Performance of the Chlorobenzene Destruction with Nonthermal Plasma: Effects of the Gas Conditions and the Gas Components

Wen-Jun Liang[1]; Guo-Bin Yin[2]; Xiu-Juan Shi[3]; Jia Liu[4]; and Feng Bin[5]

Abstract: Nonthermal plasma (NTP) is a promising technology for the decomposition of chlorobenzene (CB), but many conditions will affect the performance of NTP. The effects of gas temperature, relative humidity, oxygen content, sulfur dioxide (SO_2), and nitric oxide (NO) on the removal of CB in dielectric barrier discharge (DBD) were studied in this study. The results showed that the gas temperature with the highest CB removal rate was 50°C at 14 kV and 200°C at 18 kV. With the addition of water vapor, the removal efficiency of CB first increased and then decreased, reaching the highest (52.94%) when the relative humidity was 20% at 14 kV. The presence of oxygen inhibited the degradation of CB. At 14 kV, when the oxygen concentration increased from 0% to 25%, the degradation efficiency of CB decreased from 71.93% to 32.54%. In the discharge process, the addition of SO_2 promoted the removal of CB, while NO inhibited under the same conditions. The addition of SO_2 and NO did not change the specific energy density. For ozone (O_3), the addition of water molecules, SO_2, and NO inhibited the formation of O_3. It is of great significance to study the influence of relevant gas conditions on the degradation of CB by NTP technology, so as to provide a reference for industrial application. **DOI:** 10.1061/JOEEDU.EEENG-7179. *© 2023 American Society of Civil Engineers.*

Author keywords: Chlorobenzene (CB); Dielectric barrier discharge (DBD); Gas temperature; Relative humidity; Oxygen (O_2); Sulfur dioxide (SO_2); Nitric oxide (NO); Ozone (O_3).

Introduction

Chlorinated volatile organic compounds (CVOCs) are the main air pollutants with high toxicity, strong stability, and poor reactivity that can cause harmful effects on humans and the environment (Li et al. 2020a; Liu et al. 2019; Lu et al. 2021). Chlorobenzene (CB) is a common CVOC that is widely used in pharmaceutical, dye, and chemical industries. It can make people dizzy and neurasthenic, and aggravate the risk of cancer after long-term exposure (Li et al. 2021; Zhu et al. 2015b). CB is also one of the important precursors of dioxin (Hsu et al. 2021).

According to recent researches, the common technologies for removing CVOCs include absorption/adsorption technology, thermal catalysis technology, biological treatment technology, and catalytic oxidation technology (Li et al. 2020b). But these technologies have some limitations. There is a risk of pollutant emission in absorption and adsorption (Dahiru et al. 2021). Thermal catalysis often requires higher temperature (Kamal et al. 2016), and the biological treatment cycle is long (Barbusinski et al. 2017). The catalyst used in catalytic oxidation will be inactivated over time, and regular maintenance and treatment will increase the cost (Dahiru et al. 2021).

Nonthermal plasma (NTP) is a new technology for the decomposition of CVOCs (Yu et al. 2022). It is often used to remove VOCs of low concentration, such as chemical industry and waste incineration industry. NTP can produce highly active substances to promote the degradation of CVOCs, such as high-energy electrons, photons, ozone (O_3), hydroxide (OH), and other active free radicals (Lu et al. 2019), and it can efficiently degrade volatile organic compounds at room temperature and atmospheric pressure (Li et al. 2020c; Nguyen et al. 2018). At the same time, NTP also shows the advantages of simple operation, low investment, and operation cost (Trinh and Mok 2016; Zhu et al. 2020). However, how to reduce its energy efficiency and the formation of by-products needs to be further investigated.

Actual industrial flue gas emissions are often of a certain temperature, moisture, and oxygen level, and these gas conditions are important influencing factors in the removal of VOCs by NTP (Li et al. 2020; Lu et al. 2019). Previous studies have shown that the degradation of methane by dielectric barrier discharge (DBD) had a higher total gas number density at lower gas temperature, and free radicals could be effectively produced through electron collision (Takana et al. 2012). When NTP technology is used to destruct VOCs, the existence of water vapor cannot be ignored, because water vapor can be decomposed into the free radical OH, and its oxidation ability is stronger than other active substances (such as metastable nitrogen molecules and atoms) (Li et al. 2020b). Nguyen et al. (2018) found that under the condition of DBD, the removal efficiency of toluene was the highest when the relative humidity was 20%. Zhou et al. (2021) found that the removal

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efficiency of CB in dry air was the highest, and the decomposition of pollutants decreased with the increasing of humidity level. Because oxygen plays an important role in the degradation of VOCs by NTP, the oxygen content in discharge gas has a significant effect on the emission reduction effect of VOCs. Many studies had reported that NTP could obtain the highest VOCs removal efficiency at lower oxygen levels. For example, the oxygen content of toluene was 5% (Guo et al. 2006), the oxygen content of dichloromethane was 4% (Abd Allah et al. 2014), and the oxygen content of benzene was 3%–5% (Kim et al. 2008).

Under normal conditions, there were few experimental studies on the effect of flue gas temperature. Besides, the investigations of the effect of relative humidity and oxygen content on the degradation of VOCs by NTP were still not systematic and comprehensive enough, which needs further investigation. Furthermore, some flue gas components such as SO_2 and NO were also included in the general flue gas emissions (Cui et al. 2021; Li et al. 2022), and it is essential to investigate the effluence of the interaction between these gases and VOCs. It will contribute to exploring the possibility of a synergistic effect between desulphurization, denitrification, and VOCs degradation in practical flue gas treatment.

Taking CB as the target pollutant, this paper systematically studied the effects of gas temperature, relative humidity, and oxygen content on the degradation of CB by DBD, including CB removal efficiency, energy efficiency, specific energy density, and reactor outlet temperature. In addition, SO_2 and NO were important components in the flue gas discharged by actual industry, so in our experiment, we explored the effects of SO_2 and NO content on CB removal.

