

# NO reduction using low-temperature SCR assisted by a DBD method

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## Abstract

This paper discusses the removal of nitric oxide (NO) with low-temperature selective catalytic reduction driven by a dielectric barrier discharge with ammonia (NH<sub>3</sub>) as a reductant. We explored the effects of NH<sub>3</sub>, O<sub>2</sub>, temperature and water under different applied voltage on NO removal at atmospheric pressure. The results showed that when the gas concentration ration of NH<sub>3</sub>/NO was 0.23–0.67, the NO removal efficiency and the energy consumption was acceptable. The NO removal efficiency reached 84% under an applied voltage of 7 kV, 400 ppm NO and 90 ppm NH<sub>3</sub> at a temperature of 150 °C. Water vapor had a negative effect because NO formation reactions were strengthened and NH<sub>3</sub> was oxidized directly rather than reduced NO molecules. The outlet gas components were observed via Fourier transform infrared spectroscopy for revealing the decomposition process and mechanism.

Keywords: dielectric barrier discharge, NO, NH<sub>3</sub>, SCR

(Some figures may appear in colour only in the online journal)

## 1. Introduction

Nitric oxide (NO<sub>x</sub>), as one of the major factors responsible for serious problems such as acid rain, photochemical smog and PM<sub>2.5</sub>, does great damage to the environment and human health [1, 2]. The removal of nitric oxide has become one of the most challenging issues for the coming decade. The selective catalytic reduction (SCR) method, which generally uses NH<sub>3</sub> as a reductant and V<sub>2</sub>O<sub>5</sub>–WO<sub>3</sub>/TiO<sub>2</sub> as a cost-effective catalyst, has been wildly applied to NO removal in industrial application projects since the 1990s [3–5]. However, the typical V<sub>2</sub>O<sub>5</sub>–WO<sub>3</sub>/TiO<sub>2</sub> catalyst shows high performance only within the narrow temperature window of 300–450 °C [6–8]. In actual NO removal application, the NH<sub>3</sub>–SCR system is usually set upstream of dust precipitation and the desulfurization system, avoiding reheating the flue gas, but easily resulting in catalyst abrasion or poisoning and damage by sulfur-containing gas. Thus, an effective low-temperature NH<sub>3</sub>–SCR system is a new focus in the current study [9, 10].

To improve the low-temperature NH<sub>3</sub>–SCR system performance, many new kinds of precious metal catalysts have been developed [11–13]. Although these new catalysts have shown acceptable results under lower temperature, they could be easily damaged or deactivated by other gas components such as sulfur dioxide, water and dust [14–17]. In addition to the harsh requirements of reactant gas composition, these metal catalysts are also very expensive and not applicable currently in practical applications.

Some literature studies have shown that the plasma technique has relatively high catalytic activation under low-temperature [18–23]. Dielectric barrier discharge (DBD) can generate a non-thermal plasma at atmosphere pressure with the advantage of compact systems and fast or easy reactivity under low-temperature [24]. Recently, a combination of the NH<sub>3</sub>–SCR method and the DBD technique has been of interest for application in low-temperature SCR processes [6, 25, 26], for example, Guan *et al* found that the combination of NH<sub>3</sub>–SCR and a non-thermal plasma enhanced the overall reaction and allowed for an effective removal of NO<sub>x</sub>.

at 100 °C [6], and Wang *et al* studied NO removal in a plasma–catalyst system over CuCe/ZSM-5 catalysts and observed the highest NO and NO<sub>x</sub> removal efficiencies of 90.7% and 80.1% [26]. However, there are few reports concerning the denitration reaction mechanism and the effect of extra oxidation of NH<sub>3</sub> induced by the active particles in the DBD plasma system, and the optimum operation conditions for NO<sub>x</sub> removal is also needed to be cleared using the NH<sub>3</sub>–SCR system assisted by DBD [27, 28]. As the most widely used catalyst in practical business applications, the typical V<sub>2</sub>O<sub>5</sub>–WO<sub>3</sub>/TiO<sub>2</sub> catalyst shows high performance only within the narrow temperature window of 300–450 °C [6]. However, in the combined NH<sub>3</sub>–SCR and DBD system, the catalytic activity performance of the V<sub>2</sub>O<sub>5</sub>–WO<sub>3</sub>/TiO<sub>2</sub> catalyst at lower temperatures is needed, especially under the conditions of high gas hourly space velocity (GHSV).

In this paper, a study was performed of a NH<sub>3</sub>–SCR system assisted by an *in situ* DBD reactor. In the new integrated NH<sub>3</sub>–SCR DBD reactor, V<sub>2</sub>O<sub>5</sub>–WO<sub>3</sub>/TiO<sub>2</sub> was placed in the discharge area for achieving high catalytic activation under low temperature (25–150 °C). The effects of NH<sub>3</sub>, oxygen, temperature and H<sub>2</sub>O under varied applied voltage on NO<sub>x</sub> removal were explored. We also analyzed the reaction products to reveal the mechanism of NO<sub>x</sub> abatement. The results could provide useful information and suggestions on treating NO for industrial applications.

## 2. Experiment section

A schematic diagram of the NH<sub>3</sub>–SCR–DBD system is presented in figure 1. The inlet gases consisting of NO, N<sub>2</sub>, NH<sub>3</sub> and O<sub>2</sub>, are controlled by a mass flow controller (Horiba Stec-4400, JPN) and mixed with a gas blender. The mixed gases pass through a buffer chamber and then are led into the DBD reactor. The reactor consisted of an inner high-voltage electrode (graphite), two quartz tubes (outer tube with 30 mm inner diameter and 200 mm length, inner tube with 6 mm outer diameter and 300 mm length), and an outer electrode (aluminum foil). The quartz tubes were coaxial cylinder in shape with a 12 mm gap. The catalysts of V<sub>2</sub>O<sub>5</sub>–WO<sub>3</sub>/TiO<sub>2</sub> (Hunan Xinrui Co., CN) were packed evenly in the discharge area.

