a) Variation of NO concentration with time during NO removal using the $Na_2S_2O_8$-urea-seawater composite redox system, and (b) the corresponding mass of NO absorbed per unit volume of $Na_2S_2O_8$-urea-seawater composite redox system under different temperatures (experimental conditions: reaction temperature = 20–70 °C, $Na_2S_2O_8$ concentration = 0.1 mol·L^{-1}, urea concentration = 0.5 mol·L^{-1}, NO concentration = 1000 ppm, pH = 7).

$S_2O_8^{2-}$ was weak, and the NO removal was primarily through reactions (5–7). However, reaction (7) could release NO again, leading to poor NO removal performance. As the temperature increased, the reaction rate of reaction (3) accelerated and the activation degree of $Na_2S_2O_8$ was gradually enhanced [6]. It led to a gradual increase in the concentration of active free radicals in the solution. As a result, the oxidation ability of the scrubbing solution increased, which strengthened the oxidation processes and achieved higher NO removal performance. The thermal activation of $Na_2S_2O_8$ usually occurred at the temperature greater than 60 °C [33,44,45]. Consequently, the NO removal performance was significantly enhanced when the reaction temperature was increased to 60 °C, which was consistent with Fig. 3. Additionally, urea was hydrolyzed in an aqueous solution via reaction (8), which produced ammonium carbamate. The reaction rate of reaction (8) could increase with the increase in the reaction temperature. Compared to urea, ammonium carbamate was more likely to react with NO_2^- through reaction (9) to promote NO removal [45]. Hence, the NO removal performance of $Na_2S_2O_8$-urea-seawater composite redox system from marine exhaust gas was improved as the reaction temperature increased.

$$S_2O_8^{2-} + NO + H_2O \rightarrow 2SO_4^{2-} + 2H^+ + NO_2 \tag{5}$$

$$2NO_2 + H_2O \rightarrow HNO_2 + HNO_3 \tag{6}$$

$$3HNO_2 \rightarrow 2NO + HNO_3 + H_2O \tag{7}$$

$$(NH_2)_2CO + H_2O \rightarrow NH_2COONH_4 \tag{8}$$

$$NH_2COONH_4 + 2NO_2^- + 2H^+ \rightarrow 2N_2 + CO_2 + 4H_2O \tag{9}$$

3.3 Effect of $Na_2S_2O_8$ concentration

The $Na_2S_2O_8$ concentration was an important influencing factor for NO removal using $Na_2S_2O_8$-urea-seawater composite redox system. The variation of NO concentration with time during NO removal under different $Na_2S_2O_8$ concentrations was shown in Fig. 4(a). When the $Na_2S_2O_8$ concentration increased from 0 to 0.1 mol·L^{-1}, the normalized NO concentration decreased from 0.974 to 0.154, indicating the continuous decrease in the NO equilibrium concentration. However, when $Na_2S_2O_8$ concentration continued to increase to 0.25 mol·L^{-1}, further increases in $Na_2S_2O_8$ concentration had slight effects, with the normalized NO concentration slightly decreasing to 0.142. Meanwhile, the mass of NO absorbed per unit volume of $Na_2S_2O_8$-urea-seawater composite redox system under different $Na_2S_2O_8$ concentrations was shown in Fig. 4(b). The mass of NO absorbed per unit volume of solution increased from 0.007 to 0.095 g·L^{-1} as the $Na_2S_2O_8$ concentration increased from 0 to 0.1 mol·L^{-1}. Subsequently, the mass of NO absorbed increased marginally to 0.101 g·L^{-1} as the $Na_2S_2O_8$ concentration reached 0.25 mol·L^{-1}. It indicated that the NO removal performance initially increased and then remained stable with the increase in the $Na_2S_2O_8$ concentration. The optimal $Na_2S_2O_8$ concentration was determined to be 0.1 mol·L^{-1}.

