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# Effect of Oxidants in the Utilization of Polysulfone Hollow Fiber Membrane Module as Bubble Reactor for Simultaneously Removal of  $NO<sub>x</sub>$  and  $SO<sub>2</sub>$

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**Abstract.** Air pollution has become a global issue and contributes significantly to climate change, mainly due to the massive energy consumption in industry and the transportation sector. Emissions of harmful gases from burning fuels such as  $NO<sub>x</sub>$  and  $SO<sub>2</sub>$  are the most significant sources of environmental pollution, which have negative impacts on the environment, such as the greenhouse effect, damage to the ozone layer, photochemical smog, and acid rain, and can interfere with the respiratory system in humans. This study utilizes hollow fiber membrane modules, which act as a reactor on the shell side of the membrane module and a gas distributor by the membrane fiber to remove  $NO<sub>x</sub>$  and  $SO<sub>2</sub>$  spontaneously. The oxidant solutions used were a pair of hydrogen peroxide and sodium hydroxide  $(H_2O_2-NaOH)$  solutions, a pair of sodium chlorite and sodium hydroxide (NaClO<sub>2</sub>-NaOH) solutions, and a pair of sodium chlorate and sodium hydroxide (NaClO<sub>3</sub>-NaOH) solutions. Based on the results of experiments,  $SO<sub>2</sub>$  can be removed entirely in the process, while  $NO<sub>x</sub>$  depends on the feed gas flow rate and the concentration of the oxidant solution used. H<sub>2</sub>O<sub>2</sub> is the most effective oxidizing agent in removing  $NO<sub>x</sub>$  and  $SO<sub>2</sub>$  because of its higher oxidative properties than NaCLO<sub>2</sub> and NaClO<sub>3</sub>. The increase in feed gas flow rate resulted in a decrease in the efficiency of  $NO_x$  removal even though the  $NO_x$  mass transfer flux and  $NO_x$  loading increased. Meanwhile, an increase in the concentration of oxidants increases the efficiency of  $NO<sub>x</sub>$  removal and mass transfer flux but decreases  $N_0$  loading. Based on the experimental results, the maximum  $N_0$ removal efficiency achieved by the oxidant solutions is 93.9, 91.1, and 88.3% for  $H_2O_2$ -NaOH, NaClO<sub>2</sub>-NaOH, and NaClO<sub>3</sub>-NaOH, respectively.

*Keywords:* Climate change; Harmful gases; NO<sub>x</sub>; Removal efficiency; SO<sub>2</sub>

### **1. Introduction**

Air pollution has become a global issue and contributes significantly to climate change due to the industry and transportation sector's massive energy consumption (Manisalidis *et al.*, 2020). In many countries, such as Indonesia, the emission of air pollutants from industrial and transportation activities is increasing due to the burning of fossil fuels. Emissions of harmful gases from the fuels burning process, such as  $NO<sub>x</sub>$  and  $SO<sub>2</sub>$ , are the most significant causes of environmental pollution, which have negative impacts on the environment, such as the greenhouse effect, damage to the ozone layer, photochemical

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smog, and acid rain, and can interfere with the respiratory system in humans (Kartohardjono *et al.*, 2019; Ma *et al.*, 2019). One promising strategy to reduce exhaust gas pollutants such as NO<sup>x</sup> and SO<sup>2</sup> is to control the source of the pollution (Zhu *et al.*, 2023). Removing  $NO<sub>x</sub>$  and  $SO<sub>2</sub>$  in flue gases, such as those from coal burn boilers and marine diesel engines, is currently attracting much attention (Zhao *et al.*, 2022; Yan *et al.*, 2020). In 2020, analysis tools revealed a significant increase in the trend of  $NO<sub>x</sub>$  and  $SO<sub>2</sub>$  emissions from Indonesian coal-fired power plants. The emissions for  $SO<sub>2</sub>$  and  $NO<sub>x</sub>$  were reported as 798.5 kton/year and 120.02 kton/year, respectively (Sunarno, Purwanto, and Suryono, 2021). Considering the losses that  $SO_2$  and  $NO_x$  gases can cause, the Indonesian Government has set various regulations related to the quality standard of the two gases, which is 200 µg/Nm<sup>3</sup> or 0.16 and 0.076 ppm for NO and SO<sub>2</sub>, respectively (Ministry of Environment and Forestry, 2019).

