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# Removal of ammonia from gas streams with dielectric barrier discharge plasmas

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#### Abstract

We reported on the experimental study of gas-phase removal of ammonia (NH<sub>3</sub>) via dielectric barrier discharge (DBD) at atmospheric pressure, in which we mainly concentrated on three aspects—influence of initial NH<sub>3</sub> concentration, peak voltage, and gas residence time on NH<sub>3</sub> removal efficiency. Effectiveness, e.g. the removal efficiency, specific energy density, absolute removal amount and energy yield, of the self-made DBD reactor had also been studied. Basic analysis on DBD physical parameters and its performance was made in comparison with previous investigation. Moreover, products were detected via ion exchange chromatography (IEC). Experimental results demonstrated the application potential of DBD as an alternative technology for odor-causing gases elimination from gas streams. © 2007 Elsevier B.V. All rights reserved.

Keywords: Ammonia; Dielectric barrier discharge; Plasma; Specific energy density; Energy yield

## 1. Introduction

Ammonia (NH<sub>3</sub>) is a common inorganic odor-causing substance existing in the atmospheric environment. There are numerous sources that may emit NH<sub>3</sub> with a wide range of concentration, such as integrated coal gasification combinedcycle power generation (IGCC) systems, landfills for waste disposal, crematory, livestock farming, hog manuring, and wastewater treatment plants, etc. [1,2]. Emission of NH<sub>3</sub> without appropriate treatment is causing frequent complaints from neighboring communities. With increasing concerns on environment pollution, removal of NH<sub>3</sub> from waste streams is becoming important. Conventional methods for NH<sub>3</sub> removal from gas streams include absorption (wet scrubbing), adsorption, incineration (either thermal or catalytic), and biofiltration [3]. Nevertheless, all these methods may have technical and/or economic limitations in removing NH<sub>3</sub> from gas streams. Additionally, as the emission standards of these odor-causing substances become more stringent, the cost required to control odor problems accordingly escalates. As a result, there

0304-3894/\$ - see front matter © 2007 Elsevier B.V. All rights reserved. doi:10.1016/j.jhazmat.2007.06.070 is motivation to develop technologies, which can be accepted both economically and technically, to control  $NH_3$  problem.

In recent years, technology based on non-thermal plasmas has received considerable attention in the field of air purification due to its advantages such as low costs, high removal rate and high energy efficiency. Non-thermal plasma is classified as five categories by generating mode, pressure range and electrode geometry [4]: (a) glow discharge plasma; (b) corona discharge plasma; (c) dielectric barrier discharge (DBD) plasma; (d) radio frequency (RF) discharge plasma; (e) microwave discharge plasma. DBD was chosen for this study because of its easy operation and high efficiency in generating gas phase radicals. It is initially used as a gas discharge process to generate O<sub>3</sub> for industrial application [5]. As one kind of cold plasma and silent discharge process, it has also been widely studied in recent years for its potential in removing odor-causing gases such as SO<sub>2</sub>, H<sub>2</sub>S and NH<sub>3</sub>, as well as organic compounds such as benzene, trichloroethylene, formaldehyde and styrene from gas stream [6-13]. Couples of reports, together with our previous experiments [11,14,15] have shown the odors can be treated efficiently in DBD plasma reactor. Chang and Tseng [3], who utilized DBD plasma for removal of H<sub>2</sub>S and NH<sub>3</sub>, speculated that higher removal efficiency would

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Nomenclature			
ARA DBD EY [NH <sub>3</sub> ] ηNH <sub>3</sub> P Q	absolute removal amount of NH <sub>3</sub> ( $\mu$ g/s) dielectric barrier discharge energy yield (g/kWh) concentration of NH <sub>3</sub> (mg/m <sup>3</sup> ) NH <sub>3</sub> removal efficiency (%) power (W) gas flow rate (l/s)		
SED	specific energy density (J/l)		

be expected if using higher frequency power supply. The inspired prospect also contributes to direct the present investigation.