The higher $Na_2S_2O_8$ concentration accelerated the reaction rate of reaction (3), increasing the active free radical concentration in the solution [33]. Thus, the oxidation ability of $Na_2S_2O_8$-urea-seawater composite redox system was strengthened. At $Na_2S_2O_8$ concentration below 0.1 mol·L^{-1}, the limited $Na_2S_2O_8$ addition restrained the generation of active free radicals. The interaction between the NO molecules and active radicals was weak, with a limited number of effective collisions between them. Therefore, an increase in the $Na_2S_2O_8$ concentration enhanced the interaction between NO molecules and active radicals and strengthened the NO removal performance. However, when the $Na_2S_2O_8$ concentration exceeded 0.1 mol·L^{-1}, increasing the

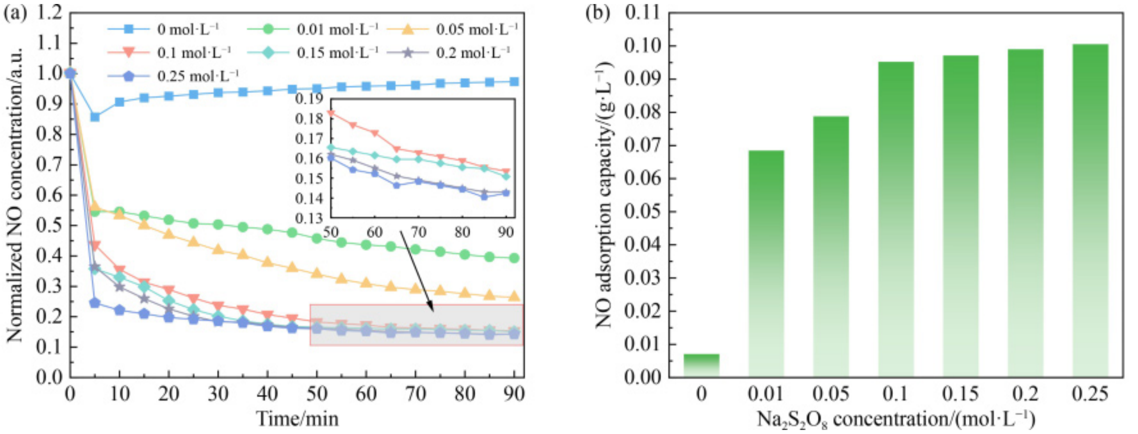

Fig. 4 (a) Variation of NO concentration with time during NO removal using the $Na_2S_2O_8$-urea-seawater composite redox system, and (b) the corresponding mass of NO absorbed per unit volume of $Na_2S_2O_8$-urea-seawater composite redox system under different $Na_2S_2O_8$ concentrations (experimental conditions: reaction temperature = 70 °C, $Na_2S_2O_8$ concentration = 0–0.25 $mol \cdot L^{-1}$, urea concentration = 0.5 $mol \cdot L^{-1}$, NO concentration = 1000 ppm, pH = 7).

$Na_2S_2O_8$ concentration did not significantly improve NO removal due to the constraints in the NO and urea concentrations. Therefore, the increased $Na_2S_2O_8$ concentration could increase the concentration of the effective oxidizing substances in the solution, which was beneficial for the NO removal using $Na_2S_2O_8$-urea-seawater composite redox system to a certain extent.

3.4 Effect of urea concentration

The influence of different urea concentrations on NO removal using $Na_2S_2O_8$-urea-seawater composite redox system was depicted in Fig. 5. An increase in the urea concentration in the range of 0–0.5 $mol \cdot L^{-1}$ decreased the normalized NO concentration from 0.455 to 0.154, and increased the corresponding mass of NO absorbed per unit volume solution from 0.057 to 0.095 $g \cdot L^{-1}$. Consequently, this indicated that the addition of urea significantly promoted NO removal. However, the normalized NO concentration and the corresponding mass of NO absorbed per unit volume solution remained stable for a further increase in the urea concentration to 0.9 $mol \cdot L^{-1}$. Therefore, the optimal urea concentration was identified as 0.5 $mol \cdot L^{-1}$.