Several technologies have been developed to control pollutant emissions in many industries, including Selective Catalytic Reduction (SCR) and Selective Non-Catalytic Reduction (SNCR) for NO<sup>x</sup> removal (Karamah *et al.*, 2021) and Flue Gas Desulfurization for SO2 removal (Sharma *et al.*, 2012). The conventional technology of SCR for NO<sup>x</sup> and FGD for SO<sup>2</sup> has been widely adopted in various countries (Xu *et al.*, 2022). With increasing environmental awareness, the government and society need strict legislation and regulations to minimize NO<sup>x</sup> and SO<sup>2</sup> emissions into the air (Jia *et al.*, 2022; Chen *et al.*, 2021). Although the removal rate is relatively high, several problems are faced, such as the catalyst used in the SCR system is quite expensive, must be replaced periodically, and requires a large area of land for its application (Guo *et al.*, 2018). Therefore, it has prompted the search for suitable alternative technologies to remove  $SO_2$  and  $NO_x$  simultaneously. The simultaneous removal of  $SO<sub>2</sub>$  and  $NO<sub>x</sub>$  through two different technologies needs high operational and investment costs because the process is becoming more complex (Zhao *et al.*, 2021b; Cheng and Zhang, 2018) and has a high working area (Zhao *et al.*, 2021a). NO<sup>x</sup> and SO<sub>2</sub> are both acidic gases, but the solubility of  $NO_x$  in water is less than SO<sub>2</sub>, so a different technique is needed to remove the two gases (Fang *et al.*, 2011).

Several approaches that can be used to remove  $NO<sub>x</sub>$  and  $SO<sub>2</sub>$  simultaneously include the oxidation approach, the reduction approach, the absorption or adsorption approach, and the microbial approach (Chen *et al.*, 2021). Oxidation approaches include gas-liquid oxidation, gas-liquid oxidation, and gas-solid oxidation. The gas oxidation approach can use ozone (Sun *et al.*, 2013), oxygen (Atkinson *et al.*, 2004), the oxidant chlorine (Cl2) and chlorine dioxide (ClO2) (Mostafa *et al.*, 2018), and non-thermal plasma (Feng *et al.*, 2018). The gas-liquid oxidation approach, also known as the wet process, includes gas-liquid oxidation using H2O<sup>2</sup> (Kartohardjono *et al.*, 2023; Waclawek *et al.*, 2017), Peroxydisulfate/Peroxymonosulfate (Matzek and Carter, 2016), and NaClO/NaClO<sub>2</sub> (Zhitao *et al.*, 2019). Meanwhile, for the gas-solid oxidation approach through a photocatalytic process using catalysts such as TiO<sup>2</sup> (Su *et al.*, 2013), ZnO (Boyjoo *et al.*, 2017), CeO<sup>2</sup> (Tsang *et al.*, 2019), Bi2WO<sup>6</sup> (Wang *et al.*, 2017), and BiOX (Cl, Br, I) (Xia *et al.*, 2015). The reduction approach includes gas-liquid reduction, gas-liquid reduction, and gas-solid reduction. Reduction of gases can use reductants such as CO (Makeev and Peskove, 2013), H<sup>2</sup> (Ge *et al.*, 2018), and CxHy (Pan *et al.*, 2015). The gas-liquid reduction can use ammonia, urea, and sodium sulfide (Na2S) (Mok and Lee, 2006), while gas-solid reduction can use carbon materials (Ma *et al.*, 2013). Absorption/adsorption approaches include Alkaline solution absorption (Sun *et al.*, 2015), complex absorption (Guo *et al.*, 2014), carbon-based adsorption (Xiong *et al.*, 2015), zeolite-based adsorption (Rezaei *et al.*, 2015), metal oxidebased adsorption (Vikrant *et al.*, 2017). Meanwhile, the microbial approach uses autotrophic micro-organisms under anoxic conditions (Xiao *et al.*, 2017).