Experimental

As shown in Fig. 1, the NTP reactor used in this experiment was a coaxial tubular DBD structure. The reactor was made of quartz glass, with a wall thickness of 1.5 mm, an outer diameter of 32 mm, and an inner diameter of 29 mm. There was a branch pipe (inner diameter of 5 mm, outer diameter of 8 mm, and branch pipe length of 40 mm) at both ends 35 mm away from the port as the gas inlet and outlet. Tungsten wire with a diameter of 0.8 mm was selected as the discharge electrode, the reactor was sealed with insulating rubber plug, and the discharge electrode was fixed in the center of the tube. The ground electrode was made of aluminum foil and tightly surrounded the outer wall of the reaction tube, with a length of 200 mm. The power supply of the DBD reactor was controlled by a variable AC transformer, the input voltage and frequency were controlled by console, and the experimental frequency was controlled at 120 Hz. The reactor was connected in series with a switch, and the current value in the circuit was measured with a multimeter with appropriate range.

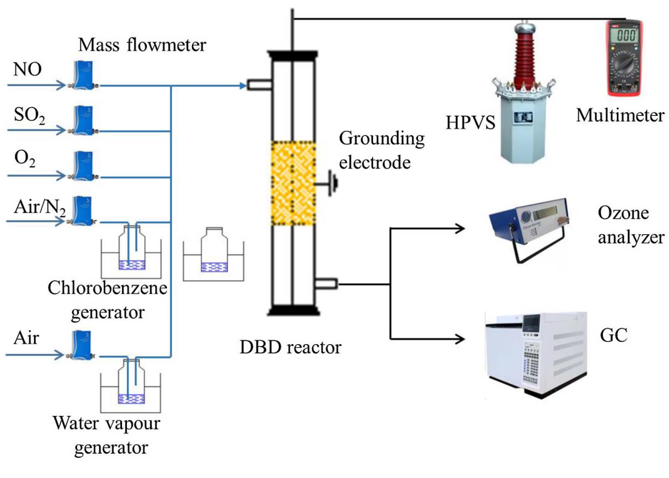


Fig. 1. Schematic diagram of the experimental setup. (Images by Guo-Bin Yin.)

The carrier gas was pumped into the reactor with an air compressor and controlled by a mass flow meter. CB was evaporated into the mixing bottle. The experiments were carried out at room temperature and pressure. The initial concentration (500 mg/m^3) and gas volume (3 L/min) of CB required for the experiment were obtained. The gas humidity was controlled by a water bath device. The whole experiment process was carried out in an insulated environment to avoid the VOCs condensation. At the same time, the purging amount and water bath temperature were controlled for humidity control, and the relative humidity was monitored by a hygrometer. In order to investigate the effect of the oxygen concentration on the VOCs removal, N_2 (78%) and O_2 (21%) cylinder gases were used, and the oxygen concentration was controlled by a mass flowmeter. The gases (SO_2 and NO) required for different experiments were controlled by a mass flowmeter to obtain the required mixed gas concentration, which was fully mixed in the mixing bottle and then entered the DBD reaction system. The concentration of CB at the inlet and outlet was determined by gas chromatography (GC) (HP6890N). The main parameters of GC were as follows: hydrogen flame ionization detector (FID), HP-5 capillary column (column length 30 m, inner diameter 0.32 mm, film thickness inside the column 0.25 um). Detection conditions of GC were as follows: oven temperature 100°C, detector temperature 200°C, sample inlet temperature 140°C. The ozone concentration of the reaction system was examined by an ozone analyzer (106-M). The SO_2 concentration at the outlet was examined by a pararosaniline spectrophotometry.

The CB degradation performance of DBD was evaluated by removal efficiency (RE), specific energy density (SED), energy efficiency (EE) and reactor outlet temperature, as shown in Eqs. (1)–(3)

$$RE(\%) = \frac{C_{in} - C_{out}}{C_{in}} \times 100\% \tag{1}$$

$$SED = \frac{U_{\max} I}{\sqrt{2}Q} \times 60 \tag{2}$$

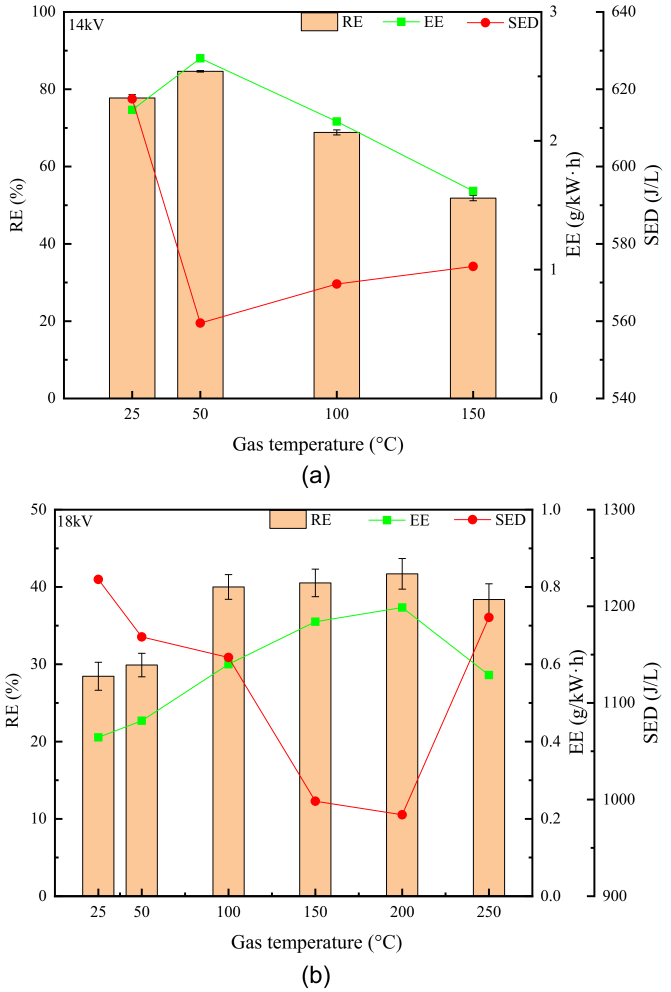
$$EE(\mathrm{g/kWh}) = \frac{C_{in} - C_{out}}{SED} \times 3.6 \tag{3}$$

where C_{in} and C_{out} are the concentration of CB (mg/m^3) of the inlet and outlet of the reactor, U is the system input voltage (kV), I is the leakage current (mA), and Q is the gas flow (3 L/min).