In the absence of urea, the NO_2^- generated during NO oxidation consumed the active free radicals via reactions (10) and (11), diminishing the oxidation ability of the composite solution [44]. In addition, NO_2^- would regenerate NO again through reaction (7), resulting in an increase of the outlet NO concentration. Therefore, the NO removal performance was weak without urea. When urea was added to the solution, urea consumed the intermediate products NO_2 and NO_2^- via reactions (12) and (13), thereby reducing the additional consumption of active free radicals and inhibiting the NO regeneration [47]. Meanwhile, urea could directly react with NO to reduce NO emission through reaction (14). Therefore, the addition of urea reduced the consumption of active

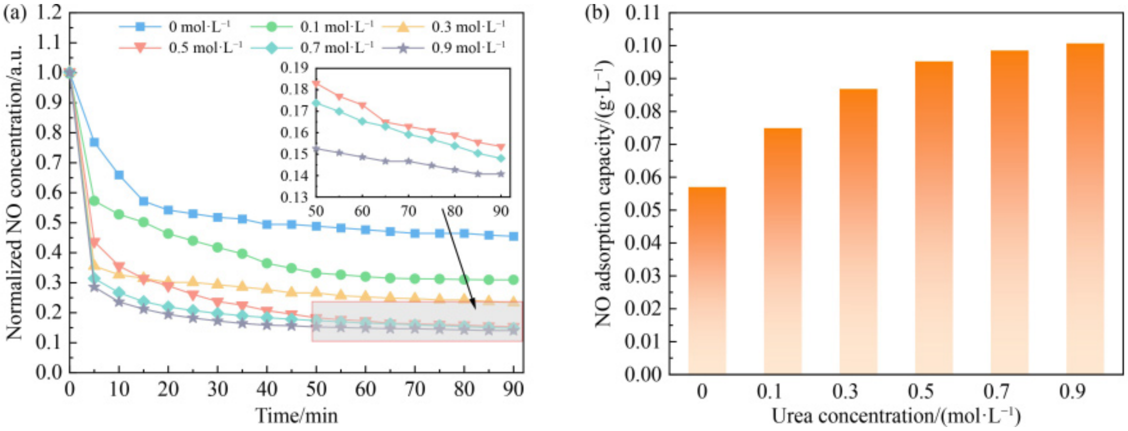

Fig. 5 (a) Variation of NO concentration with time during NO removal using the $Na_2S_2O_8$-urea-seawater composite redox system, and (b) the corresponding mass of NO absorbed per unit volume of $Na_2S_2O_8$-urea-seawater composite redox system under different urea concentrations (experimental conditions: reaction temperature = 70 °C, $Na_2S_2O_8$ concentration = 0.1 $mol \cdot L^{-1}$, urea concentration = 0–0.9 $mol \cdot L^{-1}$, NO concentration = 1000 ppm, pH = 7).

free radicals and enhanced the direct removal of NO to strengthen NO removal. Furthermore, excess urea was more inclined to react with NO_2^-, NO, and NO_2 rather than with $Na_2S_2O_8$, and thus the reaction between urea and $Na_2S_2O_8$ could be to some extent negligible, resulting in an increase in NO removal performance with increasing urea concentration [44,45]. When the urea concentration exceeded 0.5 $mol \cdot L^{-1}$, the NO removal performance did not continue to increase due to the limitation of $Na_2S_2O_8$ and NO concentrations.