### 2.1. Measurements of electrical properties

The DBD power supply can provide a sinusoidal alternating voltage varying from 5 kV to 20 kV at frequency of 10–20 kHz. The voltage and power applied was measured via the voltage–charge Lissajous figure with a 200 MHz digital phosphor oscilloscope (Tektronix, TDS2024B, USA) connected to a 1000:1 HV probe (Tektronix P6015A, USA). The current was obtained by measuring the voltage of a resistor, and the Lissajous figure was measured at the discharge electrode and 0.47 μF equivalent capacitor. Figure 2(a) shows the voltage waveforms at the discharge electrode and figure 2(b) shows the corresponding charge–voltage Lissajous figure. As shown in

figure 2, the cycle A-B-C-D-A corresponds to the variation of a discharge cycle.

### 2.2. Measurements of gas content

All the gaseous components concentrations were continuously quantified using a Fourier transform infrared absorption spectrometer (FTIR, FTIR 850, Tianjin Gangdong Co., CN). Removal efficiency ( $\eta_{\text{NO}}$ ), specific energy density ( $SED$ ), and energy consumption ( $E_{\text{NO}}$ ) were defined as follows:

$$\eta_{\text{NO}} = \frac{(\text{NO}_{\text{in}} - \text{NO}_{\text{out}})}{\text{NO}_{\text{in}}} \times 100\%$$

$$SED = \frac{P}{Q}$$

$$E_{\text{NO}} = \frac{10\,000P}{36Q(\text{NO}_{\text{in}} - \text{NO}_{\text{out}})}$$

where  $\text{NO}_{\text{in}}$  and  $\text{NO}_{\text{out}}$  are the inlet and outlet concentrations of NO (ppm), respectively;  $Q$  is the gas flow rate ( $\text{l s}^{-1}$ );  $P$  is the input power (W);  $SED$  is the specific energy density ( $\text{J l}^{-1}$ ); and  $E_{\text{NO}}$  is energy consumption ( $\text{kWh kg}^{-1}$ ).

## 3. Results and discussion

### 3.1. Ammonia (NH<sub>3</sub>) effects

Figure 3(a) showed the effect of NH<sub>3</sub> upon removal efficiency of NO and energy consumption in a packed bed DBD reactor with an initial NO concentration of 400 ppm, and an inlet NH<sub>3</sub> concentration of 90 ppm. The applied voltage was adjusted from 2 kV to 7 kV. The GHSV was 19 420 h<sup>-1</sup>. The experimental temperature was 25 ± 3 °C except for the additional requirement on the temperature.

The results indicate that the addition of NH<sub>3</sub> significantly contributed to the removal efficiency of NO. With the increase in applied voltage of 4 kV to 7 kV, the NO removal efficiency was improved from 70% to 87% in the presence of NH<sub>3</sub>. The  $E_{\text{NO}}$  (energy consumption of NO) decreased with the increasing voltage, and at the same applied voltage, NH<sub>3</sub> have a positive effect on  $E_{\text{NO}}$ .

When the applied voltage increased, the electric field of the system increased both in the absence and in the presence of NH<sub>3</sub>, and the numbers of high-energy electrons and particles were also improved as a result of higher probability of collision between NO molecules and electrons or particles. The energy consumption decreased in the presence of NH<sub>3</sub> because more NO molecules were removed under the same applied voltage. With the production of lots of new high-energy particles such as NH<sub>2</sub>·, NH·, H·, the collision reactions of NO molecules were greatly enhanced.

When the concentration of NH<sub>3</sub> varied from 0 ppm to 450 ppm, the effect of NH<sub>3</sub> concentration on NO removal efficiency and energy consumption are shown in figure 2(b) under the initial NO concentration of 400 ppm and the applied voltage of 4 kV.