The wet method approach is becoming more commonly applied to remove  $NO<sub>x</sub>$  and  $SO<sub>2</sub>$ simultaneously because of its high efficiency and low cost (Johansson, Normann, and Andersson, 2021). The wet method includes wet scrubbing technology, widely used in  $SO<sub>2</sub>$ gas removal processes, and a bubble reactor to remove NO<sup>x</sup> (Zhang *et al.*, 2021). Bubble reactors are multiphase reactors widely used in various industries, such as the chemical, petrochemical, and biochemical industries. These reactors play a pivotal role in numerous chemical processes encompassing oxidation, chlorination, alkylation, polymerization, and hydrogenation reactions. In these reactors, the feed gas is introduced into the system and then dispersed into bubbles as part of the technical process. Meanwhile, the liquid phase or liquid-solid suspension can be operated in batch mode or flowed in the direction/opposite direction of the gas flow so that contact or reaction will occur in the reactor column (Jakobsen, Linborg, and Dorao, 2005).

The wet method facilitates the removal of gaseous pollutants through contact between pollutant gas and oxidant liquid, which triggers a reaction between pollutant gas and oxidant liquid, becoming other species (Jin *et al.*, 2006). The main obstacle in removing NO<sup>x</sup> gas through the wet method is that  $NO<sub>x</sub>$  gas is a species that cannot be dissolved in the oxidant(Kang *et al.*, 2020). To address this issue, an oxidizing agent is introduced to convert the  $NO<sub>x</sub>$  species into more soluble forms, such as hydrogen peroxide (H2O2), sodium chlorite (NaClO2), and sodium chlorate (NaClO3). At the same time, an alkaline solution such as NaOH can be applied to remove SO<sub>2</sub> (Purnawan *et al.*, 2021).

Membrane technology is a non-conventional technique that can simultaneously remove  $NO<sub>x</sub>$  and  $SO<sub>2</sub>$ . The membrane is a porous medium in the form of a thin film that can diffusely transfer certain gas compounds due to a driving force in the form of concentration toward the solvent through the membrane pore (Wang and Yu, 2017). A membrane contactor has several advantages, such as ease of operation and scale-up, low separation costs and energy consumption, and high efficiency (Kartohardjono *et al.*, 2020). This study utilized a hollow fiber membrane module (HFMM) that functions as a reactor and gas distributor to remove  $NO<sub>x</sub>$  and  $SO<sub>2</sub>$  spontaneously. Using an HFMM as a bubble reactor enhances the area for gasliquid contact, providing a better removal reaction between  $NO<sub>x</sub>$  and  $SO<sub>2</sub>$  gases with the applied oxidant solutions. The oxidant solutions used were a pair of  $H_2O_2$ -NaOH solutions, a pair of NaClO<sub>2</sub>-NaOH solutions, and a pair of NaClO<sub>3</sub>-NaOH solutions.

The reactions between  $NO<sub>x</sub>$  and  $SO<sub>2</sub>$  with a pair of  $H<sub>2</sub>O<sub>2</sub>$  and NaOH solutions are presented in Equations (1) – (4) (Purnawan *et al.*, 2021; Sun, Zwolińska, and Chmielewski, 2016):

$$
NO + NO_2 + H_2O \leftrightarrow 2HNO_2 \tag{1}
$$

$$
HNO2 + H2O2 \rightarrow HNO3 + H2O
$$
 (2)

$$
SO_2 + H_2O_2 \rightarrow H_2SO_4 \tag{3}
$$

$$
HNO3 + H2SO4 + 3NaOH \rightarrow NaNO3 + Na2SO4 + 3H2O
$$
 (4)

The reactions that occur between  $NO<sub>x</sub>$  and  $SO<sub>2</sub>$  with a pair of NaClO<sub>2</sub> and NaOH solutions are presented in Equations (5) – (8) (Zhao *et al.*, 2010; Chien, Chu, and Hsueh, 2003):

$$
NO + ClO2 \rightarrow NO2 + ClO-
$$
 (5)

$$
2NO2 + ClO2- + 2OH- \to 2NO3- + ClO- + H2O
$$
 (6)

$$
4ClO_2^- + 2H^+ \to 2ClO_2 + ClO_3^- + H_2O \tag{7}
$$

$$
2NaOH + SO2 \rightarrow H2O + Na2 SO3
$$
 (8)