In this paper, a self-made DBD plasma reactor, characterized by its high frequency and high energy yield [16], was utilized to generate plasma to remove NH<sub>3</sub> from gas streams under room temperature at atmospheric pressure. The atmospheric destruction of NH<sub>3</sub> via DBD plasma, with the presence of air, depends on two removal mechanisms including (a) direct removal caused by the collision of electrons with NH<sub>3</sub> and (b) reaction between NH<sub>3</sub> and gas-phase radicals. Gas-phase NH<sub>3</sub> removal efficiency and energy yield as function of relevant controlling parameters of DBD were described, followed by comparison with previous investigations and reaction mechanism discussion.

## 2. Experiment and measurement

#### 2.1. Experimental setup

The experimental system designed for evaluating the efficiency of DBD plasmas on NH<sub>3</sub> removed from simulated gas-streams is shown in Fig. 1. The system consisted of a continuous flow gas generation system, a self-made DBD reactor, and a gas sampling and detection system. NH<sub>3</sub> vapor was generated by bubbling an aqueous solution of ammonia (25-28%, v/v) with compressed air. It was then allowed to pass through a gas mixer for a thorough mixture with air before being introduced into the DBD reactor. The gas stream should pass through the reactor for 30 min to allow the system to reach steady state. Gas flow rate and initial concentration of NH<sub>3</sub> were monitored, and then power was applied to generate plasma. Discharge process continued for 30 min to make sure that new steady state conditions had been achieved before the measurement of resulting NH<sub>3</sub> concentration in the gas stream. NH<sub>3</sub> concentration ranging from 30 to 250 mg/m<sup>3</sup>, peak voltage ranging from 3000 to 9000 V, and gas residence time ranging from 0.04 to 0.2 s were achieved by regulating the rotameter, high voltage power supply and tunable air blower, respectively. All the tests were completed at atmospheric pressure and ambient temperature  $(25 \pm 2^{\circ}C)$ .

#### 2.2. DBD reactor

The laboratory-scale DBD reactor was made of two concentric quartz tubes with wall thickness of 1.5 mm and length of 200 mm as barriers. The inner one had an outside diameter of 10 mm, while the outer one had an inside diameter of 33 mm. A sheet of stainless foil attached tightly to the inside wall of the inner tube serving as an electrode, while the other electrode made of a four-mm-wide stainless strip was wrapped around the outside of the external tube. Plasma was sustained within a volume of 155 cm<sup>3</sup> by applying an alternating-current power operated at a fixed frequency of 20 kHz, with risetime of 10 ns. A visible discharge was generally achieved with a breakdown voltage of 3000 V.

## 2.3. Chemical analysis

NH<sub>3</sub> concentration ([NH<sub>3</sub>]) before and after DBD treatment was determined via spectrometric method [17–19]. The ammonia in gas stream should be first absorbed in boracic acid (H<sub>3</sub>BO<sub>3</sub>) solution, in which ammonia reacts with hypochlorite to form chloramines. Chloramines then react with phenol to form the intermediate, named monochloroquinoimine. Finally, this intermediate couples with a second phenolic molecule to form indophenol blue. [NH<sub>3</sub>] was then determined by measuring light absorbance at 640 nm with a spectrophotometer (Model 721, Shanghai Precision & Scientific Instrument Co., Ltd.). The plot of absorbance with the amount of NH<sub>3</sub> in solution obeyed a linear relationship with a correlation coefficient of 0.998.



Fig. 1. Gas-generation system, DBD reactor, and gas-sampling and detection system.



Fig. 2.  $\eta$ NH<sub>3</sub> and ARA as a function of gas residence time. The peak voltage was 9000 V.

### 3. Results and discussion

#### 3.1. Effects of gas residence time

Fig. 2 presented the dependence of  $\eta$ NH<sub>3</sub> (solid line) and ARA (dashed line) on gas residence time ranging from 0.04 to 0.2 s. ARA is the absolute removal amount of NH<sub>3</sub> (ARA =  $C\eta Q$ ,  $\mu g/s$ ; *C* is inlet [NH<sub>3</sub>], mg/m<sup>3</sup>;  $\eta$  is the removal efficiency; *Q* is gas flow rate, l/s). The peak voltage was controlled at 9000 V. With inlet [NH<sub>3</sub>] of 250 mg/m<sup>3</sup>,  $\eta$ NH<sub>3</sub> increased with increasing gas residence time from 0.04 to 0.1 s, and reached equilibrium as residence time increased further to 0.2 s. Longer residence time provides longer time for NH<sub>3</sub> molecules to be attacked by electrons or radicals, resulting in higher  $\eta$ NH<sub>3</sub>. ARA reached its maximum at residence time of 0.1 s. The similar trends were observed with inlet [NH<sub>3</sub>] of 125 mg/m<sup>3</sup>, except that the maximum of ARA appeared at residence time of 0.067 s. The results showed that DBD technology was efficient in eliminating NH<sub>3</sub> even when the residence time was as short as 0.1 s.

