



Removal of gaseous toluene and submicron aerosol particles using a dielectric barrier discharge reactor

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ABSTRACT

A lab-scale dielectric barrier discharge (DBD) reactor was fabricated, and gaseous and particulate contaminant removal tests were carried out under a range of DBD reactor operating conditions: applied voltage (5.0–8.5 kV), frequency (60–1000 Hz), upstream toluene concentration (50–200 ppm) and gas flow rate (1–5 L min⁻¹ or 0.48–0.096 s of gas residence time). The results suggested that the toluene removal efficiency (at 1 L min⁻¹, 100 ppm) increased (up to ~46%) either with increasing voltage (at 1000 Hz) or frequency (at 8.5 kV). The overall particle collection efficiency (at 1 L min⁻¹) improved (up to ~60%) with increasing voltage (at 1000 Hz) whereas the penetration of the particles increased (up to ~40%) with increasing frequency (at 8.5 kV). The toluene removal efficiency (at 8.5 kV, 1000 Hz, 100 ppm) decreased (down to ~29%) with increasing gas flow rate while the particle collection efficiency decreased slightly (maintaining ~60%) regardless of the flow rate. In addition, the toluene removal efficiency (down to ~41%) and carbon dioxide selectivity (down to ~43%) decreased with increasing upstream toluene concentration (at 5 kV, 1000 Hz, 1 L min⁻¹).

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1. Introduction

Gas streams within occupational and industrial regions contain a range of volatile organic compounds (VOCs), polycyclic aromatic hydrocarbons (PAHs), nitric oxides (NO_x), sulfur dioxide (SO₂), and aerosols. In particular, the VOCs from some sources, such as paints, chemical plants, semiconductors, and printing industries, can be quite toxic and their emission should be reduced. Therefore, the effective removal of VOCs has become an important issue in recent years.

The use of non-thermal plasma in many industries has increased in recent years. In non-thermal plasma, free electrons gain energy from an imposed electric field and lose energy through collisions with neutral gas molecules. This transfer of energy to the molecules leads to the formation of a variety of new species, including ions, metastable species, atoms and free radicals. Non-thermal plasma processes are used widely in the electronics industry to dope semiconductors and in microcircuit fabrication to etch and deposit thin films. Non-thermal plasma processes are being considered in air purification systems to remove trace contaminants by converting

them to less harmful species through reactions in a plasma reactor [1–4].

The dielectric barrier discharge (DBD) reactor is used widely to produce non-thermal plasma in atmospheric pressure, in which the electrical energy is used to create electrons with a mean kinetic energy of 1–10 eV [5,6]. A DBD reactor has one or two dielectric barriers to distribute microdischarges throughout the DBD volume. Roland et al. [7] examined the performance of a DBD in removing organic air pollutants (especially in low concentrations) by introducing ferroelectric and catalytically active materials into the DBD zone. Seto et al. [8] developed a microscale-patterned electrode type DBD reactor to decompose VOCs. Borra [9] briefly reviewed the fundamentals and various applications of DBD reactor including environmental pollution control. In addition to gas removal, the DBD has been applied to electrostatic charging and the collection of aerosol particles [10–12]. Byeon et al. [10] studied the collection of submicron aerosol particles for a hybrid two-stage electrostatic precipitator (ESP) consisting of a DBD as the particle charger and an ESP as the charged particle collector. Byeon et al. [11] reported the charge distribution of submicron sized aerosol particles passing through a DBD reactor. Kwon et al. [12] examined the charging characteristics of a novel aerosol charge neutralizer based on a DBD.