Results and Discussion

Effect of Gas Temperature on the Removal of Chlorobenzene

Gas temperature is one of the important factors affecting the generation of NTP (Harling et al. 2007), but there are few studies on its effect on the degradation of pollutants by NTP. We carried out experiments at 14 and 18 kV, respectively. VOCs are generally removed after dust removal, and the temperature of the flue gas after dust removal usually drops to 200°C or below. Therefore, in our experiment, the maximum temperature was set at 250°C and the



(a)

(b)

Fig. 2. Effect of gas temperature on CB removal: (a) 14 kV; and (b) 18 kV.

gradient was set at 50°C to investigate the effect of gas temperature on the removal of CB.

The results are shown in Figs. 2(a and b). When the voltage was 14 kV, the removal efficiency of CB was 77.73%, 84.64%, and 51.85% at room temperature 50 and 150°C, respectively. The same situation was also obtained at 18 kV, as shown in Fig. 2(b). When the gas temperature rose from 25 to 200°C, the removal efficiency gradually increased from 28.45% to 41.7%, but that decreased to 38.38% at 250°C. The optimum temperature for CB removal was different under different discharge intensities: it was 50 and 200°C when the discharge was 14 and 18 kV, respectively. The results showed that a certain gas temperature was conducive to the degradation of CB by DBD. It had been confirmed that the moderate increase of temperature would enhance the destruction of hydrocarbons (Demidyuk et al. 2008). However, high gas temperature would inhibit the degradation of CB.

Gas temperature has two effects on the plasma discharge system: first, it changes the density of the reaction gas, and second, many reaction rate constants depend on the temperature (Soria et al. 2004). It can be seen from Figs. 2(a and b) that with the increase of gas temperature, the change of SED showed a trend of first decreased and then increased, which was just opposite to the rule of removal efficiency. When the applied voltage was 18 kV, the SED at room temperature was 1,228 J/L, while it was 984 J/L at 200°C and 1,188 J/L at 250°C. SED is mainly determined by the electron density in the reaction system. According to the results of this experiment, when the gas temperature increased, the electron density of the reaction system was affected. As the total gas density decreased, the number of electrons decreased, and then the leakage current (I) decreased, which resulted in the decrease of SED. With the increase of gas temperature, the input energy of the system increased, and more electrons with greater energy generated in the reactor, so SED rose to a certain extent. At different discharge voltages, DBD could destroy more CB molecules at lower SED, so higher EE could be obtained. A certain gas temperature could increase both RE and EE. The lower the total gas density is, the less electron impact ionization and the less free radicals there are (Takana et al. 2012), which is also considered to be one of the reasons why the increase of temperature will reduce the pollutant removal efficiency.

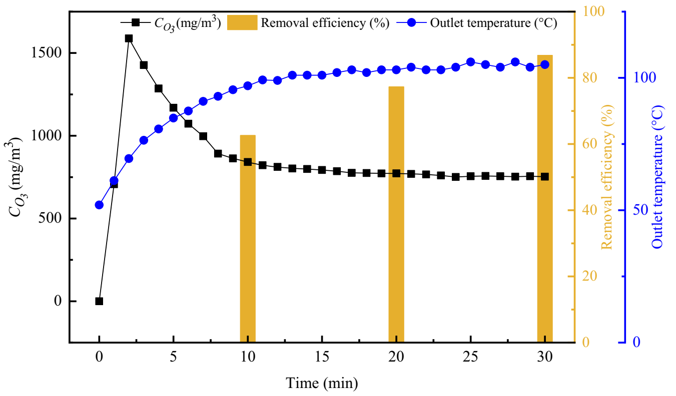


Fig. 3. Ozone concentration and outlet temperature within 30 min of 14 kV.

Temperature will affect the reaction rate between active radical and pollutant molecules in the discharge system. O_3 is one of the important active free radicals in plasma discharge system, which participates in the degradation process of pollutants (Huang and Fang 2020). O_3 can be affected easily by the temperature; when the temperature rises, the decomposition rate constant of O_3 increases and will be destroyed by the high temperature of the plasma reactor (Li et al. 2018). In our experiment, we tested the outlet temperature and O_3 concentration of the reactor with the gas temperature being 50°C under the voltage of 14 kV for 30 min, and the results are shown in Fig. 3. O_3 concentration accumulated rapidly at the beginning of discharge, which reached 1,588 $\mathrm{mg/m^3}$. The temperature of the reaction system continued to increase with the increase of the discharge, and the decomposition of ozone accelerated. The concentration was 757 $\mathrm{mg/m^3}$ at 30 min of discharge. Here, we also compared the change of O_3 concentration with the removal efficiency, and found it was gradually improved in this process. This result showed that O_3 did not promote the degradation of CB by DBD, which might be due to the low reaction rate between O_3 and VOCs in the plasma system (Jiang et al. 2013).

Effect of Relative Humidity on the Removal of Chlorobenzene

In order to investigate the influence of relative humidity on the degradation of CB in a DBD reactor, a relative humidity of 0%–50% was tested continuously with the voltage being 14, 16, 18, 20 kV at room temperature. The humidity of the experiment was controlled by the purging amount and the temperature of the water evaporation tank, which was fluctuated within a certain range (±2%). Therefore, a gradient value of 10% relative humidity was set to ensure that there was a definite difference in humidity of each group.