SED and EY were proposed in order to describe energy efficiency of DBD reactor in NH<sub>3</sub> removal. SED is the specific energy density (SED = P/Q, J/l; P is discharge power, W). EY, which is a well-accepted parameter to indicate the energy efficiency [20–22], is the energy yield (EY = 3.6 $C\eta$ /SED, g/kWh) of the DBD reactor. Table 1 showed SED and EY as function of gas residence time. With increasing of gas residence time,  $\eta$ NH<sub>3</sub> was enhanced because of longer time for collisions between NH<sub>3</sub>

Table 1 Energy utilization of DBD vs. gas residence time (9000 V, 122.41 W)

Gas residence time (s)	Q (1/s)	SED (J/l)	EY (g/kWh)	
			125 mg/m <sup>3</sup>	250 mg/m <sup>3</sup>
0.04	3.88	31.6	2.3	2.9
0.05	3.10	39.4	3.6	3.6
0.067	2.33	52.6	4.6	6.4
0.1	1.55	78.9	4.4	7.9
0.2	0.78	157.7	2.4	4.2

molecule and electron/radical. SED was also enhanced because of the reduced flux of inlet gas, which resulted in lower collision frequency of drifting electrons with gas molecules. As a result, both  $\eta$ NH<sub>3</sub> and SED increased with increasing gas residence time, which was individually of positive and negative effect to EY at fixed inlet [NH<sub>3</sub>]. As demonstrated by Table 1, EY increased first and then decreased if extending the gas residence time continuously. This indicated the balance between collision time and collision probability. Collision time played a more important role with gas residence time ranging from 0.04 to 0.1 s while collision probability hold a dominant position from 0.1 to 0.2 s with inlet [NH<sub>3</sub>] of 250 mg/m<sup>3</sup>. Similar trend was also observed with inlet [NH<sub>3</sub>] of 125 mg/m<sup>3</sup> with turning point of 0.067 s.

### 3.2. Effects of inlet [NH<sub>3</sub>]

It should be noted from Fig. 2 that with inlet [NH<sub>3</sub>] decreasing half from 250 to 125 mg/m<sup>3</sup>,  $\eta$ NH<sub>3</sub> increased relatively slightly (about 10%) while ARA increased remarkably at same gas residence time. The experimental results demonstrated that lower inlet [NH<sub>3</sub>] was a favorable factor to NH<sub>3</sub> removal efficiency but an unfavorable one to ARA. Further investigation (Fig. 3) was conducted for a specific study on the dependence of  $\eta NH_3$ and ARA on inlet [NH<sub>3</sub>] at fixed peak voltage. Inlet [NH<sub>3</sub>] was varied from 30 to 250 mg/m<sup>3</sup> while the gas residence time and peak voltage were fixed at 0.067 s and 9000 V, respectively. Experimental results demonstrated that  $\eta NH_3$  decreased with increasing inlet [NH<sub>3</sub>], while ARA grew greatly first and gradually slowed down when inlet  $[NH_3]$  was higher than 200 mg/m<sup>3</sup>. The opposite dependence of  $\eta$ NH<sub>3</sub> and ARA on inlet [NH<sub>3</sub>] suggests appropriate reaction conditions should be chosen to meet the practical requirements in industrial application of DBD technology. Additionally,  $\eta NH_3$  reached 90.9% when inlet [NH<sub>3</sub>] was as low as 30 mg/m<sup>3</sup>, indicating that DBD is very effective in removing gas wastes even when the concentration is comparatively low.