Meanwhile, the reactions that occur between  $NO<sub>x</sub>$  and  $SO<sub>2</sub>$  with a pair of NaClO<sub>3</sub> and NaOH solutions are presented in Equations (9) – (10) (Zhao *et al.*, 2020; Shi, Sun, and Cui, 2019):

$$
NaClO3 + 2NO + 2NaOH \rightarrow 2NaNO3 + NaCl + H2O
$$
 (9)

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$$
2NaOH + SO2 \rightarrow H2O + Na2SO3
$$
 (10)

#### **2. Methods**

The CV Bandung Indonesia supplied the polysulfone hollow fiber membrane module consisting of 50 fibers used in the study. The analytic grade H2O2, NaClO2, NaClO3, and NaOH are provided by Merck Indonesia. Meanwhile, the feed gas in the form of a gas mixture of 600 ppm  $NO<sub>x</sub>$  and 500 ppm  $SO<sub>2</sub>$  in nitrogen was provided by PT EIN Indonesia. The feed gas flow rate was regulated during the experiments using the CX Series mass flow controller, which can precisely control the gas flow rate. In addition, the concentration of gases entering and leaving the membrane was measured using an ECOM-D Gas analyzer.

The HFMM operates on a principle similar to that of a bubble reactor. The oxidant, which contains a pair of 200 mL solutions of  $H_2O_2$ -NaOH, NaClO<sub>2</sub>-NaOH, or NaClO<sub>3</sub>-NaOH, is located on the shell side of the HFMM. The feed gas stream containing  $SO<sub>2</sub>$  and  $NO<sub>x</sub>$  entered the membrane module through a silicone hose connection to the lumen fibers. A CX Series mass flow controller regulated the gas flow rates and made contact with oxidant solutions in the shell side of HFMM. The ECOM-D Gas Analyzer measured the  $NO<sub>x</sub>$  and SO2 composition, as it leaving the membrane module.

The  $NO<sub>x</sub>$  or  $SO<sub>2</sub>$  removal efficiency, flux, and gas loading were calculated by Equations (11-14) (Kartohardjono *et al.*, 2020):

$$
\%R = 100 \frac{c_{in} - c_{out}}{c_{in}} \tag{11}
$$

$$
J = \frac{Gas_{Abs}}{A_m} \tag{12}
$$

$$
Gas~loading = \frac{Gas_{Abs}}{c}
$$
 (13)

$$
C_{oxidant}
$$
\n
$$
G_{\text{S}} = (C - C - 1)Q_2 \frac{P}{P}
$$
\n(14)

$$
Gas_{Abs} = (C_{in} - C_{out})Q_G \frac{r}{RT}
$$
\n(14)

*C*in and *C*out are the NO<sup>x</sup> or SO<sup>2</sup> concentrations in the feed gas and gas left from the HFMM, respectively. Meanwhile, *Gas*Abs, *A*m, *C*oxidant, *Q*G, *P*, *T*, and *R* are NO<sup>x</sup> or SO<sup>2</sup> absorbed by the oxidant, membrane area, concentration of  $H_2O_2$ , NaClO<sub>2</sub>, or NaClO<sub>3</sub>, feed gas flowrate, pressure, temperature, and ideal gas constant, respectively. The series of experimental equipment is shown in Figure 1. All experiments were conducted three times, and the experimental results' standard deviation was less than 6%.



**Figure 1** Experimental equipment set up: 1. Feed gas tank, 2. Gas regulator, 3. Mass flow controler, 4. HFMM, 5. Gas Analyzer, 6. Data storage

#### **3. Results and Discussion**

This study used a feed gas with initial concentrations of  $NO<sub>x</sub>$  and  $SO<sub>2</sub>$  of 600 ppm and 500 ppm, respectively. The oxidant solutions used were H2O2-NaOH solutions, NaClO2NaOH solutions, and NaClO<sub>3</sub>-NaOH solutions with a concentration of 0.1M and 0.5M of 200 mL each. The gas flow in the experiments varied from 0.1 to 0.2 L/minute at a constant temperature and pressure of 28°C and 1 atm, respectively. The process of  $NO_x$  and  $SO_2$  gases transfer through the HFMM during the experiment occurred in three stages: (i) gas diffusion to the inner surface of the fiber membrane; (ii) gas diffusion through the membrane pores to the outer surface of the membrane fibers; and (iii) gas absorption by the oxidant (Kartohardjono *et al.*, 2019).