The results are shown in Figs. 4(a–e). Increasing the relative humidity of the gas was beneficial to improve the removal efficiency of CB. When the relative humidity was increased from 0% to 20%, the removal efficiency of CB was improved under all the voltages, among which the maximum improvement was from 41% to 53% under 14 kV. It indicated that the addition of water molecules was conducive to the degradation of CB. Zhu et al. (2020) studied the effect of humidity on trichlorobenzene and reached similar conclusions. The reason was that the addition of water could increase the amount of OH in the reaction system (Abdelaziz et al. 2018). The main process of its formation was as follows (Huang et al. 2011):

$$e + H_2O \rightarrow e + OH + H \tag{4}$$

$$O + H_2O \rightarrow OH \tag{5}$$

The free radical OH has strong oxidation ability and is considered to be one of the most potential reactants for the decomposition of gas pollutants in DBD reactor. However, as the relative humidity of the gas increased to 50%, it was found that the removal efficiency of CB in each group decreased by 6%–10%. Zhou et al. (2021) studied the degradation of CB in double DBD at different humidity, and found the remove efficiency decreased with the increasing of the humidity. If too much water vapor was introduced, the high-energy electrons generated by the discharge would be quenched by electronegative water molecules, which led to the decrease of the electron density and average energy.

Fig. 4(b) shows that the SED had little relationship with the change of the relative humidity (Nguyen et al. 2018). At 18 kV, SED fluctuated around 920 J/L with the change of humidity. Combined with the experimental phenomenon, the increase of humidity did not change the mode and intensity of DBD. In contrast, the published study showed that the addition of water vapor reduced the number of microdischarges, thus reducing the volume of reactive plasma in the system (Falkenstein and Coogan 1997). This difference might be due to the difference in reactor materials and gas components. The relationship between EE and relative humidity is shown in Fig. 4(c). It can be seen that under various humidity conditions, the EE was the highest when the discharge voltage was 18 kV and was the lowest when the discharge voltage was 20 kV. When the discharge voltage was 20 kV, more energy was needed in order to obtain the higher removal efficiency. Because the SED under each relative humidity condition was almost the same, the change of EE with relative humidity was consistent with the change of removal efficiency, which first increased and then decreased, and reached the maximum at 10%. When the voltage was 18 kV and the relative humidity was 10%, the maximum of EE was 1.83 g/kW/h. Fig. 4(d) shows the temperature change at the outlet of the reaction tube. When the relative humidity was 20%–30%, the outlet temperature of the reaction tube under each voltage was the highest, which was about 20°C higher than that under dry air conditions. Under certain humidity conditions, water molecules could not only improve the degradation efficiency of CB, but also release more heat energy. As shown in Fig. 4(e), the O_3 concentration at three relative humidity conditions with the increase of pressure was measured in our experiment. The result showed that O_3 concentration decreased with the higher relative humidity, which indicated that the addition of water molecules consumed O_3 in the system

$$OH + O_3 \rightarrow O_2 + H_2O \tag{6}$$

This leads to a reduction in the amount of O_3 and other active substances (Lu et al. 2019; Zhang et al. 2016; Zhu et al. 2015a). With the increase of voltage, the O_3 concentration decreased significantly, because the increase of voltage increased the temperature of the reaction system and accelerated the O_3 dissipation.

Effect of O_2 on the Removal of Chlorobenzene

O_2 plays a very important role in the oxidation reaction. In oxygen-containing gas mixtures, oxygen free radicals as active substances are mainly produced in NTP discharge (Abd Allah et al. 2014; Guo et al. 2006). In this experiment, the effect of oxygen content in the range of 0%–25% on the degradation of CB by DBD was investigated by continuously increasing voltage (14, 16, 18, 20 kV). Because most practical applications of removing VOCs with plasma were in environments with not much oxygen, we focused on the oxygen situation in the range of 0–5%, with an interval of 1%.

The effect of oxygen content on the removal efficiency of CB is shown in Fig. 5(a). It was found that under various voltage conditions, the higher the oxygen content was, the lower the removal efficiency was. The removal efficiency under 14, 16, 18, and 20 kV discharge voltage decreased from 100%, 95.02%, 85.78%, and 71.93% (oxygen content 0%) to 79.31%, 63.98%, 53.44%, and 32.54% (oxygen content 25%), respectively. The presence of oxygen inhibited the removal of CB, which was consistent with the experiment results of Zhu et al. (2020) and Zheng et al. (2014). In the absence of O_2, DBD will produce $N_2(A)$ metastable radical and N radical. These two important active substances are shown as follows (Abd Allah et al. 2014; Guo et al. 2006):

$$e + N_2 \rightarrow e + N_2(A) \tag{7}$$

$$e + N_2 \rightarrow N^* + N + e \tag{8}$$

where N^* is the excited nitrogen species by the electrons.

The radicals in the reactions play an important role in the removal of aromatic hydrocarbons. When O_2 enters the reactor, $N_2(A)$ is quenched by impact (Gandhi and Mok 2012; Zheng et al. 2014)

$$N_2(A) + O_2 \rightarrow N_2 + O_2 \tag{9}$$

O_2 is electronegative through electron attachment (Dahiru et al. 2021; Kim et al. 2008). The increased O_2 partial pressure also increases the consumption of oxygen atoms, which results in the reduction of removal efficiency. This process will also lead to the formation of O_3 (Huang et al. 2011). As aforementioned, the reaction rate between O_3 and VOCs is low, and it will also consume active nitrogen atoms and molecules (Abd Allah et al. 2014), resulting in less decomposition of pollutants.