Fig. 3.  $\eta$ NH<sub>3</sub> and absolute removal amount as a function of inlet [NH<sub>3</sub>]. The peak voltage was 9000 V; residence time was 0.067 s, and gas flow rate was 2.33 l/s.

Table 2 Energy utilization of DBD vs. inlet [NH<sub>3</sub>] (9000 V, 122.41 W, 0.067 s, 2.33 l/s, 52.58 J/l)

Inlet [NH <sub>3</sub> ] (mg/m <sup>3</sup> )	EY (g/kWh)
30	2.0
50	2.5
100	3.9
125	4.6
150	5.1
200	6.3
250	6.4

As shown in Table 2, EY increased monotonically from 2.0 g/kWh at inlet [NH<sub>3</sub>] of 30 mg/m<sup>3</sup> to 6.4 g/kWh at inlet  $[NH_3]$  of 250 mg/m<sup>3</sup>. Positive dependence of EY on inlet  $[NH_3]$ observed in our experiment was in good agreement with that reported by Ruan et al. [20]. At a fixed SED, the number of NH3 molecules increased along with increasing initial concentration. However, the specific energy density in the DBD reactor did not increase accordingly, which reduced the removal efficiency. On the other hand, since the lifetime of gas-phase radicals and energetic electrons, on the orders of magnitude of nanoseconds [23], is much shorter than the typical gas residence time in the DBD reactor. More NH<sub>3</sub> molecules favor more efficient use of these radicals and energetic electrons, resulting in higher EY and thus higher ARA. Therefore, although the removal efficiency reduced with increasing inlet concentration, the energy utilization increased.

## 3.3. Effects of peak voltage

Experimental tests were at last conducted to determine the dependence of  $\eta$ NH<sub>3</sub> and ARA on peak voltage. It is assumed that both the number and average energy of the electrons rise with enhancing applied voltage [23], which leads to a higher specific energy density and correspondingly a higher removal efficiency at higher applied voltage. The expected positive dependence of  $\eta$ NH<sub>3</sub> on peak voltage was observed experimentally over the range from 3000 to 9000 V as shown in Fig. 4.



Fig. 4.  $\eta$ NH<sub>3</sub> and ARA as a function of peak voltage. The inlet [NH<sub>3</sub>] was 50 mg/m<sup>3</sup>, residence time was 0.067 s, and gas flow rate was 2.33 l/s.

Table 3 Energy utilization of DBD vs. peak voltage (50 mg/m<sup>3</sup>, 0.067 s, 2.33 l/s)

Voltage (V)	Power (W)	SED (J/l)	EY (g/kWh)
3000	16.7	7.2	9.0
4500	33.0	14.2	5.5
6000	56.0	24.1	3.5
7500	85.8	36.9	3.1
9000	122.4	52.6	2.5

With inlet [NH<sub>3</sub>] of 50 mg/m<sup>3</sup> and gas residence time of 0.067 s,  $\eta$ NH<sub>3</sub> increased from 35.9% at peak voltage of 3000 V to 72.8% at peak voltage of 9000 V. ARA changed in the same manner as  $\eta$ NH<sub>3</sub>, since it was only related to removal efficiency as inlet [NH<sub>3</sub>] and gas residence time were both fixed.

Strictly speaking, influence of temperature should also be taken into account with respect to the dependence of  $\eta NH_3$  on various peak voltages. When plasma system was held for an enough long period, temperature of the whole plasma zone was detected to increase from 150 °C to 250 °C with peak voltage raising from 3000 V to 9000 V. At high peak voltage, the resulted higher temperature may promote decomposition of NH<sub>3</sub>, which also contribute to higher removal efficiency [2].

Energy utilization of DBD as a function of peak voltage was presented in Table 3. As shown in Table 3, SED increased monotonically while EY of DBD decreased with the increasing peak voltage. The phenomenon was identical to the report of Zheng et al. [16]. When the peak voltage was above breakdown voltage, low voltage was advantageous to make high EY. It could probably be explained by higher energy consumed in moving electrons/particles at higher peak voltage, other than on target gas removal. As is well known, plasma requires energy to keep electrons and other particles moving. As the voltage increased, more energy is required. Consequently, higher energy consumption on moving electrons/particles limited the energy yield on high peak voltage.