$$\cdot SO_4^- + NO_2^- \rightarrow SO_4^{2-} + NO_2 \tag{10}$$

$$\cdot OH + NO_2^- \rightarrow NO_2 + OH^- \tag{11}$$

$$6NO_2 + 4(NH_2)_2CO \rightarrow 7N_2 + 4CO_2 + 8H_2O \tag{12}$$

$$(NH_2)_2CO + 2NO_2^- + 2H^+ \rightarrow 2N_2 + CO_2 + 3H_2O \tag{13}$$

$$6NO + 2(NH_2)_2CO \rightarrow 5N_2 + 2CO_2 + 4H_2O \tag{14}$$

3.5 Effect of pH value

The effect of pH value on NO removal using $Na_2S_2O_8$-urea-seawater composite redox system was presented in Fig. 6. As the initial pH value of the $Na_2S_2O_8$-urea-seawater composite redox system increased from 4 to 7, the normalized NO concentration increased slightly from 0.102 to 0.154, and the corresponding mass of NO absorbed per unit volume of solution decreased from 0.108 to 0.095 $g \cdot L^{-1}$. The effect of the initial pH value of $Na_2S_2O_8$-urea-seawater composite redox system was not obvious. However, a further increase in the initial pH value to 8 decreased the normalized NO concentration, and the corresponding mass of NO absorbed reached 0.108 $g \cdot L^{-1}$. With a subsequent increase in the initial pH value to 11, the normalized NO concentration increased to 0.273, while the corresponding mass of NO absorbed declined to 0.079 $g \cdot L^{-1}$. Therefore, this suggested that the optimal NO removal performance was obtained when the initial pH value was set at 8.

Under acidic conditions, reactions (15)–(17) occurred. At the initial pH of 4, the acidic atmosphere facilitated the activation of $S_2O_8^{2-}$, and reactions (15) and (16) were the primary reactions. At this point, reaction (18) enhanced the NO removal process [48]. As the initial pH increased for pH values less than 7, the activity of reactions (15) and (16) was weakened and that of reaction (17) was enhanced, which increased the consumption of $Na_2S_2O_8$ and reduced the oxidizing capacity of $Na_2S_2O_8$ with constant concentration. Urea could alter the acidity of solution through reaction (19), which weakened reactions (16) and (18). Consequently, the composite solution had a reduced oxidizing capacity. When the pH was increased in the range of 7–8, the reaction (20) occurred in a weakly alkaline environment, with a higher reaction rate than reaction (4). $\cdot OH$ have superior oxidizing ability compared to $\cdot SO_4^-$ [49]. Consequently, the rapid generation rate of $\cdot OH$ increased the active free radical concentration and enhanced NO removal. As the pH continued to increase to 11, reaction (20) facilitated the NO removal, but the inert oxygen radicals ($\cdot O^-$) generated through reaction (21) in a strongly alkaline environment exhibited weaker oxidizing activity than $\cdot OH$ [50]. Therefore, the NO removal performance was weakened when the pH increased from 8 to 11.

$$S_2O_8^{2-} + H^+ \rightarrow SO_4 + HSO_4^- \tag{15}$$

$$SO_4 + H_2O \rightarrow H_2SO_5 \rightarrow H^+ + HSO_5^- \tag{16}$$

$$S_2O_8^{2-} + H_2O \xrightarrow{H^+} 2H^+ + 2SO_4^{2-} + \frac{1}{2}O_2 \tag{17}$$