In this paper, a lab-scale DBD reactor was fabricated, and gaseous and particulate contaminant removal tests were carried out under a range of DBD operating conditions: applied voltage, fre-

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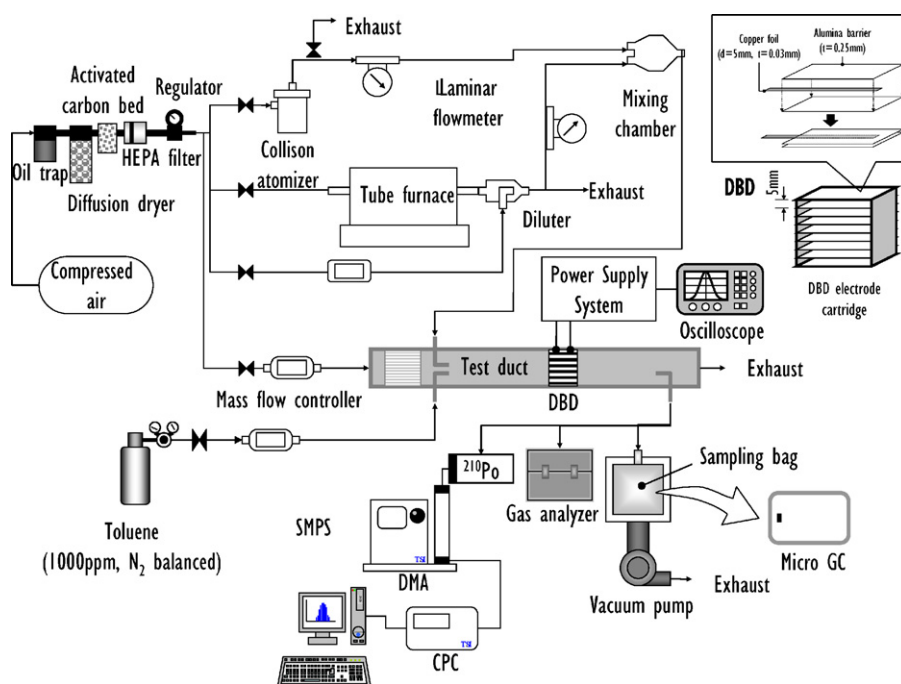


Fig. 1. Schematic diagram of the experimental set up.

quency, upstream toluene concentration, and gas flow rate. Toluene and submicron sized aerosol particles were selected as the gas-phase and particle-phase contaminants, respectively.

2. Experimental

An experimental setup was designed and constructed to evaluate the performance of a lab-made DBD reactor in removing gaseous toluene and aerosol particles from a simulated flow system (Fig. 1). The setup consisted of a continuous flow gas and particle generation systems, laboratory scale DBD reactor, and sampling and measurement systems. The temperature and relative humidity were monitored using a thermo-hygrometer (110, TESTO). Under all test conditions, the gas temperature and humidity were approximately 23 °C and 55%, respectively.

Toluene gas (1000 ppm) balanced with nitrogen (N_2) was prepared in a compressed gas cylinder and delivered to the test duct. Clean air was supplied to the test duct and mixed with toluene containing N_2 . The clean gas was realized from compressed air passing through an oil trap, activated carbon bed, and high efficiency particulate air (HEPA) filter. The desired toluene concentration at the test section was obtained by controlling the dilution ratio with clean air using mass flow controllers (Tylan).

Sodium chloride (NaCl) particles were generated stably from an electrically heated tube furnace at 700 °C. Dioctyl sebacate (DOS) particles were generated from a solution containing DOS (20 vol%) mixed with isopropyl alcohol (80 vol%) using a Collision atomizer. The particle size distribution of NaCl (geometric mean diameter of 31 nm, geometric standard deviation of 1.47, and total number concentration of 8.0×10^5 particles cm^{-3}) and DOS (geometric mean diameter of 213 nm, geometric standard deviation of 1.88, and total number concentration of 3.5×10^5 particles cm^{-3}) particles were obtained by controlling two laminar flow meters. The NaCl and DOS particles were mixed in a mixing chamber, and the mixture showed a bimodal size distribution, as described in a previous study [10].