#### 3.4. Comparison with prior studies

NH<sub>3</sub> was reported previously to be removed by coronadischarge plasmas (either non-pulse or pulse) [2,20,24], microwave plasmas [25], dielectric barrier discharge [3,26]. A comparison was made between the present work and two of these previous reports for close study of the dependence of odorcausing gas removal efficiency and DBD energy yield on DBD physical parameters, and thus probably directs future investigation. The comparison (Table 4) was based on key parameters such as removal efficiency ( $\eta$ NH<sub>3</sub>, ARA), specific energy density (SED) and energy utilized efficiency (EY).

Apparently, we achieved similar removal efficiency with lower peak voltage and shorter residence time. Chang and Tseng [3] achieved  $\eta$ NH<sub>3</sub> of 75% with peak voltage up to 17.0 kV and gas residence time of 9.6 s at inlet [NH<sub>3</sub>] of 100 mg/m<sup>3</sup>. Ruan et al. [20] achieved  $\eta$ NH<sub>3</sub> of 87% with peak voltage as high as 70 kV and gas residence time as long as 12.96 s at inlet [NH<sub>3</sub>] of 110 mg/m<sup>3</sup>, while our experiment achieved removal efficiency of 77.9% with peak voltage of only 9 kV and gas res-

Table 4				
Comparison between	previous	work an	d our	work

	Chang and Tseng [3]	Ruan et al. [20]	This study
Reaction condition			
Plasma	DBD	Pulse corona	DBD
Inlet $[NH_3] (mg/m^3)$	100	110	125
Gas residence time (s)	9.6 <sup>b</sup>	12.96 <sup>b</sup>	0.1
Peak voltage (kV)	17	70	9
$Q (m^3/h)$	0.09 <sup>b</sup>	10	8.38
SED (J/l)	_a	106	78.9
Reaction result			
ηNH <sub>3</sub> (%)	About 75 <sup>b</sup>	87	77.9
ARA (mg/h)	About 8.9 <sup>b</sup>	960	543.7
EY (g/kwh)	_a	2.99	4.5
Plasma physical parameters			
Dielectric	Pyrex		Quartz
Dielectric thickness (mm)	2	_a	1.5
Frequency (Hz)	60	_a	20000
Gas spacing thickness (mm)	14.8	a	11.5

<sup>a</sup> No original data.

<sup>b</sup> Calculated by original data.

idence time of only 0.1 s at inlet [NH<sub>3</sub>] of 125 mg/m<sup>3</sup>. Lower applied voltage would reduce the burden of the equipment and thus prolong its service lifetime. It was also noteworthy that relatively high absolute removal amount of NH<sub>3</sub> and high energy yield were achieved with short gas residence time in the present work. Such an excellent performance of the DBD employed in this study may be explained by higher plasma frequency, thinner gap spacing and dielectric materials, which have been proved by many released studies to be in favor of higher discharge intensity and/or electron intensity [7,27,28]. Luo et al. [29] found that reducing the gap spacing would promote the stability and intensity of the discharge; Cai et al. [30] found intensity of discharge raised with the increasing of dielectric constant and dropped with the increasing of barrier thickness. The results obtained from our experimental study on the dependence of DBD performance on dielectric materials with different thickness is consistent with theirs. In addition, theoretical calculation was in good agreement with the aforementioned experiments results. As long as this self-made DBD discharge wave was parallelogram with peak applied voltage of  $U_{\text{max}}$  and breakdown voltage of  $U_{\text{min}}$ , electric power P between the coaxial quartz tubes was calculated by formula (1) [27,28].

$$P = 4fC_{\rm d} \frac{C_{\rm d}}{C_{\rm d} + C_{\rm g}} U_{\rm min}(U_{\rm max} - U_{\rm min}) \tag{1}$$

where *f* is DBD frequency;  $C_d$  and  $C_g$  are capacitance of dielectric and gap, respectively, both of which were with direct proportion to  $\varepsilon/l$  ( $\varepsilon$  denoted dielectric constant and *l* denoted their thickness). Comparing Chang's work with ours, you will find that with equivalent dielectric constant of Pyrex and Quartz, formula (1) demonstrates that enhancing frequency with three orders of magnitude would reduce the applied voltage, still with equivalent or even better removal efficiency.