$$2NO + 3HSO_5^- + H_2O \rightarrow 2NO_2^- + 3SO_4^{2-} + 5H^+ \tag{18}$$

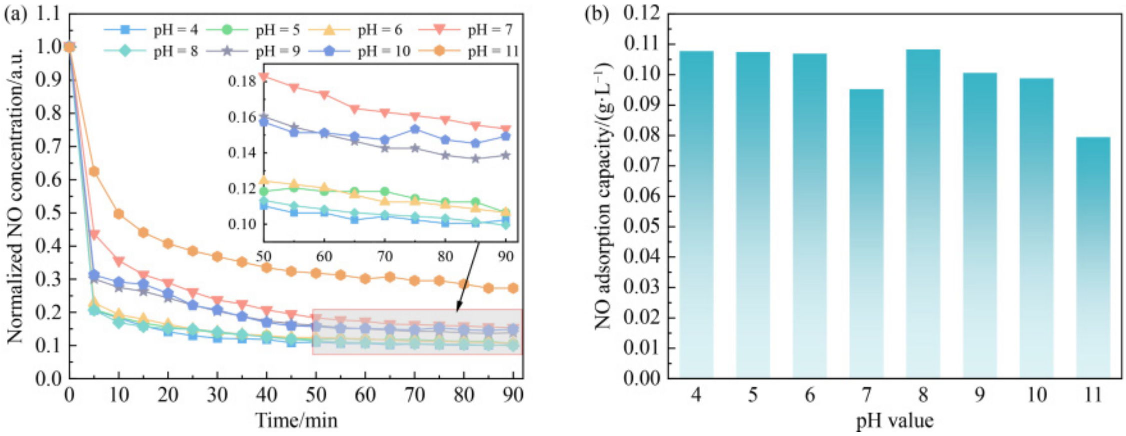

Fig. 6 (a) Variation of NO concentration with time during NO removal using the $Na_2S_2O_8$-urea-seawater composite redox system, and (b) the corresponding mass of NO absorbed per unit volume of $Na_2S_2O_8$-urea-seawater composite redox system under different initial pH (experimental conditions: reaction temperature = 70 °C, $Na_2S_2O_8$ concentration = 0.1 $mol \cdot L^{-1}$, urea concentration = 0.5 $mol \cdot L^{-1}$, NO concentration = 1000 ppm, pH = 4–11).

$$(NH_2)_2CO + H_2O + 2HSO_4^- \rightarrow 2NH_4^+ + 2SO_4^{2-} + CO_2 \quad (19)$$

$$\cdot SO_4^- + OH^- \rightarrow \cdot OH + SO_4^{2-} \quad (20)$$

$$\cdot OH + OH^- \rightarrow \cdot O^- + H_2O \quad (21)$$

3.6 Effect of initial NO concentration

Figure 7 illustrated the variation of NO concentration with time under different NO concentrations during NO removal using the $Na_2S_2O_8$-urea-seawater composite redox system. The normalized NO concentration increased from 0.144 to 0.163 as the NO concentration rose from 400 to 1500 ppm. Although the increase in the NO concentration decreased the NO removal performance, the reduction was insignificant. It suggested that the $Na_2S_2O_8$-urea-seawater composite redox system could effectively handle a wide range of NO concentrations. The increase in the initial NO concentration enhanced the gas phase partial pressure of NO, which strengthened the gas-liquid mass transfer process and increased the number of gas molecules passing through the reactor per unit of time [51]. However, the oxidizing active groups in the solution were limited when $Na_2S_2O_8$ concentration, urea concentration, and reaction temperature were constant. The increase in the number of NO gas molecules decreased the relative molar ratio of oxidizing species to NO. Thus, as the NO concentration increased, the normalized NO concentration gradually increased and the NO removal performance decreased.

3.7 The NO_3^- concentration in $Na_2S_2O_8$-urea-seawater composite redox system

As illustrated in Fig. 8, the NO_3^- concentration after the reaction was measured to investigate the effect of reaction conditions on the NO_3^- concentration in the $Na_2S_2O_8$-urea-seawater composite redox system. After the reaction, the NO_3^- concentrations in $Na_2S_2O_8$-urea-seawater composite redox system increased as the reaction temperature, $Na_2S_2O_8$ concentration, and NO concentration increased. Additionally, the NO_3^- concentrations in $Na_2S_2O_8$-urea-seawater composite redox system after the reaction decreased within the initial pH range of 4–7. Conversely, the NO_3^- concentrations increased as the initial pH value increased to 8, and the NO_3^- concentration further decreased as the initial pH value increased to 11. Notably, the $Na_2S_2O_8$-urea-seawater composite redox system had a higher NO_3^- concentration after reaction than the $Na_2S_2O_8$-urea-deionized water composite redox system. The changing trend of the NO_3^- concentration closely mirrored the NO removal performance of the reaction system. This was primarily because the oxidation capacity of $Na_2S_2O_8$-urea-seawater composite redox system was influenced by reaction temperature, $Na_2S_2O_8$ concentration, pH value, NO concentration, and solution carrier at a constant urea concentration. Under the condition of certain urea concentration, the reducing ability of urea was limited. The solution had a higher concentration of NO_3^- generated by NO oxidation when the denitrification ability of $Na_2S_2O_8$-urea-seawater composite redox system was stronger.