It can be seen from Fig. 5(b) that the change of EE of the reaction system with O_2 content was basically the same for all the voltages, and EE decreased with the increase of the O_2 content. When the O_2 content was higher than 3%, the maximum value of EE was obtained at 18 kV. When the O_2 content was less than 3%, and the maximum value of EE was obtained at 14 kV. However, it was found that under the condition of low O_2 content, especially when the O_2 content was 0%, a layer of dark brown material (petroleum or tar-like odor) (Karatum and Deshusses 2016) was attached to the inner wall of the reaction tube, which was characterized as an aromatic polymer (Guo et al. 2006). These substances affected the SED of the DBD system, as shown in Fig. 5(c). At 18 kV, when the O_2 content increased from 0% to 1%, the system SED increased from 1,267 to 1,351 J/L, and then almost remained stable. It shows that under the condition of low O_2 content, O_2 has little effect on the SED (Kim et al. 2008). In the absence of O_2, although removal efficiency and energy efficiency were high, more solid

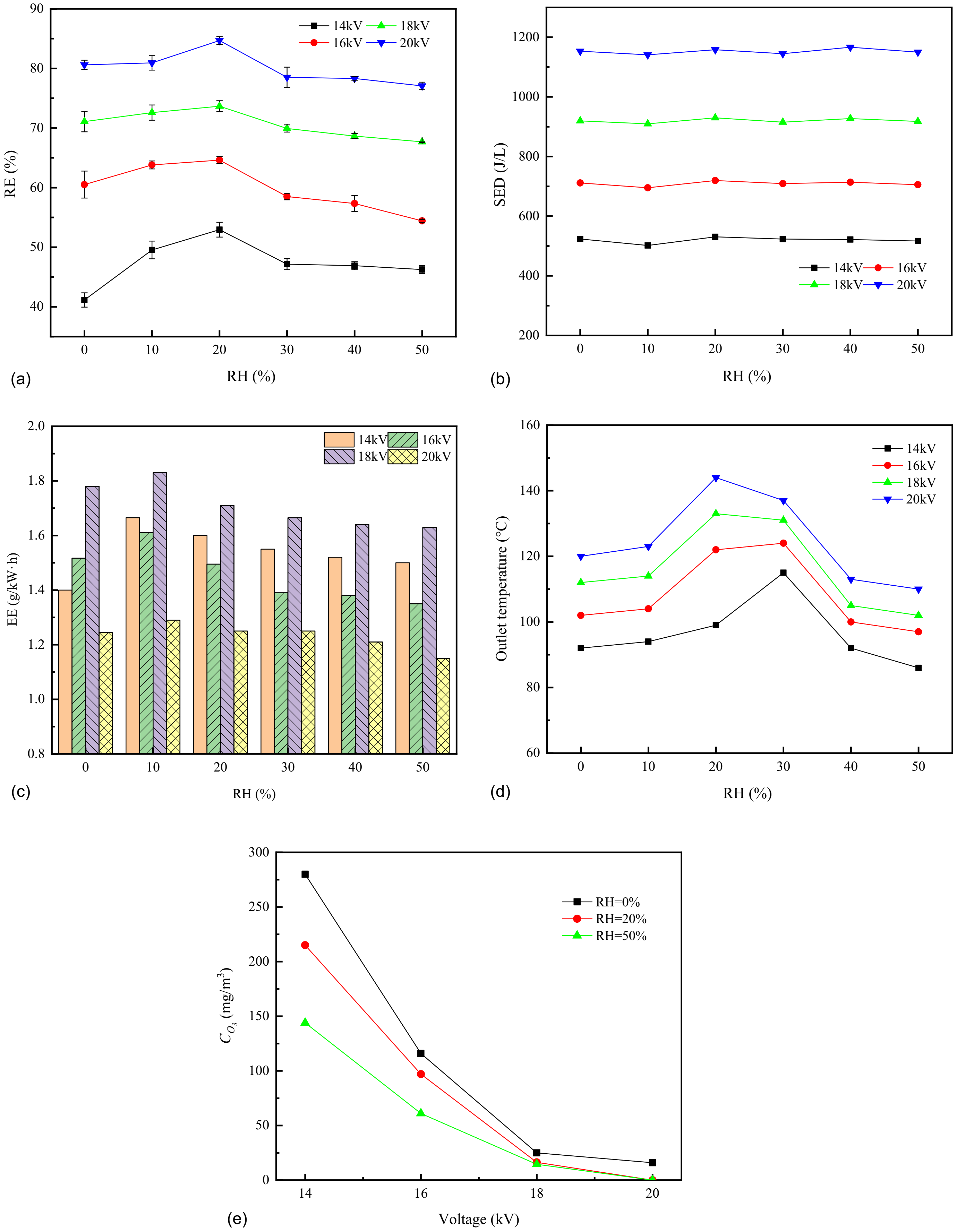


Fig. 4. Effect of relative humidity on CB removal: (a) removal efficiency; (b) energy efficiency; (c) specific energy density; (d) outlet temperature; and (e) O_3.

