Research Article

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Off-gas detection and treatment for green air-plasma process

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Abstract: Thermal plasma is a promising technology widely used in materials processing and waste treatment due to its unique properties including high temperature, high energy density, high chemical activity, and high quench rate. Air-plasma is preferentially used because air is of low price as plasma gas. The content of NO\textsubscript{x} in off-gas from air-plasma was determined using a gas analyzer, and a treatment unit was designed for the green air-plasma process. Results show that the concentration of NO\textsubscript{x} in off-gas from air-plasma was 2,489 and 9,112 ppm when the plasma input power was 50 and 150 kW, respectively. O\textsubscript{2} in the off-gas would act as an oxidant to promote NO\textsubscript{x} absorption; thus, alkali absorption method was directly used for the treatment of the present off-gas from air-plasma. The absorption efficiency could be increased to 62.2% when additional O\textsubscript{2} was provided into the off-gas to change its O\textsubscript{2} content from 20% to 50%. The absorption rate was estimated based on the experimental data and a multistage absorption unit design, which could be reduced below 100 ppm and meet the emission standard. This article presents the feasibility of thermal plasma off-gas purification, so as to truly realize the green plasma process.

Keywords: thermal plasma, off-gas, removal of NO\textsubscript{x}, green process

1 Introduction

Plasma is usually considered the fourth state of matter that is different from solid, liquid, and gaseous substances. It is a quasi-neutral gas composed of electrons, ions, neutrals in the ground state, excited species, and photons, in which the total number of positive charges is equal to that of negative charges [1,2]. Plasma can be mainly divided into high-temperature plasma and low-temperature plasma according to the ionization degree and temperature [3,4]. High-temperature plasma exhibits similar temperatures for electrons and ions and a very high ionization degree (≈1), while low-temperature plasma exhibits higher electron temperature \( T_\text{e} \) and lower ion temperature \( T_\text{i} \); in this case, a partially ionized medium [5]. Low-temperature plasma can be further classified as thermal and non-thermal plasma, where plasma with a comparable temperature of heavy ions to that of electrons is known as thermal plasma [6].

Thermal plasma is usually generated by way of making electric current pass-through gases. Many routes, such as direct current (DC), alternating current (AC), radio frequency (RF) and microwaves, have been employed to generate thermal plasma [7]. The plasma torch can be classified according to the generating source, calling DC plasma, AC plasma, and RF plasma. The DC arc plasma is usually used in two modes as non-transferred arc and transferred arc [8]. Thermal plasma has distinct properties compared to ordinary chemical reaction media, including high temperature, high energy density, and high chemical activity, which makes plasma a promising technology widely used in material processing and waste treatment, usually known as “plasma enhancement” [2,5].

Plasma processing of materials can be classified into four categories [9,10]. (i) Plasma spheroidization: powders with irregular shape fed into plasma flame were melted in the high-temperature region to form spherical liquid drops and then fast quenched (with sufficient water-cooling or gas-cooling out of the plasma flame) to form spherical powders under sharp quenching rate [11–13]. (ii) Thermal plasma evaporation and condensation: powders with large
sized were fed into plasma flame and vaporized, and ultra-fine powders were produced after fast quenching by physical vapor deposition [14–16]. (iii) Plasma-enhanced chemical vapor deposition: reaction precursors are fed in the form of gases of vapors, and plasma is introduced into the reaction zone to activate the precursors [17–21]. (iv) Plasma treatment of solid phases: target material is prepared by simply exposing the solid precursor to plasma through plasma–solid interaction [22,23].

Thermal plasma is also successfully applied to treat a variety of wastes. On the one hand, plasma can thermally decompose hazardous organic compounds into simpler fuel gas which in turn was employed for diverse applications [24]. On the other hand, plasma also helps melt solid inorganic components or, with the addition of glass former, vitrify waste to form a stable glassy slag product in which hazardous substances are trapped within the glass network. The inert glassy slag which is obtained as a by-product in the plasma-assisted waste treatment can be upgraded to value-added products such as glass ceramics, road filler material, and building construction material [25,26]. Different waste types such as municipal wastes (paper, biomass, plastic, cloth, etc.) and hazardous wastes (from industrial, agriculture, and hospitals) were processed via thermal plasma technology [27–31].