The DBD reactor installed in an acrylic duct (5 cm × 5 cm) consisted of nine parallel plate electrodes, five of which were grounded. A high rectangular alternating current (AC) voltage (rectangular

waveform) was applied to the other four electrodes. This rectangular waveform voltage was chosen over the sinusoidal and triangular waveforms because of the linear correlation between the applied voltage and the current. The gap spacing between a grounded electrode and its adjacent powered electrode was 5 mm. Each electrode was made from 0.03 mm thick copper foil (5 mm of streamwise length and 40 mm of spanwise length) sandwiched between two 0.25 mm thick dielectric plates (alumina plates, 20 mm streamwise length and 50 mm of spanwise length). Non-thermal plasma was formed in the space between the electrodes. The gaseous toluene or submicron aerosol particles passed through the plasma region.

The discharge voltage and current waveforms were measured using a 100 MHz digital scope (TDS1012, Tektronix). The AC applied voltage is expressed as the root-mean-square (RMS) amplitude according to a voltage probe (P6015A, Tektronix). The discharge currents were measured using a current monitor (2877, Pearson). The discharge power was determined from the time-averaged power waveform obtained digitally from the voltage and current waveforms.

With the reliability test performed for continuous operation (>200 h), the maximum temperature of the DBD electrodes was about 41 °C and the DBD was performed stably without thermal cracking of the electrodes.

The toluene concentrations were measured using gas chromatography (3000 Micro GC, Agilent) with a thermal conductivity detector (TCD). The analyses were completed in less than 160 s. An exhaust gas analyzer (Greenline 9000, Eurotron Instruments S.p.A.) was coupled on-line at the downstream of the DBD reactor to monitor continuously the carbon dioxide (CO_2) and carbon monoxide (CO) concentrations from the oxidation of toluene.

The toluene removal efficiency (RE) is defined as follows:

$$RE = 1 - \left(\frac{[\text{toluene}]_{\text{downstream}}}{[\text{toluene}]_{\text{upstream}}} \right) \quad (1)$$

A parameter that reflects the total oxidation of toluene is the selectivity (S) of CO_2 , which is defined as the ratio between the CO_2 concentration obtained in the experiment and the CO_2 concentration for the total oxidation of the entire amount of toluene

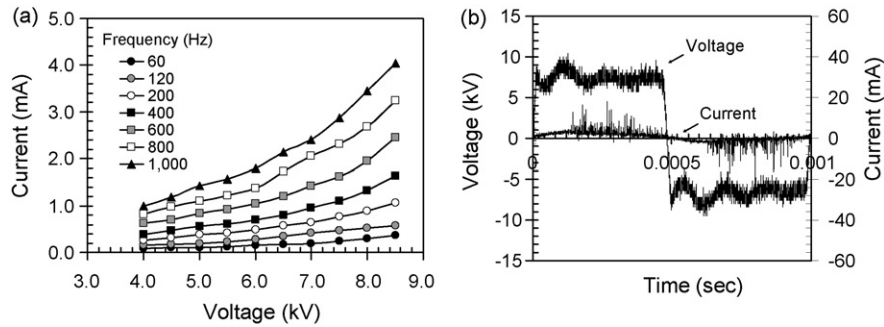


Fig. 2. Electrical characteristics of the DBD. (a) Voltage–current curve. (b) Voltage–current waveform (at 8.5 kV, 1000 Hz).

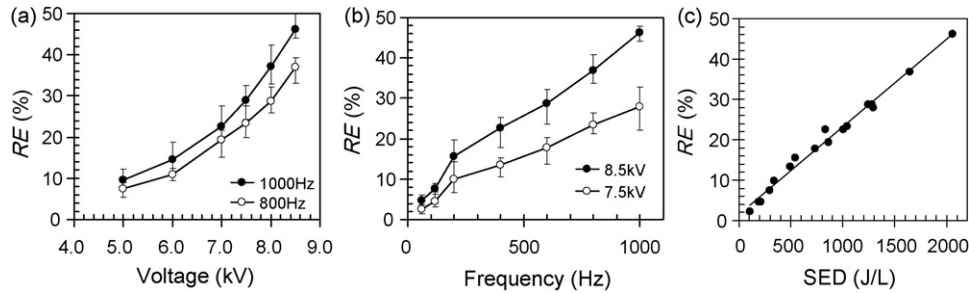


Fig. 3. Results of toluene removal at 1 L min⁻¹ flow rate and 100 ppm of toluene. (a) Function of the applied voltage. (b) Function of the voltage frequency. (c) Correlation between the SED and removal efficiency.