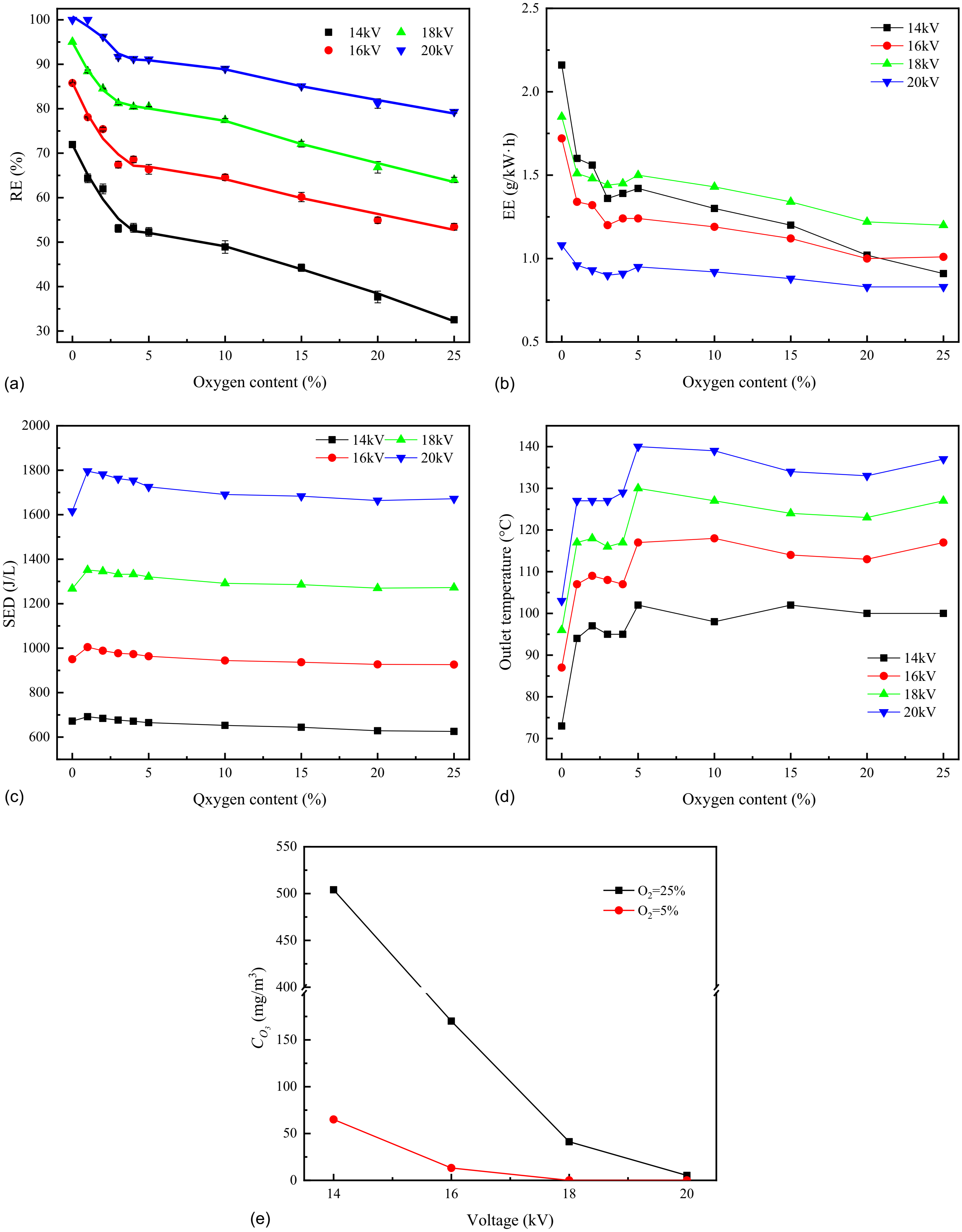


Fig. 5. Effect of O_2 on CB removal: (a) removal efficiency; (b) energy efficiency; (c) specific energy density; (d) outlet temperature; and (e) O_3.

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by-products generated, which would reduce the performance and life of the reaction system. This experiment also investigated the outlet temperature under various conditions, as shown in Fig. 5(d). The outlet temperature increased with the increase of oxygen addition, and the maximum temperature difference could reach 30°C. It indicated that under the condition of high O_2 content, more energy was used to increase the temperature of the reaction system, which reduced the removal efficiency of CB. As shown in Fig. 5(e), the O_3 concentration with oxygen content of 5% and 25% was determined. The results showed that the O_3 concentration of the oxygen content being 25% was significantly higher than that of 5%, because O_2 provides free radical O for O_3 generation. When the carrier gas is only nitrogen, there has no O and no O_3 in the plasma system, which is consistent with our detection results.

Effect of SO_2 on the Removal of Chlorobenzene

Generally, SO_2 and NO are the most common pollutants in the flue gas, and their concentration levels are generally several hundred mg/m^3. Therefore, the average concentrations of SO_2 being 0–500 mg/m^3 and NO being 0–600 mg/m^3 were investigated in the study.

Fig. 6(a) shows the relationship between the removal efficiency of CB and the SO_2 concentration. With the concentration of SO_2 increased from 0 to 500 mg/m^3, the removal efficiency was obviously improved. At 20 kV voltage, the removal efficiency of CB was 85.76% when the SO_2 concentration was 0 mg/m^3. When the SO_2 concentration rose to 400 mg/m^3, CB could not be detected at the outlet of the reactor. The emergence of SO_2 activated a series of basic reactions, resulting in the generation of a large number of activated radicals (Obradović et al. 2011), thus could improve the degradation of CB.

Fig. 6(b) shows the change of SO_2 removal efficiency under the same conditions. With the increase of SO_2 concentration, the SO_2 removal efficiency of DBD decreased gradually. In a separate NTP system, SO_2 is mainly oxidized into high valence oxides through OH, such as SO_3, HSO_3, and HSO_4 (Hong et al. 2019; Obradović et al. 2011). However, this experiment was carried out under dry conditions, and there was little OH in the reaction system, and other excited particles in the reaction system had little effect on the oxidation of SO_2 in the gas phase (Li et al. 2013). During the experiment, a yellow substance produced on the inner wall of the reaction tube, and the organic and inorganic experiments were carried out on it, respectively. After SO_2 was introduced and reacted, the by-product on the inner wall of the reactor was washed off with acetone and analyzed through the GC analysis. The results showed that only CB could be detected besides acetone. At the same time, the inner wall of the reaction tube was washed with deionized water. From the results, we found that the liquid was yellow. A large amount of white sediment was produced through adding barium chloride solution into the liquid, which proved the existence of SO_4^{2-}. SO_2 led to the formation of particles with SO_4^{2-} on the inner wall of the reaction tube. These particles would adsorb the CB and its by-products' molecules in the reaction tube. The removal of CB in the whole reaction included the degradation in the discharge process and the adsorption of particles, which improved the degradation efficiency of CB. As shown in Fig. 6(e), when 500 mg/m^3 SO_2 was introduced, the concentration of O_3 was the lowest, indicating that the addition of SO_2 consumed the O_3 of the system. We used ion chromatography to detect the anions in the tail gas, and found the existence of SO_4^{2-}, indicating that SO_2 reacts with O in the system, thereby inhibiting the formation of O_3. The introduction of SO_2 improved the EE to a certain extent, as shown in Fig. 6(c). The law was consistent with the removal efficiency of CB under various voltage conditions. As shown in Fig. 6(d), SED under each voltage fluctuated within a certain range, and the presence of SO_2 had little effect on it.