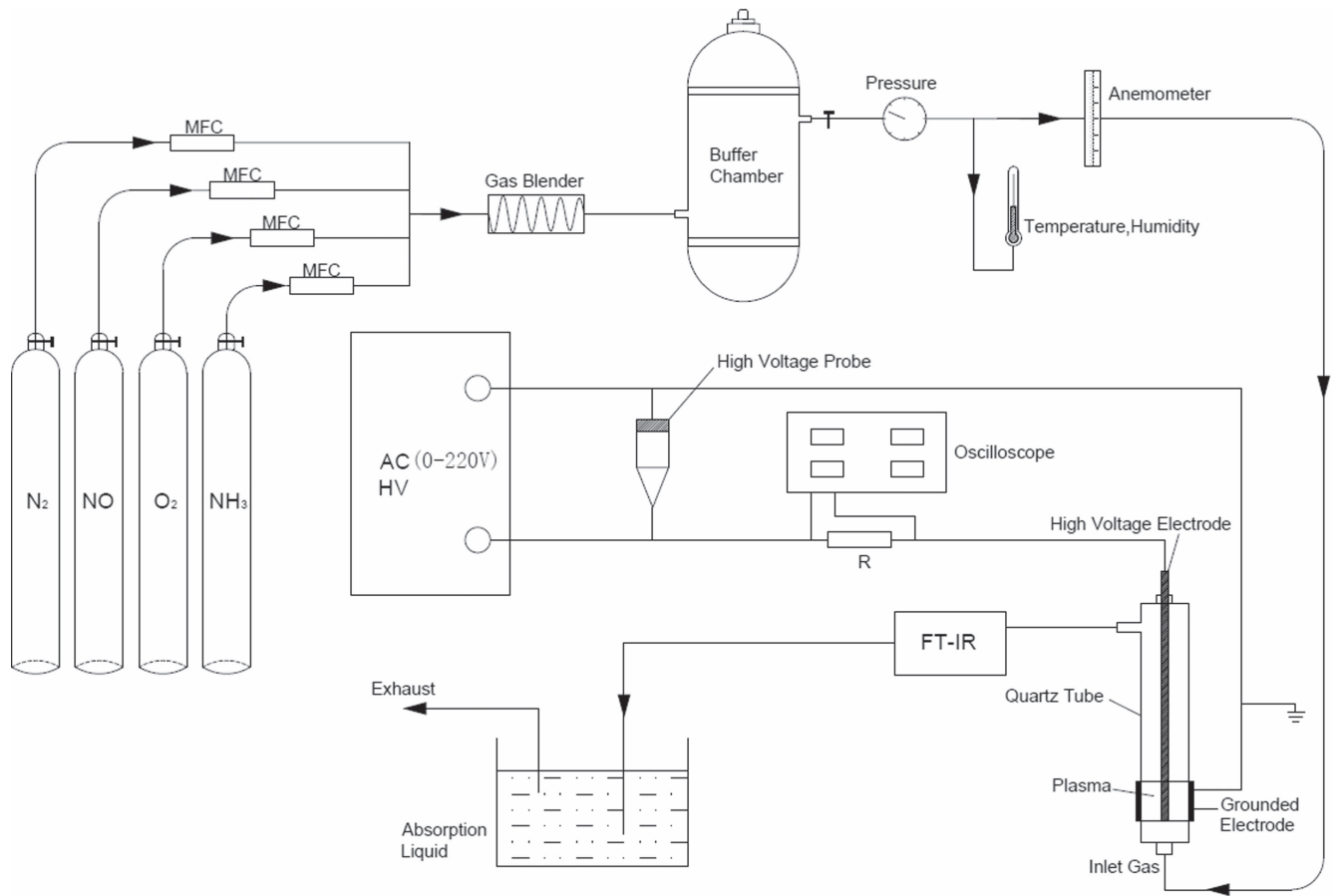


Figure 1. Schematic diagram of the experimental setup of the NH<sub>3</sub>-SCR-DBD system.

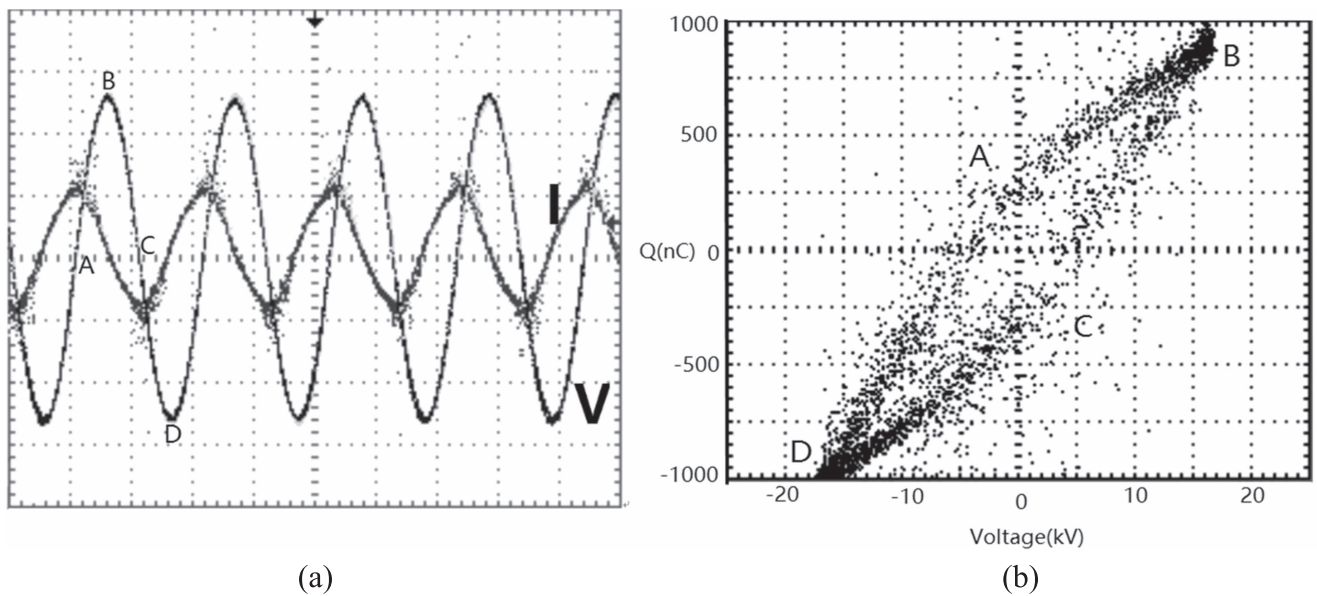
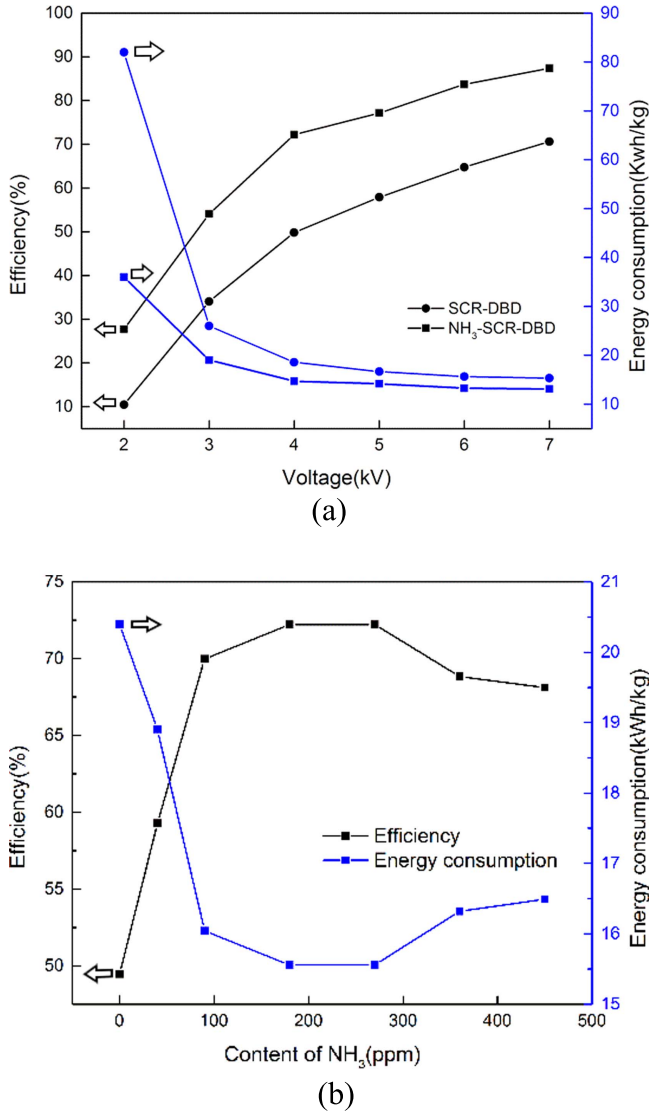