In a project of thermal plasma processing, one of the main parameters which need to take into account is the system pressure. In principle, atmospheric-pressure plasma devices can provide a crucial advantage over low-pressure plasma because they eliminate complications introduced by the need for vacuum. Therefore, atmospheric pressure plasma has been mostly employed for material processing and waste treatment. Air is the most common gas used, but several inert gases (N₂, He, Ar, etc.) have also been widely used as plasma gas [32–34]. When air is used, it must be subjected to a large differential in electrical potential in order for air to conduct electricity. Otherwise, Ar is usually used at the initial stage of the arc ignition process and switched to air after the plasma operates stably.

Another problem with the use of air as a thermal plasma gas is that N₂ and O₂ would react with each other to form NOₓ in the unordinary chemical reaction media provided by thermal plasma. When a plasma wind tunnel was used for ablation of ultrahigh temperature ceramic materials, there existed NOₓ in the exhaust gas which could be preliminarily judged through the color and smell.

This article will discuss the exhaust gas treatment based on the data accumulation. The content of NOₓ in off-gas from air-plasma was determined using a gas analyzer, removal methods and their important parameters were examined according to the characteristics of plasma off-gas, and a treatment unit was designed so as to truly realize the green plasma process.

2 Experimental

Different thermal plasma devices including DC plasma, RF plasma, and three-phase AC plasma were designed and used in our laboratory. The DC plasma and three-phase AC plasma are usually applied for waste treatment, and the RF plasma is commonly used for functional powder material synthesis. DC plasma is also used to test the ablation behavior of ultra-high-temperature materials. When the air was used as plasma gas during these processes, the off-gas was a light brown pungent gas. We supposed that something had to be conducted for the plasma technique both in experimental conduction and in future application.

Figure 1 illustrates the off-gas treatment unit connected with the thermal plasma reactor. The green lines exhibit the gas flow path, while the blue lines are the circulating absorption liquid pipeline. Air is supplied by the air compressor as the plasma gas, and the off-gas is exhausted by an air pump and sent into the off-gas treatment unit. The gas to be treated enters from the bottom of the absorption tower and runs upward, while the absorption liquid enters from the top of the absorption tower and runs downward driven by a circulating pump. In order to enhance their contact and absorption efficiency, we set up sieve plates and porous suspended ball fillers in the absorption tower. The gas before and after absorption was tested using a gas analyzer. The off-gas after absorption was exhausted to the atmosphere or introduced into a secondary stage absorption.

Figure 2 shows the pictures of off-gas treatment unit connected with RF (Figure 2a) and DC (Figure 2b) plasma reactor, respectively. Plasma is usually ignited using Ar as a working gas and switched to air afterward. The off-gas is pumped out of the plasma reactor so that the system pressure maintains at negative pressure, which is conducive to the stable operation of thermal plasma for a long time. In the present work, the plasma equipment runs without any materials treated, so that all NOₓ comes from the plasma itself. The off-gas was tested online using a flue gas analyzer (Testo-350, Testoterm).
3 Results and discussion

3.1 Formation of NO\textsubscript{x} in air-plasma

NO\textsubscript{x} includes N\textsubscript{2}O, NO, NO\textsubscript{2}, N\textsubscript{2}O\textsubscript{3}, N\textsubscript{2}O\textsubscript{4}, and N\textsubscript{2}O\textsubscript{5}, in which NO and NO\textsubscript{2} are often referred to as air pollutants. The formation of NO\textsubscript{x} can be ruled by the following reactions between N\textsubscript{2} and O\textsubscript{2}:

\begin{align*}
N\textsubscript{2} + O\textsubscript{2} & \rightarrow 2NO \\
N\textsubscript{2} + 2O\textsubscript{2} & \rightarrow 2NO\textsubscript{2} \\
2N\textsubscript{2} + O\textsubscript{2} & \rightarrow 2N\textsubscript{2}O \quad (3) \\
2N\textsubscript{2} + 3O\textsubscript{2} & \rightarrow 2N\textsubscript{2}O\textsubscript{3} \\
N\textsubscript{2} + 2O\textsubscript{2} & \rightarrow N\textsubscript{2}O\textsubscript{4} \quad (5) \\
2N\textsubscript{2} + 5O\textsubscript{2} & \rightarrow 2N\textsubscript{2}O\textsubscript{5} \quad (6)
\end{align*}

Whether a chemical reaction occurs spontaneously is usually judged by the change of the Gibbs free energy \(\Delta G\). Thermodynamic calculation has been made based on the data given in the literature [35]. Figure 3 shows \(\Delta G\) of the above reactions as a function of temperature.