For all experiments, the  $SO<sub>2</sub>$  removal efficiency is generally 100%, as it has a high solubility in water and better chemical reactivity (Liu, Shi, and Wang, 2022), so its presence in the feed gas will be examined to see the influence on  $NO<sub>x</sub>$  removal. Figure 2 shows the impact of varying feed gas flow rates on  $NO<sub>x</sub>$  gas's absorption efficiency (%R) with various oxidants.

As demonstrated in Figure 2, the removal efficiency of  $NO<sub>x</sub>$  for all oxidants decreases with increasing feed gas flow. Increasing the feed gas flow causes an increase in the  $NO<sub>x</sub>$ absorbed by the oxidant solutions, thereby increasing the efficiency of  $NO<sub>x</sub>$  removal. However, increasing the feed gas flow led to less gas residence time in the HFMM, which caused a decrease in the removal efficiency of NOx. The decline in the removal efficiency of  $NO<sub>x</sub>$  to the gas flow indicates that the effect of gas residence time in the membrane module is more influential than the increase in the adsorbed NO<sup>x</sup> (Xu *et al.*, 2022). The removal efficiency of NO<sub>x</sub> decreased from 93.9 to 81.3%, 91.1 to 79.5%, and 88.3 to 71.0% for H<sub>2</sub>O<sub>2</sub>-NaOH, NaClO2-NaOH, and NaClO3-NaOH adsorbents, respectively. Oxidant solutions containing  $H_2O_2$  have the highest removal efficiency because of their higher oxidative properties than NaClO<sub>2</sub> and NaClO<sub>3</sub>. The standard reduction potentials for H<sub>2</sub>O<sub>2</sub>, NaClO<sub>2</sub>, and NaClO<sup>3</sup> are 1.77, 0.76, and 0.62 Volt, respectively (Purnawan *et al.*, 2021; Lide, 2004). Previous studies showed a slight decrease in the removal efficiency of  $NO<sub>x</sub>$  from about 99.8 to 98.8%, 99.4 to 98.6%, and 99.3 to 98.3% for H2O2-HNO3, NaClO2-NaOH, and NaClO3- NaOH oxidant pairs, respectively, under the same conditions as this study using feed gas containing 600 ppm  $NO_x$  without  $SO_2$  and flow rates from 100 to 200 mL/min (Purnawan *et al.*, 2021). Thus, it is clear that the presence of SO<sub>2</sub> in the feed gas reduces the NO<sub>x</sub> removal efficiency due to the influence of competition in consuming the oxidant solution (Kartohardjono *et al.*, 2023), as shown in Equations (3), (8), and (10). In addition, the wet method has the disadvantage that it can only be used indirectly if the exhaust gas temperature is high enough because the wet process is only adaptable to operate at ambient temperature.