Figure 2. (a) Voltage waveforms measured at the discharge electrode. (b) Corresponding charge-voltage Lissajous figure.

Figure 3(b) shows that with the increase in initial NH<sub>3</sub> concentration, the removal efficiency of NO increased but energy consumption decreased. However, when the NH<sub>3</sub> concentration was more than 270 ppm, the NO removal efficiency decreased while  $E_{NO}$  increased slightly.  $\eta_{NO}$  decreased to 67% and  $E_{NO}$  was 16.49 kWh kg<sup>-1</sup> when the content of

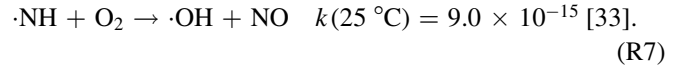
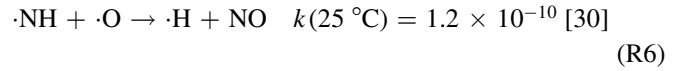
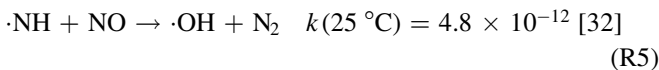
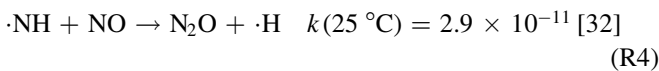
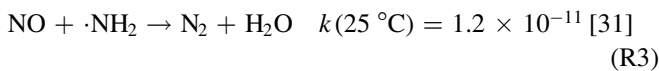
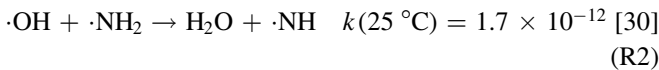
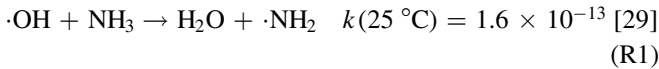
NH<sub>3</sub> was 450 ppm. When the NH<sub>3</sub> concentration was 270 ppm,  $\eta_{NO}$  reached a maximum of 72% and the  $E_{NO}$  was 15.62 kWh kg<sup>-1</sup>.

With more NH<sub>3</sub> molecules induced into the reaction system, the total quantity of collision particles increased, and more active free radicals such as NH<sub>2</sub><sup>·</sup>, NH<sup>·</sup>, H<sup>·</sup> were also



**Figure 3.** (a) The effect of NH<sub>3</sub> on the removal efficiency of NO and energy consumption. (b) The effect of different NH<sub>3</sub> concentration.

involved in the reaction process, which could significantly contribute to the removal of NO molecules (reactions R3, R4 and R5). However, when the NH<sub>3</sub> concentration was more than 270 ppm, superfluous ammonia molecules could be oxidized by oxygen, which could lead to NO formation (reactions R6 and R7).

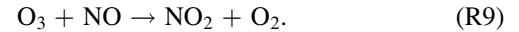


Consequently, the removal efficiency of NO could decrease slightly and  $E_{\text{NO}}$  increased when too many NH<sub>3</sub> molecules were led into the reactor. We could achieve an attractive removal efficiency with low energy consumption under atmospheric pressure and low-temperature conditions when the concentration of NH<sub>3</sub> varied from 90 ppm to 270 ppm. The gas concentration ratio of NH<sub>3</sub>/NO at 0.23–0.67 proved to be very attractive.

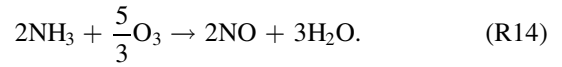
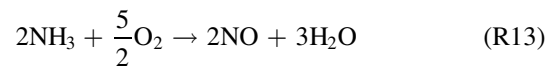
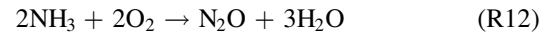
### 3.2. Oxygen (O<sub>2</sub>) effects

Figure 4(a) shows the relationship between applied voltage and removal efficiency of NO under different percentages of oxygen (0%, 2% and 8%) conditions. The initial concentration of NO and NH<sub>3</sub> were 400 ppm and 90 ppm, respectively. The applied voltage varied from 2 kV to 7 kV. Figure 3(b) shows NO and NO<sub>2</sub> concentration under the same conditions.