\(\Delta G\) value for reaction in Eq. 1 decreases as the temperature increases and becomes lower than zero at the temperature around 7,300°C, indicating that reaction in Eq. 1 could take place spontaneously when the temperature is above 7,300°C. \(\Delta G\) value for other reactions increases as the temperature increases and keeps positive. Thermodynamic calculation shows that the earlier reactions to form NO\textsubscript{x} cannot occur under conventional conditions.

![Figure 1: Illustration of off-gas treatment unit connected with thermal plasma reactor.](image1)

![Figure 2: Picture of off-gas treatment unit connected with (a) RF and (b) DC plasma reactor.](image2)

![Figure 3: \(\Delta G\) of the above reactions as a function of temperature.](image3)
NO$_x$ in nature mainly originates from lightning, which is a form of plasma. Plasma can provide highly chemically active species, promoting reactions that cannot occur under normal conditions. That is the reason why NO$_x$ can be detected in the off-gas of thermal plasma.

### 3.2 Detection of NO$_x$ from air-plasma

Detection of NO$_x$ was first conducted using the 50 kW air-plasma. Detailed plasma parameters are listed in Table 1.

Figure 4 shows the NO$_x$ concentration in the off-gas from the 50 kW air-plasma. The concentration increased and reached a stable value 12 s after the beginning of the test. The time lag is because one needs to pump the off-gas into the analyzer for analysis.

The mean concentration was calculated based on the last four data as shown in Table 2. The mean concentration of NO and NO$_2$ was 2,101 and 388 ppm, respectively, and the total NO$_x$ content is 2,489 ppm. The ratio of NO/NO$_2$ after stabilization is about 5.4.

In order to examine the influence of the plasma power on the NO$_x$ concentration in off-gas, a 150 kW air-plasma was applied. Detailed plasma parameters are listed in Table 3.

Figure 5 shows the NO$_x$ concentration in the off-gas from the 150 kW air-plasma. The air-plasma with the input power of 150 kW was equipped with a plasma wind tunnel, and the detection point was 30 m away from the plasma flame. There was less than 3 m between the plasma flame and detection point when the laboratory had 50 kW plasma. The long distance provided more time for the conversion from NO to NO$_2$ in the air.
Table 4 shows the NO$_x$ concentration in the off-gas from the air-plasma with the input power of 150 kW. This test was started from the moment when the plasma ignited. It can be seen that the concentration of NO and NO$_2$ raised from zero and reached a stable value of 100 s after the beginning of the test.

The mean concentration was also calculated based on the last four data provided in Table 4. The mean concentration of NO and NO$_2$ was 6,968 and 2,143 ppm, respectively, and the total NO$_x$ concentration was 9,112 ppm. The ratio of NO/NO$_2$ after stabilization was about 3.25. The decrease in NO/NO$_2$ ratio from 5.4 to 3.25 was related to the detection position.

### 3.3 Removal of NO$_x$ from air-plasma

#### 3.3.1 Selection of adsorption method

NO$_x$ removal technology mainly includes selective catalytic reduction, selective non-catalytic reduction, solid adsorption method, liquid absorption method, and microbiological method [36]. Among them, the liquid absorption method uses simple equipment to obtain high removal efficiency and large adsorption capacity. According to the type of absorbent and purification principle, the liquid absorption method can be divided into water absorption method, acid absorption method, alkali absorption method, oxidation absorption method, and absorption reduction method [37,38].

At the beginning of this work, the water absorption method was first selected in consideration that water is cheap and pollution-free. Otherwise, it could avoid the corrosive effect of acid and alkali solutions on the equipment. A water circulating pump was used to extract the plasma tail gas and maintain the negative pressure state of the system. The water in the circulating pump also acted as the absorption liquid for NO$_x$ removal. It turned out that the absorption effect was not obvious, because the laboratory was still filled with a pungent smell after water absorption. We terminated the experiment immediately and used alkali absorption method instead.

The alkali solution absorption method uses the alkali solution (NaOH, KOH, or Na$_2$CO$_3$) to react with NO$_x$ and generate nitrate and nitrite, so as to remove NO$_x$. However, the solubility of NO in water and solution is very low, so the alkali solution absorption method is generally only applicable to off-gas with high NO$_x$ content. The off-gas from thermal air-plasma is typically of high NO content as the testing results in Section 3.2, which does not seem apparently to be suitable for alkali absorption treatment.