**Figure 2** NO<sup>x</sup> removal efficiency, *R-NOx*, at various feed gas flow rates, *Q*<sup>G</sup>

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The  $NO<sub>x</sub>$  mass transfer flux, as presented in Figure 3, rises with increasing the feed gas flow, indicating that increasing gas flow contributes to an increase in oxidant performance in absorbing  $NO<sub>x</sub>$  passing through the membrane. With the feed gas flow increase from 100 CC/min to 200 CC/min, the NO<sub>x</sub> mass transfer flux rose from 4.9 to  $8.4 \times 10^{-8}$  mmol/cm<sup>2</sup>.s, 4.7 to 8.2  $\times$ 10<sup>-8</sup> mmol/cm<sup>2</sup>.s, and 4.6 to 7.4  $\times$ 10<sup>-8</sup> mmol/cm2.s, for the H<sub>2</sub>O<sub>2</sub>-NaOH, NaClO<sub>2</sub>-NaOH, and NaClO3-NaOH oxidant pairs, respectively. Increasing the gas flow enhances the absorbed  $NO<sub>x</sub>$ , as presented in Figure 3, so it increases the flux in the end. A similar phenomenon also occurs for  $NO<sub>x</sub>$  loading, the ratio between  $NO<sub>x</sub>$  absorbed and the amount of oxidant (H<sub>2</sub>O<sub>2</sub>, NaClO<sub>2</sub>, or NaClO<sub>3</sub>), where the  $NO<sub>x</sub>$  loading appears to increase with the higher feed gas flow rate, indicating that the feed gas flow also contributes to the rise in the uptake of  $NO<sub>x</sub>$  by the oxidant solutions, as presented in Figure 4. When the feed flow raised from 100 to 200 CC/min, the  $NO<sub>x</sub>$  loading increased from 0.0019 to 0.0033 mmol/mol.s, 0.0019 to 0.0032 mmol/mol.s, and 0.0018 to 0.0026 mmol/mol.s, for the  $H_2O_2$ -NaOH, NaClO<sub>2</sub>-NaOH, and NaClO<sub>3</sub>-NaOH solvent pairs, respectively. In previous studies, under the same conditions using feed gas containing 600 ppm  $NO<sub>x</sub>$  without  $SO<sub>2</sub>$  and flow rates from 100 to CC mL/min, the mass transfer flux increased from about 0.54 to 1.1 ×10−<sup>7</sup> mmol/cm<sup>2</sup>.s for all pairs of oxidants as their  $NO<sub>x</sub>$  removal efficiency only slightly different. Meanwhile,  $NO_x$  loading increased from 0.002 to 0.004 mmol/mol.s for all pairs of oxidants (Purnawan *et al.*, 2021). It reveals that the NO<sup>x</sup> mass transfer flux and NO<sup>x</sup> loading using feed gas without  $SO_2$  is higher than that in the feed gas with  $SO_2$  due to the competition in oxidant consumption, as shown in Equations  $(5)$ ,  $(6)$ ,  $(11)$ ,  $(14)$ , and  $(19)$ .



**Figure 3** NO<sup>x</sup> mass transfer flux, *J*, and NO<sup>x</sup> absorbed at various feed gas flow rates, *Q*<sup>G</sup>



**Figure 4** NO<sup>x</sup> Loading at various feed gas flow rates, *Q*<sup>G</sup>

Figure 5 shows the effect of oxidant concentration on  $NO<sub>x</sub>$  removal efficiency and mass transfer flux. The absorption efficiency of  $NO_x$  by the oxidant solution increases with raising the oxidant solution concentration. The higher the concentration of the oxidant solution, the more chemical compounds are available to react with  $NO<sub>x</sub>$ ; thereby, it can increase the number of chemical reactions between  $NO<sub>x</sub>$  and chemical compounds in the oxidant to boost the NO<sub>x</sub> removal efficiency. The increase in NO<sub>x</sub> mass transfer flux is also proportional to the increase in  $NO<sub>x</sub>$  removal efficiency, as the feed gas flow rate used is the same for each concentration of the oxidant solution (Zhao *et al.*, 2020). NO<sup>x</sup> removal efficiency and flux increased significantly at oxidant concentrations between 0.01 and 0.1 M while only slightly increased at oxidant concentrations greater than  $0.1$  M. The efficiency of  $NO<sub>x</sub>$  removal is still relatively low, around 75.6, 88.3, and 91.6% for NaClO<sub>3</sub>, NaClO<sub>2</sub>, and H<sub>2</sub>O<sub>2</sub>, respectively, with a concentration of around 0.01 M. Hence, an increase in oxidant concentration up to 0.1 M still gives a significant increase. However, at 0.1 M oxidant concentration, the  $NO<sub>x</sub>$ removal efficiency was relatively high, around 88.3, 91.1, and 93.5% for NaClO<sub>3</sub>, NaClO<sub>2</sub>, and H2O2, respectively. Hence, an increase in oxidant concentration above 0.1 M gave a not as sharp rise in NO<sup>x</sup> removal efficiency as in the oxidant concentration area between 0.01 and 0.1 M. Similar findings were also reported in the previous studies using  $NO<sub>x</sub>$  feed gas without  $SO_2$ , where  $NO<sub>x</sub>$  removal increased with increasing oxidant concentration using a PVDF HFMM consists of 40 fibers. The  $NO<sub>x</sub>$  removal efficiency increased from 93.3 to 99.0%, 98.7 to 99.2%, and 98.9 to 99.7% with the raised of oxidant concentration from 0.05 to 0.25M, 0.01 to 0.05M, and 0.015 to 0.075 M, for the oxidants NaClO<sub>3</sub>, NaClO<sub>2</sub>, and H<sub>2</sub>O<sub>2</sub>, respectively (Purnawan *et al.*, 2021). Shi *et al.* reported a rise in NO<sup>x</sup> removal from about  $34.5$  to  $91.7\%$  when the concentration of NaClO<sub>3</sub> solution as an oxidant increased from 0.005 to 0.1 M in a bubble column reactor (Shi, Sun, and Cui, 2019). Meanwhile, Zhitao *et al.* reported that increasing the NaClO<sup>2</sup> concentration from 0.005 to 0.15 M could improve the efficiency of the NO removal process with an initial concentration of 800 ppm through a cyclic scrubbing process from 62.5 to 85% (Zhitao *et al.*, 2019). It is seen that the presence of  $SO<sub>2</sub>$  in the feed gas affects reducing the efficiency of  $NO<sub>x</sub>$  removal.