In short, less quantity of electrons resulted from relatively lower electric power in gap spacing in Chang's reactor limited the removal efficiency and ARA of NH<sub>3</sub>, without consideration of thinner dielectric and gap spacing. Additionally, less gas flow rate also contributes to lower ARA.

### 3.5. Reaction mechanism

Some white sediment was observed deposited on reaction tube as  $NH_3$  gas stream passing by the discharge area. Such needle-like bright crystal was also reported to be observed during removal of  $NH_3$  via non-pulse corona-discharge by Wiwut et al. [2]. The white sediment was identified to be  $NH_4NO_3$ and  $NH_4NO_2$  through ion exchange chromatography (IEC, DX-500). The detection result was in good agreement with Ruan et al. [20] and Marek et al. [25]. It is reported [2,31] that at room temperature,  $NH_4NO_2$  and  $NH_4NO_3$  are formed by reaction with  $O_3$  as:

$$2NH_3 + 4O_3 \rightarrow NH_4NO_2 + H_2O_2 + 4O_2 \tag{R1}$$

$$NH_4NO_2 + H_2O_2 \rightarrow NH_4NO_3 + H_2O$$
(R2)

However, as mentioned in Section 3.3, temperature inside DBD reactor was about  $250 \,^{\circ}$ C at peak voltage of 9000 V. Ozone is unstable and is suppressed at this temperature [2,32]. Therefore, oxidation of NH<sub>4</sub>NO<sub>2</sub> and NH<sub>4</sub>NO<sub>3</sub> by O<sub>3</sub> can be neglected. Instead, O, OH, N and H radicals are produced at high temperature and contribute to NH<sub>3</sub> removal [27]. As a result, O, OH, N and H radicals, who are produced by attacking with energetic electrons with presence of air [7,20,24], are considered to play more important role in the formation of NH<sub>4</sub>NO<sub>2</sub> and NH<sub>4</sub>NO<sub>3</sub>. The main reactions were supposed to be as follows [26,32,33]:

NO + OH + N<sub>2</sub> 
$$\xrightarrow{k_3}$$
 HNO<sub>2</sub> + N<sub>2</sub>  
 $k_3[N_2] = 3.61 \times 10^{-18} \text{ m}^3/\text{s}$  (R3)

,

Table 5 Production concentration analysis via IEC (9000 V of applied voltage, 0.067 s of residence time)

	Concentration (mg/l)			
	$[\rm NH_3] = 0  mg/m^3$		$[NH_3] = 125 \text{ mg/m}^3$	
	$\overline{NO_2^-}$	NO <sub>3</sub> <sup>-</sup>	$\overline{NO_2^-}$	$NO_3^-$
Gas-phase product Solid-phase product	0.63 0	2.12 0	0.53 1.26	2.79 2600

$$NO_2 + OH + N_2 \xrightarrow{k_4} HNO_3 + N_2$$
  
$$k_4[N_2] = 6.94 \times 10^{-18} \text{m}^3/\text{s}$$
(R4)

 $NH_3 + HNO_2 \rightarrow NH_4NO_2$  (R5)

$$NH_3 + HNO_3 \rightarrow NH_4NO_3$$
 (R6)

One of the major concerns in the application of non-thermal plasma in NH<sub>3</sub> removal is the production of  $NO_x$ . Quantitive investigation on the source and sinks of N should be made to study the fate of  $NO_x$ . However, only preliminary exploration of the utilization of DBD to remove NH3 from gas stream was possible in this study due to apparatus and conditions constraints. The open system without excluding air made it difficult to measure  $NO_x$  converted from  $NH_3$ , too. Instead, we measured the concentration of NO<sub>3</sub><sup>-</sup> ([NO<sub>3</sub><sup>-</sup>]) and NO<sub>2</sub><sup>-</sup> ([NO<sub>2</sub><sup>-</sup>]) in products in both pure air gas stream and NH<sub>3</sub>-air gas stream after discharge, aiming to analyse sinks of NH<sub>3</sub>-N. One litre gas products was sampled into 20 ml distilled water, while white sediments in outpour tube were collected and dissolved in 100 ml distilled water after discharging for 1 h at applied voltage of 9000 V, inlet [NH<sub>3</sub>] of 0 and 125 mg/m<sup>3</sup>, and gas residence time of 0.067 s. Samples were detected by means of IEC. The results were listed in Table 5.