The data shown in figure 4(a) indicate that the removal efficiency of NO increased gradually with the increase in applied voltage, and decreased with the oxygen concentration under the same applied voltage. In the presence of oxygen, the DBD process produces ozone and degrades NO

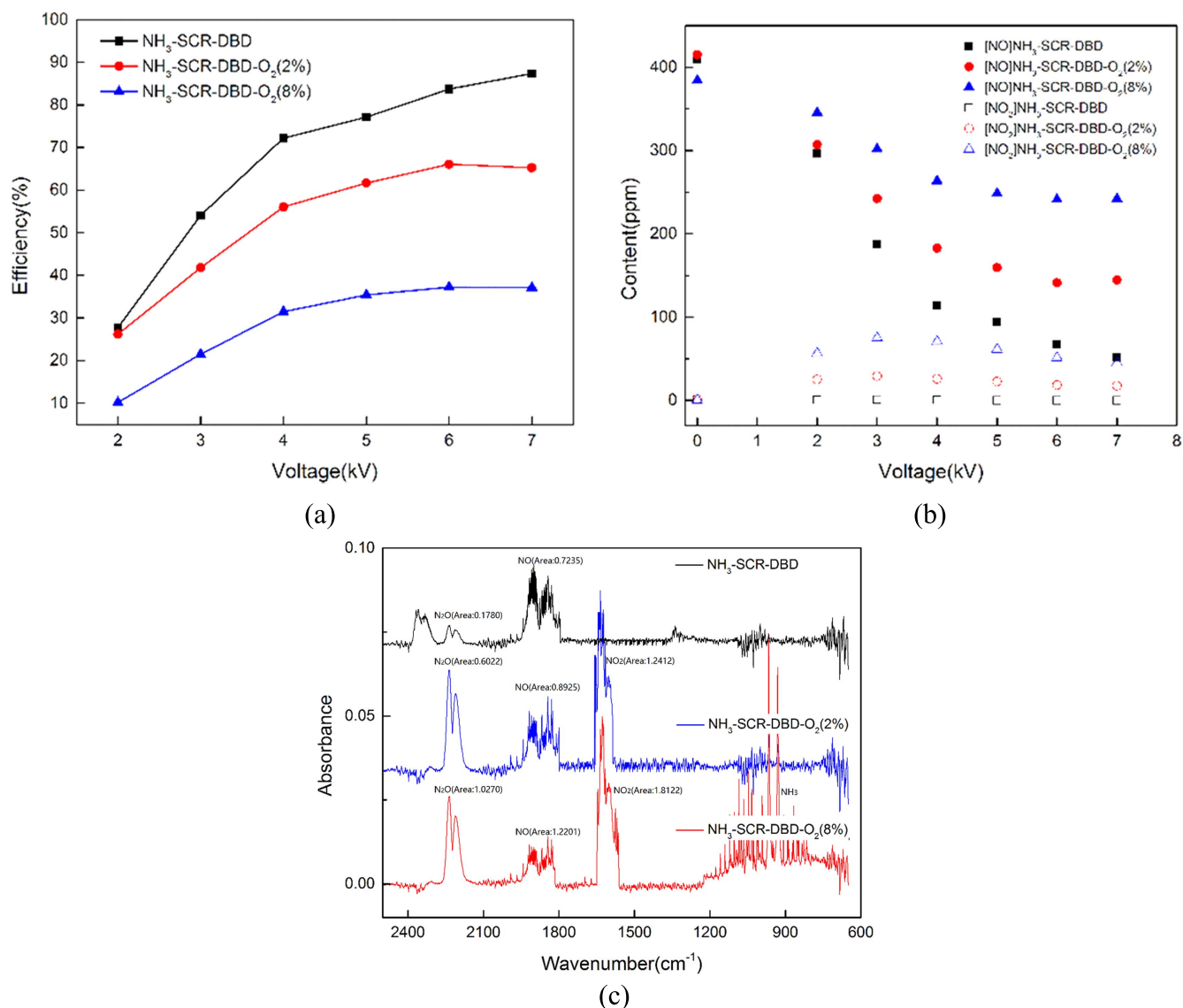


However, from the results, the NO removal process was suppressed because NO and NH<sub>3</sub> were partially oxidized to new NO and NO<sub>2</sub>. Therefore, the removal efficiency of NO decreased. The main reactions occurred in the presence of O<sub>2</sub> and O<sub>3</sub> and are listed as follows:



Therefore, when more oxygen molecules were induced into the reaction system, lots of active radicals such as  $\cdot\text{O}$  were generated. Then, large quantities of NH<sub>3</sub> were oxidized to NO molecules, which acted as a negative effect on NO removal.

As shown in figure 4(b), the concentration of NO decreased quickly and NO<sub>2</sub> increased slightly in the presence of O<sub>2</sub> molecules when the applied voltage was below 3 kV. Then, with the applied voltage continually increasing, the NO and NO<sub>2</sub> concentrations decreased slightly. Initially, under the low O<sub>2</sub> concentration, lots of NO molecules were degraded (as shown in R3 to R5). However, higher O<sub>2</sub> concentration led to NO<sub>2</sub> molecule formation, and, at the same time, with higher applied voltage, the electric field of the reaction system was strengthened, and more NH<sub>3</sub> molecules participated in the reactions with O<sub>2</sub> and  $\cdot\text{O}$ , which led to new



**Figure 4.** (a) Effects of different oxygen concentration on NO removal efficiency. (b) Effects of different oxygen concentration on NO and NO<sub>2</sub> content. (c) Final products observed using FTIR under different oxygen conditions.