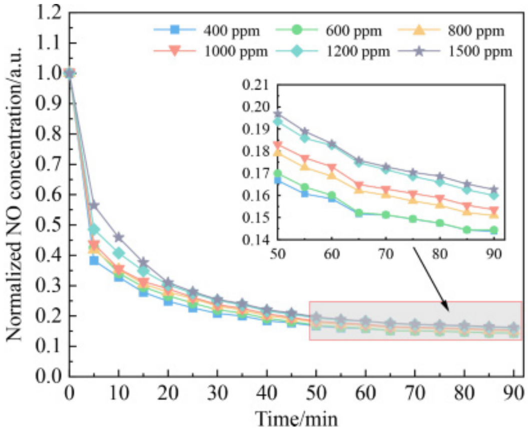

Fig. 7 Variation of NO concentration with time under different NO concentrations during NO removal using the $Na_2S_2O_8$-urea-seawater composite redox system (experimental conditions: reaction temperature = 70 °C, $Na_2S_2O_8$ concentration = 0.1 $mol \cdot L^{-1}$, urea concentration = 0.5 $mol \cdot L^{-1}$, NO concentration = 400–1500 ppm, pH = 7).

The urea concentration constrained the reduction process of the solution. Specifically, higher urea concentrations reduced NO_3^- concentrations in the $Na_2S_2O_8$-urea-seawater composite redox system, which could be attributed to two factors. On the one hand, the increased urea concentration promoted the consumption of NO_3^- through reaction (22). On the other hand, the reduction of NO_2^-, NO_2, and NO reduced the production of NO_3^- via reactions (12–14) [45]. Therefore, urea could effectively inhibit NO_3^- formation, ensuring that the NO_3^- concentration in $Na_2S_2O_8$-urea-seawater composite redox system met the relevant emission standards (< 60 $mg \cdot L^{-1}$).

$$5(NH_2)_2CO + 6NO_3^- + 6H^+ \rightarrow 8N_2 + 5CO_2 + 13H_2O \quad (22)$$

3.8 Denitrification mechanism of $Na_2S_2O_8$-urea-seawater composite redox system

The schematic diagram of the denitrification mechanism using $Na_2S_2O_8$-urea composite redox system is shown in Fig. 9.

The ionization of $Na_2S_2O_8$ in water could produce $S_2O_8^{2-}$, which could decompose into $\cdot SO_4^-$ via reaction (3). Furthermore, some $\cdot SO_4^-$ ions generated $\cdot OH$ through reaction (4). The radicals exhibited higher oxidation potential than $S_2O_8^{2-}$ [33,52]. The generated $\cdot SO_4^-$ and $\cdot OH$ reacted efficiently with NO to form NO_2^- via reactions (23) and (24). Subsequently, $\cdot SO_4^-$ and $\cdot OH$ oxidized the NO_2^- to form NO_2 and NO_3^-, thus