introduced ($C_6H_5CH_3 + 9O_2 \rightarrow 4H_2O + 7CO_2$):

$$S = \frac{[CO_2]_{downstream}}{[toluene]_{upstream} \times 7} \quad (2)$$

A scanning mobility particle sizer (SMPS, 3936, TSI), consisting of a differential mobility analyzer (DMA, 3081 TSI) and a condensation particle counter (CPC, 3022A, TSI), was used as the particle measurement system. An aerosol charge neutralizer containing a radioactive source ²¹⁰Po (2U500, NRD) was used in this study. The flow rates of the sample and sheath of the SMPS system were 0.3 and 3.0 L min⁻¹, respectively.

The fractional (grade) collection efficiency (CE) of the DBD reactor, excluding diffusional wall loss and including only electrostatic deposition, is defined as follows:

$$CE_{DBD}(d_p) = 1 - \left[\frac{C_{downstream}(d_p)}{C_{upstream}(d_p)} \right] \quad (3)$$

where $C_{downstream}(d_p)$ and $C_{upstream}(d_p)$ are the concentrations of the particle sizes (electrical mobility equivalent diameter, d_p) measured at downstream and upstream of the DBD reactor, respec-

tively. The overall collection efficiency is defined as follows:

$$CE_{DBD,overall} = \frac{\int_0^\infty CE_{downstream}(d_p)C_{upstream}(d_p)dd_p}{\int_0^\infty C_{upstream}(d_p)dd_p} \quad (4)$$

3. Results and discussion

Fig. 2a shows the voltage–current characteristics of the DBD. The discharge current increased with increasing applied voltage. Bluish filaments were visible when the voltages were >5 kV. Fig. 2b presents temporal current and voltage distributions showing pulse-like waveforms.

Fig. 3a and b shows the effects of DBD on the toluene removal efficiency (RE) at voltages of 5.0–8.5 kV and frequencies of 60–1000 Hz, respectively. The gas flow rate and upstream toluene concentration were 1 L min⁻¹ and 100 ppm, respectively. A higher voltage or frequency resulted in higher efficiency. The data shown in Fig. 3a and b are plotted in Fig. 3c with respect to the specific energy densities (SED, J/L), which are defined as follows

$$SED = \frac{\text{Discharge power}(W)}{\text{Gas flow rate}(L \text{ min}^{-1})} \times 60 \quad (5)$$

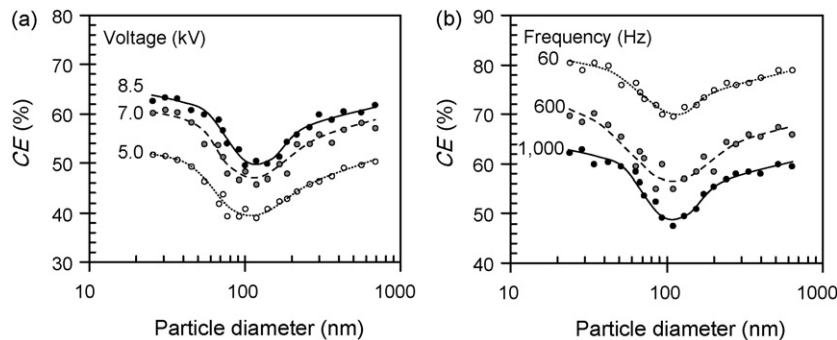


Fig. 4. Results of particle collection at 1 L min⁻¹. (a) Function of the applied voltage at 1,000 Hz. (b) Function of the voltage frequency at 8.5 kV.