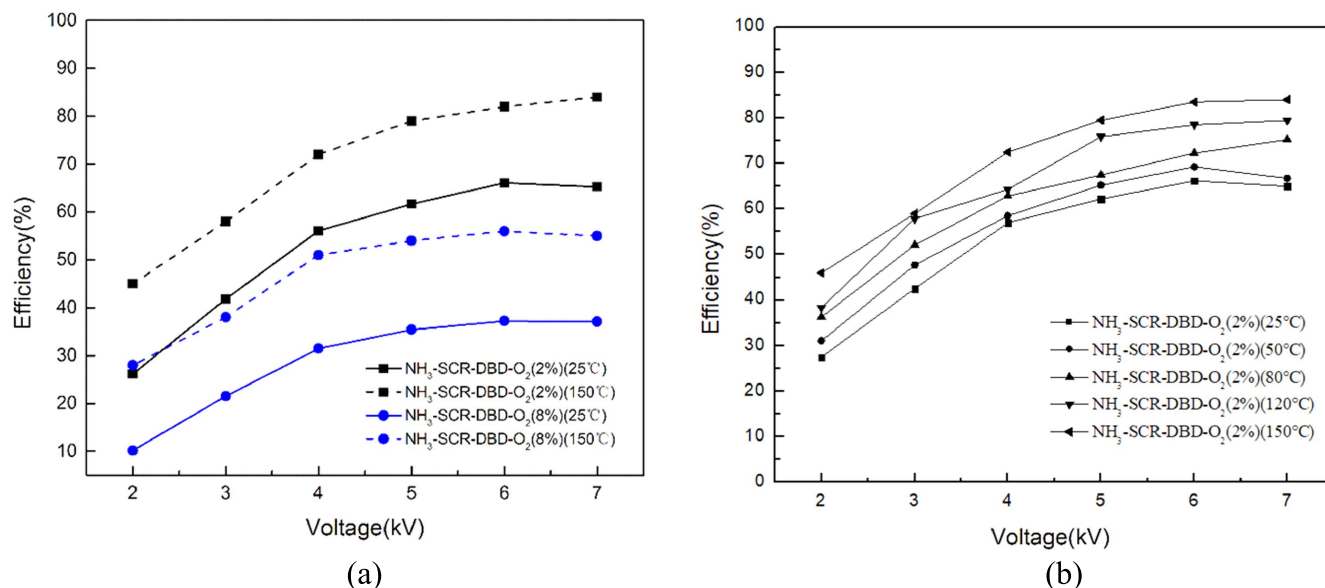
**Table 1.** Principle reactions involved.

	NO	NO <sub>2</sub>
Applied voltage ≤ 3 kV	$\text{NO} + \text{O}\cdot \rightarrow \text{O}_2 + \text{N}_2$ $\text{NO} + \cdot\text{OH} \rightarrow \text{HNO}_2$ $\text{NO} + \cdot\text{OH} \rightarrow \text{HNO}_2$ $4\text{NH}_3 + 4\text{NO} + \text{O}_2 \rightarrow 4\text{N}_2 + 6\text{H}_2\text{O}$ $4\text{NH}_3 + 4\text{NO} + 3\text{O}_2 \rightarrow 4\text{N}_2\text{O} + 6\text{H}_2\text{O}$	$\text{NO} + \cdot\text{O} \rightarrow \text{NO}_2$
Applied voltage > 3 kV	$\text{NH} + \cdot\text{O} \rightarrow \text{H}\cdot + \text{NO}$ $\text{NH} + \text{O}_2 \rightarrow \cdot\text{OH} + \text{NO}$ $\text{NO}_2 + \cdot\text{O} \rightarrow \cdot\text{OH} + \text{NO}$	$\text{NH} + \text{NO}_2 \rightarrow \text{N}_2\text{O} + \cdot\text{OH}$ $\text{NH} + \text{NO}_2 \rightarrow \text{HO}_2\cdot + \text{N}_2$ $\text{NH} + \text{NO}_2 \rightarrow \text{HNO}\cdot + \text{NO}$

NO molecule production through R6, R7 and R10. The principle reactions are listed in table 1.

Figure 4(c) shows the outlet gas components observed by FTIR under different oxygen conditions at fixed applied voltage of 3 kV. It indicates that the more oxygen was led into

the gas stream, the higher the concentrations of N<sub>2</sub>O, NO and NO<sub>2</sub> that were observed. The results also indicated that with higher concentration of O<sub>2</sub>, NO removal efficiency decreased, but N<sub>2</sub>O and NO<sub>2</sub> were increased by the oxidizing reactions of NO and NH<sub>3</sub> molecules in the reactor.



**Figure 5.** (a) Effects of temperature on NO removal efficiency under different oxygen content. (b) Effects of temperature on NO removal efficiency at 2% oxygen content.

### 3.3. Temperature effects

Figure 5(a) shows the relationship of NO removal efficiency and applied voltage under different oxygen content (2% and 8%) and temperature (25 °C and 150 °C), and figure 5(b) shows the relationship of NO removal efficiency and applied voltage under different temperatures (25 °C, 50 °C, 80 °C, 120 °C and 150 °C) at 2% oxygen content. The initial concentrations of NO and NH<sub>3</sub> were 400 ppm and 90 ppm, respectively. The applied voltage varied from 2 kV to 7 kV.

The data in figure 5 show that higher temperature conditions were good for the removal efficiency of NO. Under the applied voltage of 7 kV,  $\eta_{\text{NO}}$  was improved from 65% to 84% and 37% to 55% in the presence of O<sub>2</sub> of 2% and 8%, respectively. NO removal was influenced by temperature for two main reasons:  $E/N$  (electrical field strength divided by the total gas density) and reaction rate [34].  $E/N$  increases when the electric field strength ( $E$ ) is proportional to the applied voltage if the plasma reactor parameters remain constant and the gas density  $N$  decreases with the increase in the temperature [35]. A rising  $E/N$  means that more energy is transferred to the particles, which promotes the ionization and excitation process. Then, more active species such as  $\cdot\text{O}$ ,  $\cdot\text{H}$ ,  $\cdot\text{OH}$ , and  $\text{NH}\cdot$  are induced into the reactor system and play a role in NO removal. At high temperature conditions, electron detachment becomes significant so that radicals of  $\cdot\text{O}$ ,  $\cdot\text{H}$ ,  $\cdot\text{OH}$ , and  $\text{NH}\cdot$  have more negligible effects than their anionic counterparts [36].

Meanwhile, temperature affects the reaction rate coefficients directly, which dominate the chemical kinetic in gas phase reactions. The main reactions involved are shown in table 2.