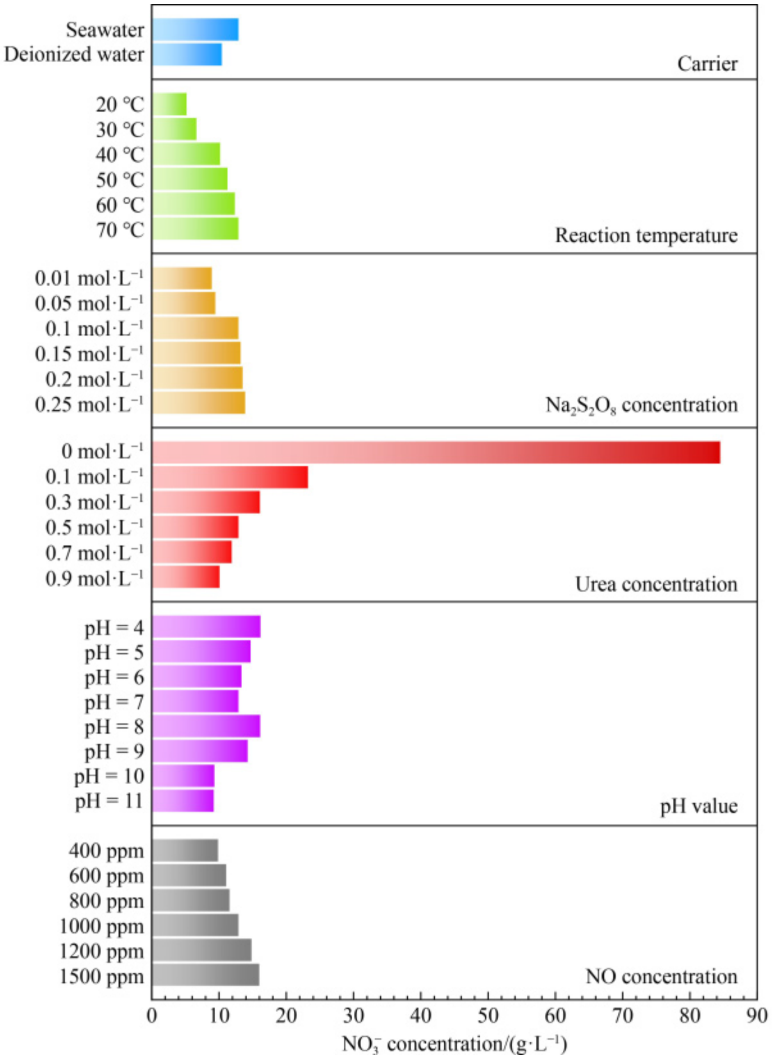

Fig. 8 The NO_3^- concentrations in $Na_2S_2O_8$-urea-seawater composite redox system after reaction.

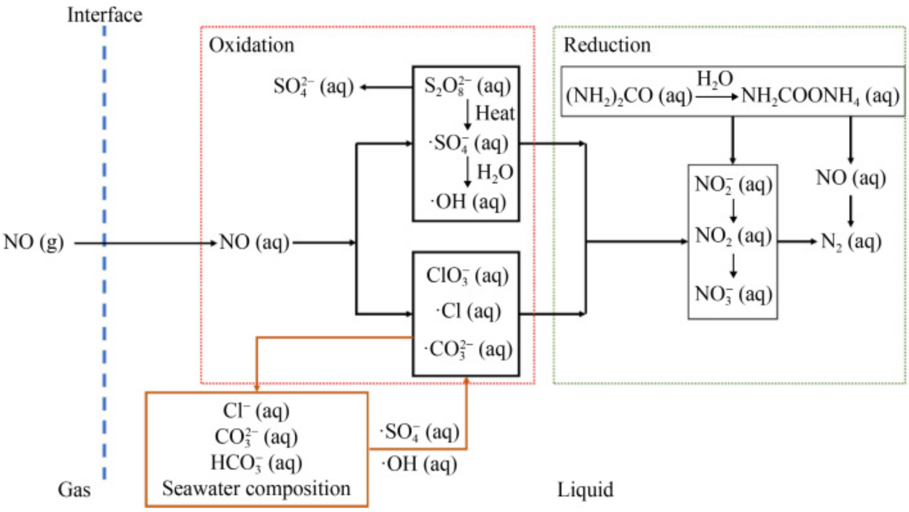

Fig. 9 The schematic diagram of the denitrification mechanism using $Na_2S_2O_8$-urea composite redox system.

contributing to the NO removal from marine exhaust gas via reactions (10), (11), (25), and (26). Simultaneously, $S_2O_8^{2-}$ could directly oxidize NO to remove NO through reaction (5).

$$\cdot SO_4^{-} + H_2O + NO \rightarrow SO_4^{2-} + NO_2^{-} + 2H^{+} \qquad (23)$$

$$\cdot OH + NO \rightarrow NO_2^- + H^+ \tag{24}$$

$$\cdot SO_4^- + NO_2 + H_2O \rightarrow SO_4^{2-} + NO_3^- + 2H^+ \tag{25}$$

$$\cdot OH + NO_2 \rightarrow NO_3^- + H^+ \tag{26}$$

It should be noted that Cl^-, HCO_3^-, and CO_3^{2-} in seawater could react with $\cdot SO_4^-$ and $\cdot OH$, which affect the distribution pattern and selectivity of active free radicals [30,53]. Cl^- could produce active $\cdot Cl$ and ClO_3^- via reactions (27–29), augmenting the overall concentration of active free radicals in the solution. Subsequently, $\cdot Cl$ and ClO_3^- could efficiently react with NO through reactions (30–33) [30]. It was inferred that $\cdot Cl$ and ClO_3^- had a good oxidation effect on NO, which improved NO removal performance. Furthermore, the CO_3^{2-} and HCO_3^- could be complexed with the $\cdot OH$ and $\cdot SO_4^-$ radicals to form $\cdot CO_3^-$ with sufficient oxidation potential via reactions (34–37).