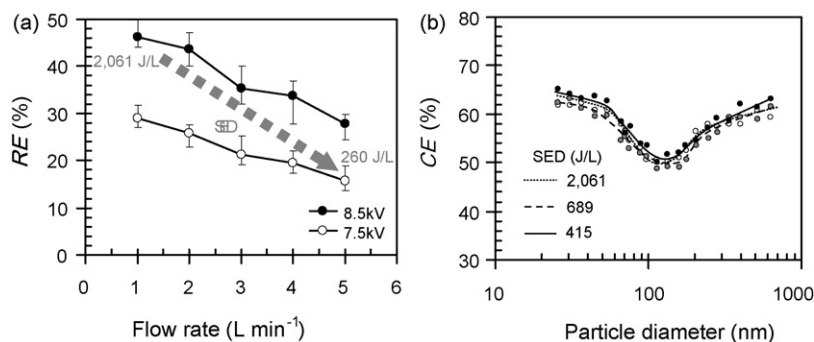


Fig. 5. (a) Toluene removal (at 1000 Hz, 100 ppm toluene) as a function of flow rate and (b) particle collection (at 8.5 kV, 1000 Hz) as a function of the flow rate.

For a higher SED, more electrons reacted with toluene and broke the bonds between toluene molecules [13], resulting in high toluene removal efficiency. The performance of a DBD can be evaluated using the parameter, β (J/L), from the simple equation [14–18]

$$\ln \frac{[\text{toluene}]_{\text{downstream}}}{[\text{toluene}]_{\text{upstream}}} = -\frac{SED}{\beta} \quad (6)$$

As shown in Fig. 3c, the parameter was approximately 3.7×10^3 J/L.

Fig. 4a shows the fractional particle collection efficiencies, CE_{DBD} , as a function of the particle diameter for voltages of 5.0–8.5 kV. The results were obtained at a frequency of 1000 Hz and an air flow rate of 1 L min^{-1} . The results suggest that the AC power applied to the DBD reactor charged the aerosol particles, which precipitated on the DBD reactor. The collection efficiencies for all particle sizes increased with increasing applied voltage because of the higher particle charging corresponding to enhanced particle motion in the DBD reactor. For details of the particle charge distribution in the DBD reactor, please refer to Ref. [11]. The fractional collection efficiency of the DBD reactor can be expressed using the following equation by assuming well-mixed flow caused by turbulence:

$$CE_{\text{DBD}}(d_p) = 1 - e^{-kEZ} \quad (7)$$

where E is the intensity of the applied electric field, k is a constant depending on the flow characteristics, and Z is the electrical mobility. At any applied voltage, for $d_p < \sim 100 \text{ nm}$, Z is proportional to $d_p^{-1/2}$, hence CE_{DBD} decreases with increasing d_p , while for $d_p > \sim 100 \text{ nm}$, Z is proportional to $d_p^{1/2}$ and thus CE_{DBD} increases with increasing d_p .

Fig. 4b shows the effect of the voltage frequency (60–1000 Hz) on CE_{DBD} . The results were obtained at an applied voltage and flow rate of 8.5 kV of 1 L min^{-1} , respectively. The data shows that the collection efficiency decreased with increasing frequency. In a DBD reactor, the fraction of aerosol particles collected depends on the amplitude of particle motion, Z_p , which is related to the particle electrical mobility, applied voltage, and voltage frequency of the DBD reactor. Indeed, the amplitude of particle motion was previously approximated in Refs. [9,19,20] for particles $< 100 \text{ nm}$, as follows:

$$Z_p \sim \frac{ZE}{2\pi F} \quad (8)$$

where F is the voltage frequency. Therefore, the voltage frequency affects the migration of charged particles in the alternating field cycle. For example, as discussed by Byeon et al. [10], at lower frequencies, the particles will spend most of each half-cycle migrating toward an electrode. At higher frequencies, the particles spend a significant fraction of each half-cycle with ions, which neutralize the charge existing at the end of the previous half-cycle. This

can cause migration in the wrong direction, resulting in little net progress toward the DBD electrode. Therefore, an increase in frequency has an adverse effect on the particle collection efficiency of the DBD reactor.