Among those reactions in table 2, compared to the data at temperatures of 25 °C, the larger rate coefficients of reactions of R1–R2 and R14–R17 at a temperature of 150 °C contribute

to the reactions of active radicals with NO. In this system, the removal of NO mainly depends on reactions R17–R22, R3 and R5. More active radicals such as HO<sub>2</sub> $\cdot$  and  $\cdot\text{O}$ , contribute significantly to the conversion of NO into NO<sub>2</sub> [37]. Moreover, with increasing the temperature, the rate constants of R24 and R25 were decreased, resulting in less NO molecule formation. In general, the generation of active species and the rate coefficients of NO removal reactions were both promoted with increased temperature, and then contributed to  $\eta_{\text{NO}}$  considerably. In addition, previous studies [17] showed that at temperatures of about 100 °C, the catalytic activity of the V<sub>2</sub>O<sub>5</sub>–WO<sub>3</sub>/TiO<sub>2</sub> catalyst increases with the increase in temperature; it also promoted the removal of NO.

### 3.4. Humidity effects

Figure 6 shows the effect of water vapor on NO removal under varied applied voltages of 2–7 kV. The initial concentrations of NO and NH<sub>3</sub> were set at 400 ppm and 90 ppm, respectively. All experiments were conducted at room temperature of 25 °C. The relative humidity in our DBD reactor was 85% in the presence of H<sub>2</sub>O.

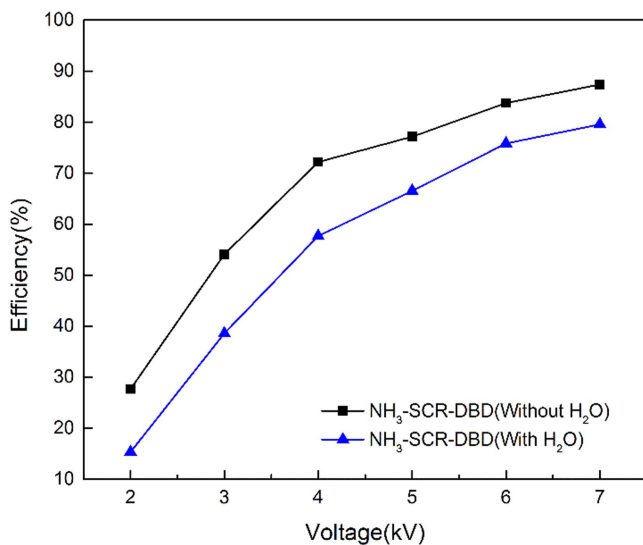
The results shown in figure 6 indicate that the NO removal efficiency was only 57% in the presence of H<sub>2</sub>O, but increased to 72% without H<sub>2</sub>O under an applied voltage of 4 kV. In other words, H<sub>2</sub>O has an obvious suppression effect on NO removal. The reasons for this phenomenon might be considered as follows.

Firstly, with the addition of H<sub>2</sub>O, the electronegative molecules lead to large quantities of electrons absorbed, which decrease the discharge power. In the experiment process we found that the frequency was 14.0 kHz without H<sub>2</sub>O and 10.5 kHz after adding H<sub>2</sub>O. Previous studies [38] have shown that with H<sub>2</sub>O addition, the number of high-energy electrons decreases, which indicates that the collisions

**Table 2.** Major reactions and their corresponding rate coefficients.

Reactions	Rate coefficients (cm <sup>3</sup> s <sup>-1</sup> )			Reaction number
	25 °C (298 K)	150 °C (423 K)	Formula	
e + O <sub>2</sub> → e + ·O + ·O			f(E/N)	R15
e + H <sub>2</sub> O → ·H + ·OH + e			f(E/N)	R16
e + H <sub>2</sub> O → H <sup>-</sup> + OH			f(E/N)	R17
e + N <sub>2</sub> → e + ·N + ·N			f(E/N)	R18
O <sub>2</sub> + ·O → O <sub>3</sub>	6.0 × 10 <sup>-34</sup>	9.0 × 10 <sup>-34</sup>	6.0 × 10 <sup>-34</sup> exp(T/300) <sup>-2.6</sup>	R8
O <sub>2</sub> + H → HO <sub>2</sub> ·	9.5 × 10 <sup>-11</sup>	1.1 × 10 <sup>-10</sup>	9.47 × 10 <sup>-11</sup> (T/298) <sup>0.44</sup>	R19
·O + H <sub>2</sub> O → ·OH + ·OH	7.3 × 10 <sup>-24</sup>	4.1 × 10 <sup>-20</sup>	1.84 × 10 <sup>-11</sup> (T/298) <sup>0.95</sup> exp(-71 255/RT)	R20
·OH + NH <sub>3</sub> → H <sub>2</sub> O + ·NH <sub>2</sub>	1.6 × 10 <sup>-13</sup>	4.0 × 10 <sup>-13</sup>	3.5 × 10 <sup>-12</sup> (T/298)exp(-7691/RT)	R1
NH <sub>3</sub> + ·O → ·OH + ·NH <sub>2</sub>	4.4 × 10 <sup>-17</sup>	7.5 × 10 <sup>-16</sup>	2.9 × 10 <sup>-13</sup> (T/298) <sup>2.1</sup> exp(-21 784/RT)	R2
NO removal				
NO + ·N → N <sub>2</sub> + ·O	2.1 × 10 <sup>-11</sup>	3.1 × 10 <sup>-11</sup>	8.2 × 10 <sup>-11</sup> exp(-3409/RT)	R21
O <sub>3</sub> + NO → O <sub>2</sub> + NO <sub>2</sub>	1.8 × 10 <sup>-14</sup>	6.3 × 10 <sup>-14</sup>	1.4 × 10 <sup>-12</sup> exp(-1310/T)	R22
HO <sub>2</sub> + NO → NO <sub>2</sub> + ·OH	2.7 × 10 <sup>-13</sup>	1.2 × 10 <sup>-12</sup>	3.3 × 10 <sup>-11</sup> exp(-11 890/RT)	R23
·O + NO → NO <sub>2</sub>	3.0 × 10 <sup>-11</sup>	3.3 × 10 <sup>-11</sup>	3.0 × 10 <sup>-11</sup> (T/298) <sup>0.6</sup>	R24
·NH + NO → N <sub>2</sub> O + ·H	2.9 × 10 <sup>-11</sup>	3.1 × 10 <sup>-11</sup>	1.17 × 10 <sup>-10</sup> (T/298) <sup>-1.03</sup> exp(-3492/RT)	R25
·OH + NO → HNO <sub>2</sub>	7.4 × 10 <sup>-31</sup>	3.2 × 10 <sup>-31</sup>	7.4 × 10 <sup>-31</sup> (T/300) <sup>-2.4</sup>	R26
NO + NH <sub>2</sub> → N <sub>2</sub> + H <sub>2</sub> O	1.2 × 10 <sup>-11</sup>	9.0 × 10 <sup>-12</sup>	2.07 × 10 <sup>-11</sup> (T/298) <sup>-1.61</sup> exp(-1247/RT)	R3
·NH + NO → ·OH + N <sub>2</sub>	4.8 × 10 <sup>-12</sup>	4.3 × 10 <sup>-12</sup>	5.86 × 10 <sup>-12</sup> (T/298) <sup>-0.5</sup> exp(-499/RT)	R5
NO formation				
·O + NO <sub>2</sub> → NO + O <sub>2</sub>	9.7 × 10 <sup>-12</sup>	8.6 × 10 <sup>-12</sup>	6.51 × 10 <sup>-12</sup> exp(998/RT)	R27
NH <sub>2</sub> + HNO → NH <sub>3</sub> + NO	3.6 × 10 <sup>-12</sup>	2.9 × 10 <sup>-12</sup>	1.1 × 10 <sup>-12</sup> (T/298) <sup>0.41</sup> exp(2935/RT)	R28
O <sub>2</sub> + ·NH → ·OH + NO	9.0 × 10 <sup>-15</sup>	1.9 × 10 <sup>-14</sup>	6.74 × 10 <sup>-14</sup> (T/298) <sup>0.79</sup> exp(-4997/RT)	R7
·O + NH <sub>2</sub> → H <sub>2</sub> + NO	2.1 × 10 <sup>-13</sup>	2.3 × 10 <sup>-13</sup>	7.42 × 10 <sup>-14</sup> (T/298) <sup>1.02</sup> exp(2627/RT)	R29
NH <sub>3</sub> + NO <sub>2</sub> → HNO <sub>2</sub> + ·NH <sub>2</sub>	2.2 × 10 <sup>-37</sup>	1.5 × 10 <sup>-31</sup>	1.11 × 10 <sup>-15</sup> (T/298) <sup>3.41</sup> exp(-12 4717/RT)	R30