$$\cdot SO_4^- + Cl^- \rightarrow SO_4^{2-} + \cdot Cl \tag{27}$$

$$6 \cdot SO_4^- + Cl^- + 3H_2O \rightarrow ClO_3^- + 6H^+ + 6SO_4^{2-} \tag{28}$$

$$\cdot OH + Cl^- \rightarrow OH^- + \cdot Cl \tag{29}$$

$$NO + 2 \cdot Cl + H_2O \rightarrow NO_2 + 2Cl^- + 2H^+ \tag{30}$$

$$3NO + ClO_3^- \rightarrow 3NO_2 + Cl^- \tag{31}$$

$$NO_2 + \cdot Cl + H_2O \rightarrow NO_3^- + Cl^- + 2H^+ \tag{32}$$

$$6NO_2 + ClO_3^- + 3H_2O \rightarrow 6NO_3^- + Cl^- + 6H^+ \tag{33}$$

$$\cdot SO_4^- + CO_3^{2-} \rightarrow \cdot CO_3^- + SO_4^{2-} \tag{34}$$

$$\cdot SO_4^- + HCO_3^- \rightarrow \cdot CO_3^- + SO_4^{2-} + H^+ \tag{35}$$

$$\cdot OH + CO_3^{2-} \rightarrow \cdot CO_3^- + OH^- \tag{36}$$

$$\cdot OH + HCO_3^- \rightarrow \cdot CO_3^- + H_2O \tag{37}$$

The presence of urea in $Na_2S_2O_8$-urea-seawater composite redox system altered the conversion path of element N during the denitrification of $Na_2S_2O_8$. On the one hand, urea reduced the NO_3^- to N_2 through reaction (22), decreasing the NO_3^- concentration in the solution. On the other hand, the urea could react with the intermediates (NO_2^- and NO_2) of the NO oxidation process through reactions (12) and (13), which reduced NO_3^- production [44]. Additionally, urea in the composite solution could be hydrolyzed to produce ammonium carbamate through reaction (8), which could further react with NO_2^- through reaction (9). The urea could also directly react with NO via reaction (14), avoiding the formation of NO_3^-. Therefore, from the perspective of mechanism analysis, $Na_2S_2O_8$-urea-seawater composite redox system can effectively reduce the emission of NO and NO_3^- on ships.

4 Conclusions

The $Na_2S_2O_8$-urea-seawater composite redox system was proposed as an effective scrubbing solution for NO removal from marine exhaust gas. The NO removal performance was systematically studied under different solution carriers, reaction temperatures, $Na_2S_2O_8$ concentrations, urea concentrations, pH values of the solution, and NO concentrations. The experimental results revealed that the seawater carrier was advantageous for NO oxidation. Additionally, the seawater carrier increased the active free radical content and provided an alkaline buffer system to promote NO removal. The increase in reaction temperature and $Na_2S_2O_8$ concentration could augment the active free radical concentration in $Na_2S_2O_8$-urea-seawater composite redox system to strengthen the NO removal performance. The addition of urea reduced the consumption of active free radicals and enhanced the direct NO removal, with the overall NO removal performance increasing in proportion to urea concentration. Furthermore, the NO removal performance decreased as the pH values increased within the pH ranges of 4–7 and 8–11. However, the pH values in the range of 7–8 could promote NO removal. The limitation of oxidizing active groups weakened the NO removal performance as the NO concentration increased. The addition of urea decreased significantly the NO_3^- concentration, allowing it to meet emission standards over an extended period. It was clear that $Na_2S_2O_8$-urea-seawater composite redox system had an outstanding NO removal performance and ability to reduce NO_3^- emission. The $Na_2S_2O_8$-urea-seawater composite redox system was a promising scrubbing solution for NO removal from marine exhaust gases.

Competing interests The authors declare that they have no competing interests.

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