Removal of NO\textsubscript{2} and O\textsubscript{3} generated from corona discharge in indoor air cleaning with MnO\textsubscript{2} catalyst

H Ge \textsuperscript{1}, R Yu \textsuperscript{1}, D Mi \textsuperscript{2}, Y M Zhu \textsuperscript{1*}

1. Institute of Environment of Remediation, Dalian Maritime University., Lingnan Road Dalian, China
2. Department of Physics, Dalian Maritime University., Lingnan Road Dalian, China

E-mail: ntp@dlmu.edu.cn

Abstract. The production rules and removal efficiency of harmful byproducts such as NO\textsubscript{2} and O\textsubscript{3} generated from DC corona discharge in indoor air cleaning were investigated. The production behaviours of NO\textsubscript{2} and O\textsubscript{3} and the relationship between the amount of catalyst (MnO\textsubscript{2}) and the removal rate of harmful byproducts were experimentally studied. Further, indoor application tests were carried out in a closed room with 90 m\textsuperscript{3}. The results showed that the concentrations of NO\textsubscript{2} and O\textsubscript{3} produced by corona discharge linearly increased with discharge time. The NO\textsubscript{2} yield is larger than O\textsubscript{3} by almost one order of magnitude under the same discharge power. To satisfy the demand of Standard of Indoor Air Quality (GB/T18883-2002), the power consumption of unit volume should be less than 1 W m\textsuperscript{-3} and the catalyst MnO\textsubscript{2} consumptions in positive-negative corona discharge were 200 cm\textsuperscript{3} W\textsuperscript{-1} and 100 cm\textsuperscript{3} W\textsuperscript{-1}, respectively.

1. Introduction
Corona discharge in air could produce many active species, and then could be widely applied in environmental pollution control [1-5]. Corona discharge has multiple purification effects in indoor air cleaning process, including particulate matter trapping, sterilization as well as VOCs removal. It also has many advantages comparing with other plasma technologies, such as low energy consumption, discharge stability and easy to use etc. However, the harmful by-products O\textsubscript{3} and NO\textsubscript{2} are also produced simultaneously by corona discharge during purification process. If these by-products can not be effectively eliminated, the practical application of corona discharge in air pollution control will be subject to considerable restrictions.

Some studies show that MnO\textsubscript{2} could accelerate the conversion of O\textsubscript{3} to O\textsubscript{2}, and then can be used as...
plasma air purification aftertreatment to improve discharge quality [6]. Mix oxides of MnO₂ and Fe₂O₃ attached to active carbon (AC) can effectively decompose O₃ [7]. As far as O₃ catalytic decomposition ability is concerned, as active component, MnO₂ is better than CuO and Fe₂O₃. Meanwhile, AC is a kind of superior carrier over γ-Al₂O₃ and molecular sieve [8]. In addition, MnO₂ can also be used to effectively remove NO₂ produced by discharge [9].

In this study, the production behavior of O₃ and NO₂ generated by corona discharge in a closed chamber was investigated. On this base, a manganese catalyst was used to decompose the harmful by-products O₃ and NO₂. Finally, the applied research was carried out in a closed 84 m³ room to study the decomposition law and removal method of by-products.

2. Experiment Method

Figure 1 shows the experimental setup of multi-needle-to-plate corona discharge in a closed chamber of 0.4 m³. The needle-to-plate distance is 25 mm, and the needle-to-needle distance is 20 mm. There are 97 needles in the matrix with needle radius of 1 mm and length of 5 mm. Discharge power is supplied by ZGF-DC high-voltage generator and the applied voltage is measured by a Tek P6015A divider connected with an HP 54503 digital oscilloscope. The flow rate in electrical field is kept at 2.8 m³/s, which is measured by ZRQF anemometer. The manganese oxides catalyst is of 10 mm thickness per layer with hexagon holes of 2 mm side length. The accumulated concentrations of NO₂ and O₃ in the closed chamber are measured by PGM-7840 Five-Gas Monitors (RAE Co.) with resolution of 0.1 ppm and GASTEC Detector tube(0.05~0.6 ppm, 0.01 ppm), respectively.

As shown in figure 2, the indoor air is cycled and purified in the following way by the air cleaner: firstly, air is taken into the air cleaner by blower fan, then it is processed in the discharge area and catalyst layers, and finally, it returns to the room with dimension of 5×6×3 m³. The discharge reactor possesses 10 channels with discharge distance of 25 mm, and each channel has 194 needles in the matrix. The air circulation can be calculated and it is about 400 m³ h⁻¹.
3. Results and Discussion

3.1. Production of NO₂ and O₃ generated by corona discharge

Figure 3 shows the effect of discharge time on the productions of NO₂ and O₃ under different electrode polarity. The temperature and relative humidity in closed chamber are approximately kept at 20 °C and 83 %. The applied voltage in positive corona discharge is 15.6 kV and the corresponding current and power are 0.04 mA and 0.624 W, respectively. Two sets of experiments were made in the case of negative corona discharge, and the measured voltage, current and power are 17 kV, 0.045 mA, 0.765 W and 17.4 kV, 0.05 mA, 0.87 W, respectively.