As shown in Fig. 5a, the gas flow rate affected the toluene removal efficiency. The toluene removal efficiency decreased monotonously from ~ 50 to $\sim 30\%$ for an upstream toluene concentration of 100 ppm, an applied voltage of 8.5 kV, and a frequency of 1000 Hz with increasing gas flow rate from 1 to 5 L min^{-1} . Increasing the gas flow rate (decreasing the residence time) caused that more amount of toluene was passing through the plasma zone during same time, so the ratio between the oxidized molecules and the total amount of toluene molecules would be lower [13], which resulted in a lower toluene removal efficiency. Fig. 5a shows the toluene removal efficiency and specific energy density as a function of the flow rate.

On the other hand, the particle collection efficiencies remained $> 60\%$ regardless of the flow rate (see Fig. 5b). Jidenko et al. [20] reported that the collection efficiency is independent of the gas flow rate at a longer gas residence time than a voltage period. In the present study, the residence times were $0.48\text{--}0.096 \text{ s}$ ($1\text{--}5 \text{ L min}^{-1}$ of flow rate), which were longer than the periods of applied voltages ($0.017\text{--}0.001 \text{ s}$ for 60–1000 Hz). Moreover, these residence times were sufficiently longer than those ($0.048\text{--}0.016 \text{ s}$) in a previous study [10] and the discharge currents did not vary significantly, ranging from 3.96 to 4.05 mA.

Correspondingly, the air ion concentration (N_{ion}), which can be estimated from the following expression:

$$N_{\text{ion}} = \frac{I}{eZ_{\text{ion}}EA} \quad (9)$$

where I is the measured discharge current, e is the elementary charge ($1.6 \times 10^{-19} \text{ C}$), Z_{ion} is the mobility of air ions ($1.6 \times 10^{-4} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1}$) and A is the area of the electrode, ranged $2.20\text{--}2.64 \times 10^{10} \text{ ions cm}^{-3}$ for gas flow rates of $1\text{--}5 \text{ L min}^{-1}$. For various particle sizes, the effect of flow rate on average particle charge was investigated from classical diffusion and field charging theories. The results showed that the average particle charges did not change much for gas flow rates of $1\text{--}5 \text{ L min}^{-1}$.

Fig. 6a shows the toluene removal efficiency as a function of the upstream toluene concentration between 50 and 200 ppm. The experiments were carried out at applied voltages of 7.5 and 8.5 kV with a constant voltage frequency of 1000 Hz. The removal efficiency of toluene decreased with increasing upstream toluene concentration. Fig. 6b shows the β -value as a function of the upstream toluene concentration:

$$\beta \sim 330 \sqrt{[\text{toluene}]_{\text{upstream}}} \quad (10)$$

Table 1 summarizes the operating parameters and results of the performance evaluation of the DBD reactor in this study, which

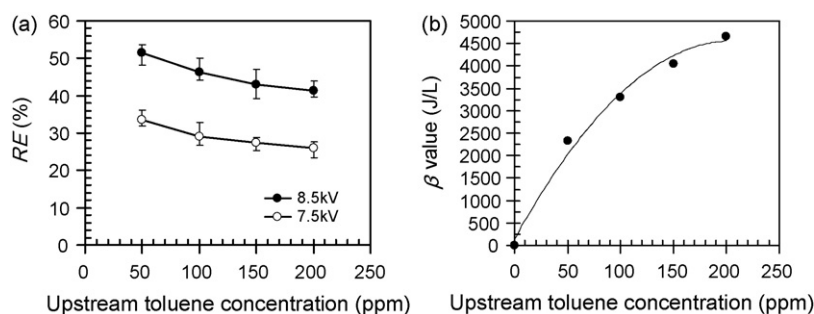


Fig. 6. (a) Toluene removal as a function of the upstream toluene concentration at 1000 Hz of frequency and 1 L min^{-1} of flow rate and (b) β parameter as a function of the upstream toluene concentration.

Table 1

Performance comparison between the present and other studies.