Oxidation absorption is a method that oxidizes NO to NO$_2$ with an oxidant, in order to promote NO$_x$ absorption in an alkali solution. Oxidants mainly include gas-phase oxidants such as O$_2$, Cl$_2$, and ClO$_2$, and liquid-phase oxidants such as KMnO$_4$, NaClO$_2$, NaClO, H$_2$O$_2$, HNO$_3$, and Na$_2$CrO$_4$. The reactions can be formulated as follows when O$_2$ is used as oxidant and NaOH is used in absorption solution:

$$2\text{NO} + \text{O}_2 \rightarrow 2\text{NO}_2$$  \hspace{1cm} (7)

$$2\text{HNO}_3 + \text{NaOH} \rightarrow \text{NaNO}_3 + \text{H}_2\text{O}$$  \hspace{1cm} (8)

It is gratifying that the off-gas from thermal air-plasma is still mainly composed of O$_2$ and N$_2$. In order to determine the O$_2$ and N$_2$ content, the off-gas was collected from the plasma reactor using gas bags and characterized by gas chromatography. Table 5 shows the ratio of O$_2$/N$_2$ in pure air and the off-gas from air-plasma with the input power of 150 kW. The values are 0.276 and

| Serial number | Time (s) | NO (ppm) | NO$_2$ (ppm) | NO$_x$ (ppm) |
|---------------|----------|----------|--------------|--------------|
| 01            | 10       | 0.0      | 0.0          | 0.0          |
| 02            | 15       | 0.0      | 0.0          | 0.0          |
| 03            | 20       | 0.0      | 0.2          | 0.2          |
| 04            | 25       | 0.0      | 0.0          | 0.0          |
| 05            | 30       | 414.0    | 86.2         | 500.2        |
| 06            | 35       | 1,484.0  | 257.6        | 1,741.6      |
| 07            | 40       | 2,068.0  | 336.1        | 2,404.1      |
| 08            | 45       | 2,952.0  | 680.0        | 3,632.0      |
| 09            | 50       | 2,995.0  | 482.3        | 3,477.3      |
| 10            | 55       | 3,289.0  | 812.1        | 4,101.1      |
| 11            | 60       | 3,819.0  | 971.6        | 4,790.6      |
| 12            | 65       | 2,711.0  | 844.6        | 3,555.6      |
| 13            | 70       | 3,890.0  | 1,142.1      | 5,032.1      |
| 14            | 75       | 6,008.0  | 1,837.5      | 7,845.5      |
| 15            | 80       | 6,821.0  | 2,145.8      | 8,966.8      |
| 16            | 85       | 7,084.0  | 2,083.6      | 9,167.6      |
| 17            | 90       | 6,987.0  | 2,139.3      | 9,126.3      |
| 18            | 95       | 6,938.0  | 2,164.1      | 9,102.1      |
| 19            | 100      | 6,866.0  | 2,186.1      | 9,052.1      |

### Table 5: Ratio of O$_2$/N$_2$ in pure air to the off-gas

| Sample     | O$_2$ (vol%) | N$_2$ (vol%) | Ratio of O$_2$/N$_2$ |
|------------|--------------|--------------|----------------------|
| Pure air   | 21.6         | 78.4         | 0.276                |
| Off-gas    | 20.2         | 79.8         | 0.253                |
0.253, respectively. The atomic ratio of O/N in NOx (whether 1/1 in NO or 2/1 in NO2) is greater than that in the air (0.276). The reaction between O2 and N2 to form NOx would consume more O2 than N2 and result in a decrease of O2 and an increase of N2 content. That is the reason why the off-gas exhibited a lower O2 content and higher N2 content than air.

Now that there is O2 in the off-gas that can act as an oxidant to promote NO absorption, the alkali absorption method can be directly used for treatment of the present off-gas from air-plasma. In consideration that the alkali solution would corrode the water circulating pump, a spray tower (shown in Figure 1) was built.

### 3.3.2 Effect of gas flow

The concentration of NaOH solution as absorption liquid is 16 wt%. The flow of the liquid pump for NaOH solution circulation was 30 L·min⁻¹. The torch power was kept constant, and the off-gas was pumped and introduced into the absorption tower through a branch connected to the exhaust pipeline. The gas flow was regulated by a rotameter from 2.5 to 20.0 m³·h⁻¹. Figure 6 shows the effect of gas flow on the absorption efficiency.

The absorptivity of NOx increased obviously from 6.5% to 61.2% when the gas flow decreased from 20 to 2.5 m³·h⁻¹. The absorptivity of NO also increased from 5.3% to 52.5%. However, the concentration of NOx increased instead of decreased and showed an opposite changing trend. The absorptivity of NO2 was -4.7% with the gas flow of 20 m³·h⁻¹ and decreased to -16.0% with the gas flow of 2.5 m³·h⁻¹.