Under this condition,  $\eta$ NH<sub>3</sub> obtained at inlet [NH<sub>3</sub>] of 125 mg/m<sup>3</sup> was 53%, according to the experimental results shown in Fig. 2. The total volume of gas passing the reaction zone was 8.38 m<sup>3</sup> in 1 h and total amount of NH<sub>3</sub> was thus 1047.6 mg. From Table 5, we can conclude as following: (1) the amount of NO<sub>2</sub><sup>-</sup> and NO<sub>3</sub><sup>-</sup> was few in pure air gas stream after discharge. As a result, NO<sub>2</sub><sup>-</sup> and NO<sub>3</sub><sup>-</sup> converted from air-N could be neglected in NH<sub>3</sub>-included gas stream. (2) NO<sub>3</sub><sup>-</sup>-N (including NH<sub>4</sub><sup>+</sup>-N in solid NH<sub>4</sub>NO<sub>3</sub>) accounted for about 48.8% of NH<sub>3</sub> destructed. (3) NO<sub>2</sub><sup>-</sup>-N (including NH<sub>4</sub><sup>+</sup>-N in solid NH<sub>4</sub>NO<sub>2</sub>) accounted for about 5.9%.

 $NO_x$  inevitably formed during discharge. Fortunately,  $NO_x$  may be further decomposed by extended discharge through (R3), (R4) and the following reactions [7,20,24,32,33]:

$$NO + OH + N_2 \xrightarrow{k_3} HNO_2 + N_2$$
  

$$k_3[N_2] = 3.61 \times 10^{-18} \text{ m}^3/\text{s}$$
(R7)

$$NO_2 + OH + N_2 \xrightarrow{k_4} HNO_3 + N_2$$
  

$$k_4[N_2] = 6.94 \times 10^{-18} \text{ m}^3/\text{s}$$
(R8)

NO+NH<sub>2</sub>
$$\xrightarrow{k_7}$$
N<sub>2</sub> + H<sub>2</sub>O  $k_7 = 9.11 \times 10^{-18} \text{ m}^3/\text{s}$  (R9)

NO + NH 
$$\xrightarrow{k_8}$$
 N<sub>2</sub> + OH  $k_8 = 5.15 \times 10^{-17} \text{ m}^3/\text{s}$  (R10)

NO + N<sub>2</sub>H
$$\xrightarrow{k_9}$$
N<sub>2</sub> + HNO  $k_9 = 8.3 \times 10^{-17} \text{ m}^3/\text{s}$  (R11)

$$NO_2 + OH + N_2 \xrightarrow{k_{10}} HNO_3 + N_2$$
  

$$k_{10}[N_2] = 6.94 \times 10^{-18} \text{ m}^3/\text{s}$$
(R12)

The formed acids in (R7)–(R12) can combine with NH<sub>3</sub> to form ammonium salts NH<sub>4</sub>NO<sub>3</sub> and NH<sub>4</sub>NO<sub>2</sub> which could be removed easily through physical means.

## 4. Conclusion

Ammonia ( $NH_3$ ) is an odorous gas and has wide industrial applications. Our experimental results show that DBD is useful for the removal of  $NH_3$  even when gas residence time is as short as 0.1 s and inlet gas concentration is fairly low. This self-made DBD is also of great advantage when the inlet  $NH_3$  concentration is unstable.

 $NH_3$  could be decomposed completely via DBD plasma. About 50%  $NH_3$  destructed to  $NO_3^-$ , the possibly toxic products such as  $NO_x$  can be decomposed by extended discharge.

The DBD reactor used in this investigation shows fairly superior performance in NH<sub>3</sub> removal in comparison with other non-thermal plasma reactors employed previously. It could probably be explained by much higher frequency of power supply. DBD with high frequency may be suitable for the removal of waste gas contaminants.

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