Effect of NO on the Removal of Chlorobenzene

It can be seen from Fig. 7(a) that the introduction of NO reduced the removal efficiency of CB. When NO concentration rose from 0 to 600 mg/m^3, the removal efficiency decreased from 41.31 to 22.89% at 14 kV. At 16 kV, the removal efficiency decreased from 60.66% to 32.89%. At 18 kV, the removal efficiency decreased from 72.71% to 44.3%. At 20 kV, the removal efficiency decreased from 83.14% to 58.02%. The removal efficiency of CB decreased by about 20% under all voltages. The existence of NO inhibited the degradation of CB and there was competition between NO and CB molecules in the NTP system. The bond energy of NO is 941.69 kJ/mol, which can not to be dissociated easily. Therefore, the higher its concentration is, the easier it is to inhibit the degradation of CB. Some literatures had studied the reaction kinetics of the system containing NO, and experimentally measured the generation and extinction trend of active groups such as O, OH in the discharge process. It was found that NO could consume O in the discharge process and generated oxides of NO_2, NO_3, N_2O_5, and other higher valence states (Cui et al. 2018). To study whether NO consumed O, this experiment measured the O_3 at different NO concentrations. As shown in Fig. 7(d), the addition of NO reduced the concentration of O_3 in the system. During the discharge process, O in the system was consumed, which led to the reduction of O_3 concentration. O is an important free radical for the removal of CB, so the existence of NO will compete with CB for the energy and free radicals required for degradation, and finally inhibit the degradation of CB. Different from SO_2, NO did not form particulate matter that could adsorb CB. The addition of NO significantly could reduce the EE, as shown in Fig. 7(b). The highest of EE could be gained at 18 kV and the lowest of EE could be gained at 20 kV. Fig. 7(c) shows that SED was not affected by NO and remained at the same level, which proved that NO did not affect the intensity of discharge in this DBD reactor.

Conclusion

The effects of gas temperature, relative humidity, O_2, SO_2, and NO on the removal of CB were studied by DBD. The main experimental results were as follows:

1. The removal efficiency of CB would first increase and then decrease with the increasing of the gas temperature. The gas temperature corresponding to the best removal efficiency under different voltages was also different, such as 50°C at 14 kV and 200°C at 18 kV.
2. When the relative humidity was controlled lower than 50%, the best removal efficiency of CB could be gained when the relative humidity was 20%. Moreover, a certain humidity would also improve energy efficiency and gas temperature at the outlet. The increase of humidity would reduce the concentration of O_3.
3. With the increase of oxygen content (0%–25%), the removal efficiency and energy efficiency of CB gradually decreased, but the outlet temperature increased to a certain extent. The increase of oxygen content promoted the formation of O_3.
4. After SO_2 was introduced into the reaction system, it was found that it could promote the degradation of CB and improve the energy efficiency, but it did not affect the input energy density. The addition of SO_2 inhibited the formation of O_3.
5. On the contrary to SO_2, the introduction of NO into the reaction system could inhibit the removal efficiency of CB and reduce