Both the decrease of NO and increase of NO2 during the absorption process were related to the conversion rate of NO to NO2. When the gas flow decreased from 20 to 2.5 m³·h⁻¹, the residence time was prolonged by eight times, which provided more sufficient time for the conversion from NO to NO2. In order to give a clear explanation, we suppose that the conversion from NO to NO2 is divided into two stages, before and after absorption. The residence time was prolonged for both stages. At the first stage before absorption, a longer residence time was attributed to the higher conversion from NO to NO2, resulting in an absorption efficiency, which is the reason why the absorptivity of NO increased obviously with the gas flow decrease. At the second stage after absorption, a longer residence time was still attributed to the higher conversion from NO to NO2, resulting in an increase in NO2 concentration.

### 3.3.3 Effect of O2 content

It is demonstrated from the above results that O2 in the off-gas must have played an important role so that alkali absorption method can be directly used for the treatment of the off-gas from air-plasma with high NO content. Inspired by this O2-enhanced absorption process, we provided additional O2 into the off-gas to increase its O2 content. Figure 7 shows the absorption efficiency with and without additional O2. The O2 flow was 1.5 m³·h⁻¹ when the off-gas was 2.5 m³·h⁻¹. The concentration of O2 was increased from about 20% to about 50%. The absorptivity of NO increased slightly from 61.2% to 69.7%, while the absorptivity of NO2 increased obviously from -16% to 16%. The obvious increase in NO2 absorptivity cannot be explained clearly through the change of residence time.

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**Figure 6:** Effect of gas flow on the absorption efficiency.

**Figure 7:** The absorption efficiency with and without additional O2.
It indicated that O₂ might have played a synergistic absorption role except that it can convert NO to NO₂ as an oxidant during different stages. The total NOₓ absorptivity increased from 52.5% to 62.2%.

### 3.3.4 Design of multistage unit

Taking the gas flow and O₂ content effects into account, the content of NO and NO₂ was reduced to a fairly low level at 436 and 197.2 ppm, respectively. The total content of NOₓ was 633.2 ppm, which has not reached the emission standard of 200 ppm. Multistage absorption is a commonly used strategy in industrial production to increase absorption efficiency.

The number of cycles of absorption can be estimated from the absorption rate. In the above absorption experiment, the initial concentration of NOₓ was 1,674.9 ppm, and the concentration after one cycle absorption was 633.2 ppm. The absorption rate was 62.2%. The NOₓ concentration was detected at different positions as shown in Figure 8: the inlet of the first absorption tower, the outlet of the first absorption tower, the outlet of the second absorption tower, and the outlet of the third absorption tower. The NOₓ concentration and the absorption rate after different cycles of absorption were estimated, and the results are shown in Table 6.

It can be estimated that NOₓ concentration after a three-cycle absorption can be reduced below 100 ppm which can meet the emission standard. Based on the assessment, we have designed a multistage unit for the treatment of off-gas from air plasma, as shown in Figure 8.

| Cycle number | Cumulative absorption rate (%) | NOₓ concentration (ppm) |
|--------------|--------------------------------|-------------------------|
| 0            | 0                              | 1,674.9                 |
| 1            | 62.2                           | 633.2                   |
| 2            | 85.7                           | 239.2                   |
| 3            | 94.6                           | 90.5                    |

This article intends to present the feasibility of thermal plasma off-gas purification. Further testing and application according to detailed plasma running parameters are still in progress.

### 4 Conclusions

The concentration of NOₓ in off-gas from air-plasma was determined by a gas analyzer, removal methods were selected according to the characteristics of plasma off-gas, different influencing parameters were examined, and a treatment unit was designed for the green air-plasma process. Several conclusions can be drawn from the results and discussion:
1. N₂ and O₂ would react and form NOₓ in the unordinary chemical reaction media provided by thermal plasma, and the concentration of NOₓ in off-gas from air plasma increased with the increase in plasma power.
2. The content of O₂ in the off-gas was close to that in air, which would act as an oxidant to promote NOₓ.

![Figure 8: Illustration of a multistage off-gas treatment unit.](image-url)
absorption; thus, alkali absorption method can be directly used for the treatment of the present off-gas from air-plasma.

3. The absorption efficiency was 52.5% when the gas flow was 2.5 m³/h⁻¹, and it could be further increased to 62.2% when additional O₂ was provided into the off-gas to increase its O₂ content from 20% to 50%.

4. It was estimated that the NOₓ concentration can be reduced below 100 ppm when a multistage absorption unit was designed, which can meet the emission standard and present a feasible technical scheme for the green plasma process.

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