**Figure 5** NO<sup>x</sup> removal efficiency, R, and NO<sup>x</sup> mass transfer flux, *J*, at various concentration of oxidant present in oxidant solutions, *C*Abs

 $NO<sub>x</sub>$  loading in the  $NO<sub>x</sub>$  removal process using an oxidant solution is the ratio between the absorbed  $NO<sub>x</sub>$  by the oxidant solution and the number of moles of oxidant in the oxidant solution. As presented in Figure 6, an increase in the concentration of oxidants in the  $NO<sub>x</sub>$ removal process decreases gas loading because more oxidants are used, while the increase in NO<sup>x</sup> absorbed is much smaller (Karamah *et al.*, 2021). These results indicate that a low

oxidant concentration is preferable because it provides a high  $NO<sub>x</sub>$  loading. However, the desired  $NO<sub>x</sub>$  removal target also influences the decision to determine the oxidant concentration in the oxidant solution used. In this study, the  $NO<sub>x</sub>$  loading declined from around 0.015 to 0.0002 mmol/mol.s, 0.018 to 0.0002 mmol/mol.s, and 0.019 to 0.0002 mmol/mol.s for NaClO<sub>3</sub>, NaClO<sub>2</sub>, and H<sub>2</sub>O<sub>2</sub>, respectively, when the oxidant concentration in the oxidant solutions was increased from 0.01 to 1 M. Figure 6 also demonstrates that the three oxidants used have almost the same  $NO<sub>x</sub>$  loading, so the images coincide. It indicates that the type of oxidant used does not have a significant effect on  $NO<sub>x</sub>$  loading due to the insignificant difference in the amount of  $NO<sub>x</sub>$  absorbed, as also reported previously (Purnawan *et al.*, 2021). Table 1 summarizes the experimental results at a feed gas flow rate of 100 mL/min and an oxidant concentration of 0.1 M.



**Figure 6** NO<sub>x</sub> loading at various concentration of oxidant

**Table 1** The results of NO<sub>x</sub> removal efficiency, flux, and NO<sub>x</sub> loading at the concentration of the oxidant 0.1 M and feed gas flow rate of 100 mL/min



## **4. Conclusions**

 $H<sub>2</sub>O<sub>2</sub>$ , NaClO<sub>2</sub>, and NaClO<sub>3</sub> are all capable of removing NO<sub>x</sub> and SO<sub>2</sub> from flue gases, but their effectiveness depends on feed gas flow and concentration. All experimental results show that the efficiency of  $SO<sub>2</sub>$  removal is generally 100% due to its high solubility in water and better chemical reactivity.  $H_2O_2$  is a highly effective oxidizing agent and has been shown to be capable of removing both  $NO<sub>x</sub>$  and  $SO<sub>2</sub>$  because of its higher oxidative properties than NaClO<sup>2</sup> and NaClO3. Based on the experimental results, it can be seen that a rise in the feed gas flow rate decreases the  $N_{\alpha}$  removal efficiency even though the  $N_{\alpha}$ mass transfer flux and  $NO<sub>x</sub>$  loading increase. Meanwhile, increasing the oxidant concentration increases  $NO<sub>x</sub>$  removal efficiency and mass transfer flux but decreases  $NO<sub>x</sub>$ loading. The three oxidant solutions used relatively have the same  $NO<sub>x</sub>$  loading at the same oxidizing concentration.

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