As shown in figure 3, the concentrations of both NO₂ and O₃ produced by positive-negative corona discharge increase linearly with discharge time at initial stage, then slightly increase after 1 h, and finally achieve a dynamic balance at 1.5 h. The productions of NO₂ and O₃ generated by negative corona discharge are a little more than that by positive corona discharge. This means that the concentrations of NO₂ and O₃ at dynamic balance are related to discharge power but not electrode polarity. The production of NO₂ is one order of magnitude higher than O₃.

![Figure 3. Concentrations of NO₂ and O₃ produced as a function of time in corona discharge.](image)

The standard values of NO₂ and O₃ for 1 h in indoor air quality standard (GB-T18883-2002) are 0.24 mg m⁻³ (0.12 ppm) and 0.16 mg m⁻³ (0.07 ppm), respectively. In indoor air purification research, it is thought that generating amount of gas pollutant should be one order of magnitude less than those in GB-T18883-2002, because only the results obtained from experiment are more meaningful and representative for practical application. Thus, under present experimental condition, the discharge power in positive-negative corona discharge should be a little higher than 0.5 W when the closed chamber is 0.4 m³ and the estimated power consumption of unit volume should be less than 1 W m⁻³.

3.2. Catalytic removal of NO₂ and O₃
Figure 4 shows the effect of two layer manganese catalyst on decomposition of NO₂ and O₃ when discharge power is lower than 1 W in the closed chamber with 0.4 m³. As shown in figure 4, the concentration of O₃ generated by positive-negative corona discharge reach dynamic balance and kept at 0.05 ppm after discharge 10 min, satisfying the national standard of indoor air quality (O₃, 0.07 ppm, 1 h). The concentration of NO₂ simultaneously produced with O₃ also reach dynamic balance after discharge for a period of time, and kept at 0.2 ppm for positive discharge as well as 0.3 ppm for negative discharge, slightly exceeding the national standard of indoor air quality (NO₂, 0.12 ppm, 1 h).

![Figure 4](image)

**Figure 4.** Concentrations of NO₂ and O₃ produced in corona discharge with 2-layer catalyst.

- ■ NO₂ in negative corona (0.765 W);
- ▲ O₃ in negative corona (0.765 W);
- □ NO₂ in positive corona (0.624 W);
- △ O₃ in positive corona (0.624 W).

3.3. **Practical Application of decomposing NO₂ and O₃**

In practical application of catalytic decomposition of NO₂ and O₃, using two layer catalyst, the production behavior of by-products during discharge under different electrode polarity was studied.

As shown in figure 5(a), the concentrations of NO₂ and O₃ produced by positive corona discharge under different discharge power reach a dynamic balance after 1 h. Under the conditions of both 4.8 W and 8.58 W, the concentrations of O₃ all satisfy GB-T18883-2002. However, the concentrations of NO₂ slightly exceed the standard for 4.8 W. Especially, the concentrations of NO₂ are two or three times higher than the standard value for 8.58 W. Therefore, the discharge power of positive corona discharge should be less than 4 W under this experiment condition.

The variation trend of concentrations of NO₂ and O₃ produced by negative corona discharge is shown in figure 5(b). The concentrations of both by-products satisfy the standard at 8.25 W when their values reach a dynamic balance. The concentration of O₃ is less than the standard value, while the concentration of NO₂ slightly exceeds the standard at 12 W. Thus, the discharge power of negative corona discharge should be less than 12 W under this experiment condition.

In order to satisfy the standard GB-T18883-2002, the estimated values of the dosage of manganese for decomposing NO₂ and O₃ produced by positive or negative corona discharge are as follows: 200 cm³ W⁻¹ for positive corona discharge and 100 cm³ W⁻¹ for negative corona discharge.
Figure 5. NO\textsubscript{2} and O\textsubscript{3} reduced by catalyst in indoor air cleaning.
1. +4.8 W NO\textsubscript{2}(◇); 2. +4.8 W O\textsubscript{3}(□); 3. +8.58 W NO\textsubscript{2} (△); 4. +8.58 W O\textsubscript{3}(○)
5. -8.58 W NO\textsubscript{2} (◆); 6. -8.58 W O\textsubscript{3}(■); 7. -12.0 W NO\textsubscript{2}(▲); 8. -12.0 W O\textsubscript{3}(●)

4. Conclusion
(1) The concentration of O\textsubscript{3} produced by needle matrix to plate corona discharge linearly increases in 1.5 h and its production is directly related to discharge power. The production of NO\textsubscript{2} is one order of magnitude higher than O\textsubscript{3} under the same discharge power.
(2) The power consumption of unit volume in our experimental condition is less than 1 W m\textsuperscript{-3}.
(3) The O\textsubscript{3} and NO\textsubscript{2} produced by corona discharge can be rapidly and easily decomposed by MnO\textsubscript{2} catalyst. The dosages of catalyst in positive and negative corona discharge in our experimental condition are approximately 200 cm\textsuperscript{3} W\textsuperscript{-1} and 100 cm\textsuperscript{3} W\textsuperscript{-1}, respectively.

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