The rate coefficients were obtained from the NIST chemical kinetics database.



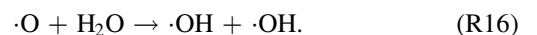
**Figure 6.** Effects of water on NO removal efficiency.

between electrons and molecules decrease. As fewer electrons are produced, the electric field of the discharge system decreases, and the energy is not high enough to break the bond of neutral molecules such as H<sub>2</sub>O and O<sub>2</sub>. Furthermore, more H<sub>2</sub>O molecules leads to fewer vital active species such as HO·, HO<sub>2</sub>· and ·O [39].

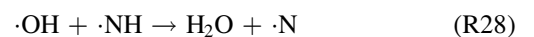
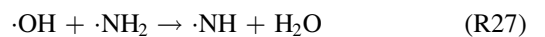
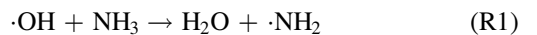
Secondly, the formation of active particles induced by the collision between electrons and H<sub>2</sub>O molecules such as ·H

and ·OH lowers electron abundance. Therefore, the remainder of the electrons do not reach the energy to produce HO<sub>2</sub>· and ·O, and the number of active species participating in the NO reduction reaction are reduced, which leads to a decrease in NO removal efficiency.

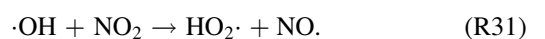
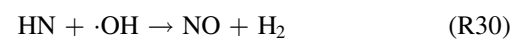
Thirdly, in the presence of H<sub>2</sub>O, more ·OH radicals are produced:



The ·OH radicals react with NH<sub>3</sub>:



and the reaction of H<sub>2</sub>O with N is intensified by producing more ·HN to contribute to NO formations via the following reactions:



In addition, previous work [40] showed that the presence of H<sub>2</sub>O has an inhibiting effect on the catalyst activity, which is caused by competitive adsorption of H<sub>2</sub>O and the reactants such as NH<sub>3</sub> and/or NO.

As demonstrated above, water plays a negative effect on NO removal in high relative humidity conditions.


#### 4. Conclusions

Our study mainly investigated NO removal using a new integrated NH<sub>3</sub>-SCR-DBD reactor with ammonia (NH<sub>3</sub>) as a reductant. Catalysts (V<sub>2</sub>O<sub>5</sub>-WO<sub>3</sub>/TiO<sub>2</sub>) were inserted in the discharge area. In this paper, the effects of NH<sub>3</sub>, O<sub>2</sub>, temperature and H<sub>2</sub>O on NO removal were discussed. Results showed that the NO removal efficiency increases with the increase in NH<sub>3</sub> and temperature, but decreased with the increase in O<sub>2</sub> and H<sub>2</sub>O. Meanwhile, when the gas concentration ratio of NH<sub>3</sub>/NO was 0.23–0.67, we obtained acceptable NO removal efficiency with low energy consumption. When the temperature increased to 150 °C, more active species such as ·O, ·H, ·OH and NH· were produced, which promoted NO removal. In the presence of H<sub>2</sub>O, the discharge state was suppressed and ·OH generated from the dissociation of H<sub>2</sub>O led to new NO formation. The combination of NH<sub>3</sub>-SCR assisted by the DBD reactor method was demonstrated to be a very attractive and promising method for NO removal application.

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