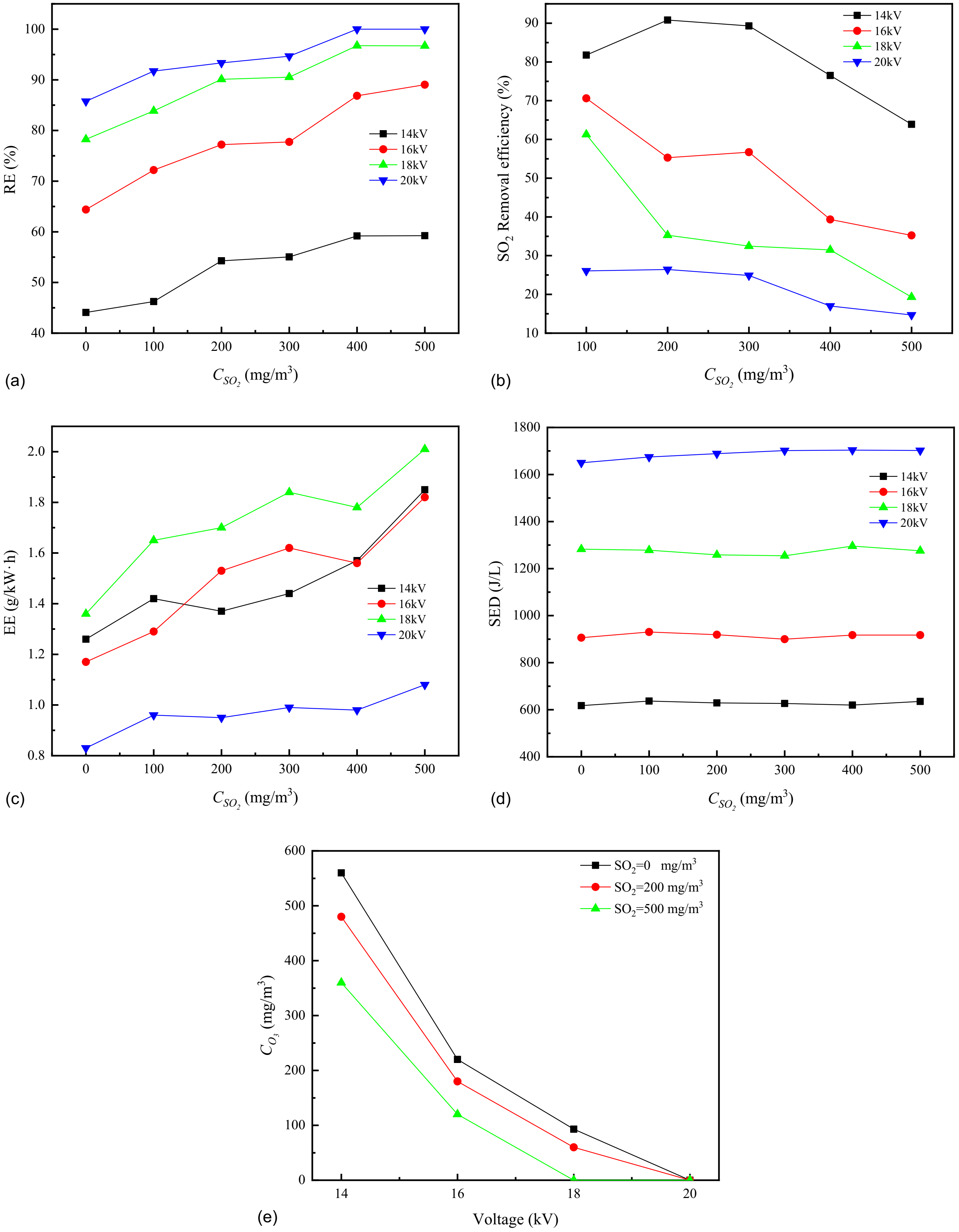

Fig. 6. Effect of SO_2 on CB removal: (a) removal efficiency; (b) SO_2 removal efficiency; (c) energy efficiency; (d) specific energy density; and (e) O_3.

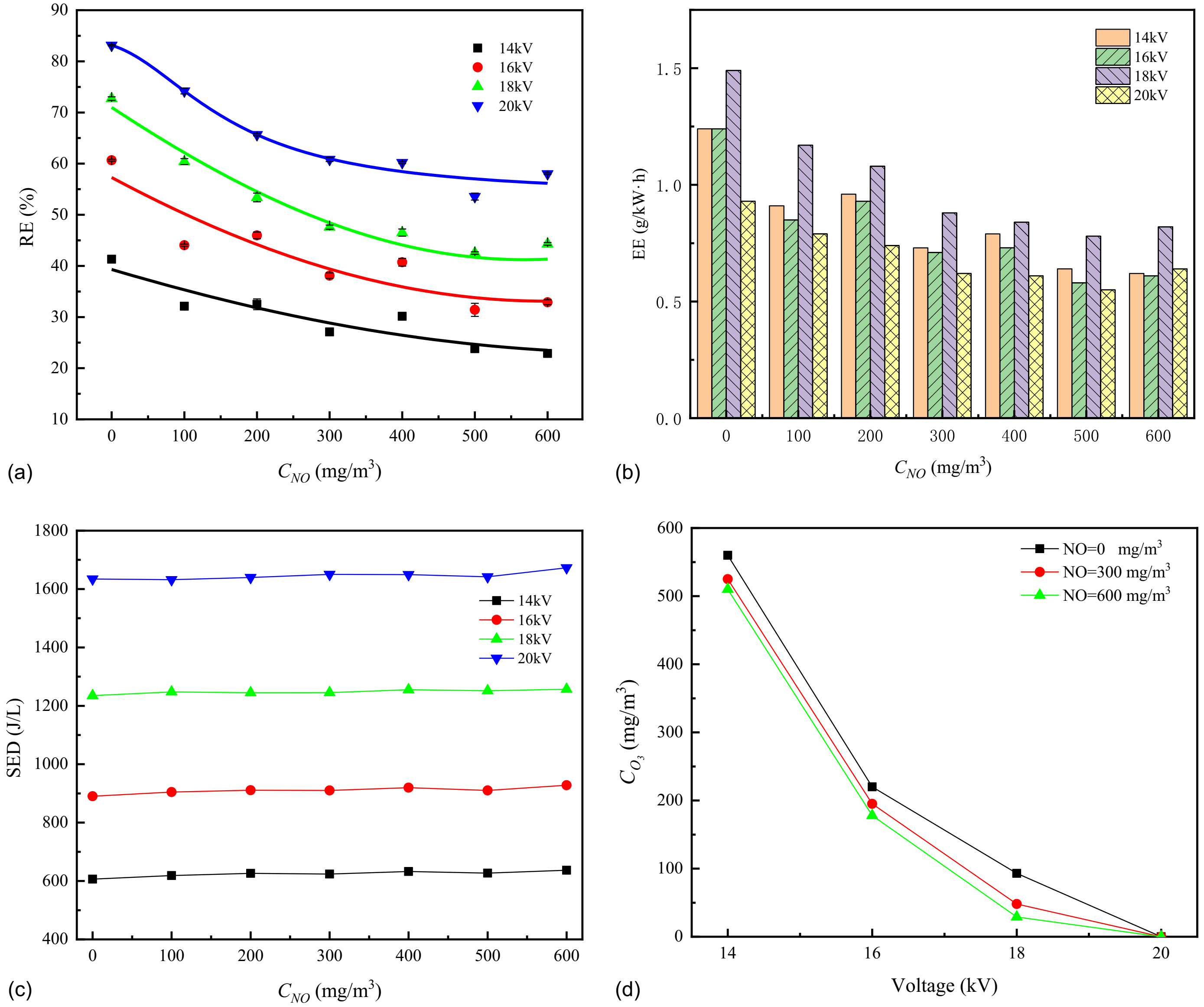

Fig. 7. Effect of NO on CB removal: (a) removal efficiency; (b) energy efficiency; (c) specific energy density; and (d) O_3.

the energy efficiency of the reaction. The addition of NO also inhibited the formation of O_3.

Through the experimental results, the influence laws of various factors in the degradation of CB by DBD could be obtained. A certain temperature and relative humidity would benefit to improve the removal efficiency and energy efficiency. The removal effect of CB would be better under the condition of low oxygen content. SO_2 would promote the removal of CB and NO would inhibit that.

Data Availability Statement

All data used during the study appear in the published paper.

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