	Upstream toluene concentration (ppm)	Flow rate (L min^{-1})	Residence time (s)	SED (J/L)	RE (%)	Removed toluene/discharge power ($\mu\text{g/W}$)
Our study	50–200	1–5	0.096–0.48	259–2064	15–51	14.2–17.9
Guo et al. [5]	50	0.1	27	200–750	20–62	10.2–12.4
Delagrangé et al. [18]	240	0.315	>10	172	36	12.4
Magureanu et al. [21]	560	0.05–0.15	2.21–6.36	900–2700	5–25	6.3–12.3

are also compared with those of previous studies using DBD reactors with different configurations. The comparison shows that the amount of toluene removal (μg) per unit discharge power (W) in this study was the highest. This was attributed to the difference in DBD reactor configuration. The DBD reactor in this study was designed with a complete volume discharge (fully filled plasma in DBD volume).

The CO_2 selectivity (S) may be another useful parameter to assess the performance of the DBD reactor towards toluene removal. Table 2 shows the dependence of the selectivity on the upstream toluene concentration varying from 50 to 200 ppm, The applied voltage, frequency, and gas flow rate were 8.5 kV, 1000 Hz, and 1 L min^{-1} , respectively. For 50 ppm toluene, the selectivity to CO_2 was $\sim 66\%$ but decreased to $\sim 43\%$ with 200 ppm of toluene. The selectivity depends on a ratio between the amounts of active species (O radicals, etc., to be reacted with carbon atoms in toluene molecules) and toluene molecules, for example, a higher ratio causes a higher probability of CO_2 formation. During DBD operation, a relatively low oxygen concentration (i.e. relatively high upstream toluene concentration) led to the formation of CO due to the incomplete oxidation reaction between the active species and toluene related molecules [22,23]. Hence, the CO_2 : $\text{CO}_2 + \text{CO}$ ratio decreased with increasing upstream toluene concentration.

In addition, yellowish particles were observed on the surfaces of the DBD electrodes after toluene removal tests. This implied that the DBD electrically charged particles probably produced during the toluene removal in the discharge zone, which were, thereafter, collected on the electrodes. The production of the particles was confirmed using a scanning mobility particle sizer (SMPS) system; number concentration of 7×10^2 – 4×10^4 particles cm^{-3} and mean diameter of 15–90 nm. These particles could be oxi-

dized by operating the DBD reactor with or without toluene injection.

4. Concluding remarks

This study examined the removal of gaseous toluene and submicron aerosol particles using a plate-to-plate type DBD reactor under an AC electric field. For higher voltage and frequency, more electrons reacted with toluene and broke the bonds between toluene molecules, resulting in higher toluene removal efficiency. The collection efficiency increased with increasing applied voltage due to higher particle charging corresponding to an enhanced particle motion in the DBD reactor. On the other hand, the penetration of particles increased with increasing frequency. The collection of aerosol particles depended on the amplitude of particle motion, which is related to the particle electrical mobility; the amplitude is inversely proportional to the frequency. An increase in gas flow rate caused that more amount of toluene was passing through the plasma zone during the same time. Even though the amount of toluene molecules affected by the electron collision was expected to do not change since the nature of the plasma did not change, the ratio between the oxidized molecules and the total amount of toluene molecules would be lower, and then the toluene conversion decreased. The particle collection efficiency did not decrease much regardless of the flow rate, which was attributed to the longer residence times than periods of applied voltages. In addition, the CO_2 selectivity decreased with increasing upstream toluene concentration because the relatively low concentration of active species led to the formation of incompletely oxidized species, such as CO.

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Table 2

CO_2 selectivity at different upstream toluene concentrations at 8.5 kV of voltage, 1000 Hz of frequency, and 1 L min^{-1} of flow rate.

Upstream toluene concentration (ppm)	CO_2 (ppm)	CO (ppm)	$\text{CO}_2/\text{CO}_2 + \text{CO}$ (%)	S (%)
50	230	46	83.3	65.7
100	420	112	78.9	60.0
150	590	190	75.6	56.2
200	600	308	66.1	42.9

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