

Removal Characteristics of Benzene in Dielectric Barrier Discharge Process

Jae-Woo Chung*

Department of Environmental Engineering, Jinju National University, Jinju 660-758, Korea

(Received 14 January 2002; accepted 28 March 2002)

Abstract

The electrical and chemical properties of the dielectric barrier discharge (DBD) process for the benzene removal were investigated. The benzene removal was initiated with the applied voltage higher than the discharge onset value. The removal efficiency over 95 % was obtained at approximately $1.6 \text{ kJ liter}^{-1}$ of the electrical energy density. The increase of the inlet concentration decreased the removal efficiency. However, the benzene decomposition rate increased with the inlet concentration. While the increase of the gas retention time enhanced the removal efficiency, the decomposition rate decreased. Identification of the optimum condition between the decomposition rate and the removal efficiency is required for field applications of the DBD process.

Key words : Benzene removal, Electrical discharges, Operating parameter, Removal efficiency, Decomposition rate

1. INTRODUCTION

Volatile organic compounds (VOCs) are typical emission gases from automobile, painting, storing and pumping of gasoline, and oil refinery and chemical processes. VOCs have adverse effects on human health and environments by generating photochemical smog and lots of diseases, such as headache, dizziness, sore throats, etc. In particular, some VOCs such as benzene and chloroform are known as potential carcinogen compounds. Various technologies have been investigated for decomposing the VOCs, including catalytic oxidation, thermal decomposition, carbon adsorption, condensation and biological treatment. The application of nonthermal plasmas generated by electrical discharges has been an active research topic in recent years

(Lee and Jung, 2000; Heo *et al.*, 2000; Chang and Chang, 1997). The electrons in nonthermal plasmas have much higher energies than gas molecules. Therefore, the chemical reactions which do not occur under the normal condition can take place in gas streams. Since little energy is lost in heating gas molecules, the nonthermal plasma process can be more efficient method in the removal of gaseous pollutants than the conventional processes. Various techniques have been chosen to remove VOCs from gas streams by generating nonthermal plasmas, such as an electron beam, corona discharge, dielectric barrier discharge (DBD) and others (Won *et al.*, 2001; Urashima and Chang, 2000; Futamura *et al.*, 1997; Oda *et al.*, 1995). The DBD process can generate very stable discharges under the low temperature and atmospheric pressure. The DBD is characterized by a dielectric layer covers at least one of the electrodes, sometimes both. The dielectric material in DBD process effectively limits the

* Corresponding author.
E-mail : jwchung@jinju.ac.kr

charges in plasma so that the transition to filamentary or arc discharge may be prevented (Eliasson and Kogelschatz, 1991).

To date, the research trend on the discharge process for the removal of pollutants has been directed to identify pollutant removal efficiency and mechanism. However, electrical characteristics of the process are very important because discharge conditions dominantly affect the pollutant removal. Therefore, the simultaneous focusing on the electrical and chemical aspects of the process should be taken for the effective process development. The purpose of this study is the identification of electrical and chemical properties of the DBD reactor for the benzene removal. The effect of operating parameters such as applied voltage, electrical energy density, inlet concentration and gas retention time on the benzene removal was evaluated.

2. EXPERIMENTAL

Fig. 1 shows the schematic diagram of the experimental set-up, which consists of gas injection parts, a DBD reactor, an alternating current (AC) power supply

and analyzing instruments for electrical and chemical properties. The gas streams were controlled by MFC (mass flow controller, MKS 1179) in order to obtain the desired gas flow rate and inlet benzene concentration. Gaseous benzene was generated by passing a small amount of gas streams through the liquid benzene sustained in a constant temperature and mixed with the main gas streams.

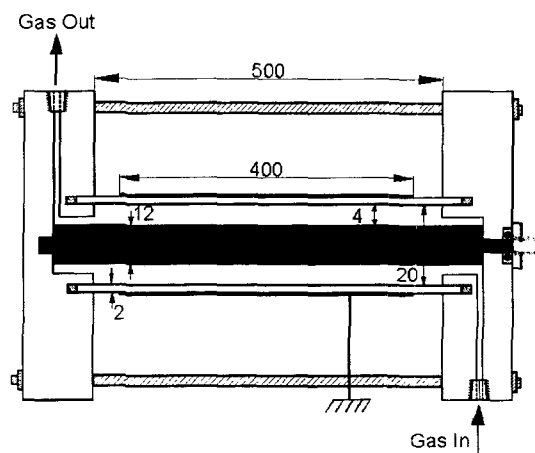


Fig. 2. Geometry of DBD reactor.

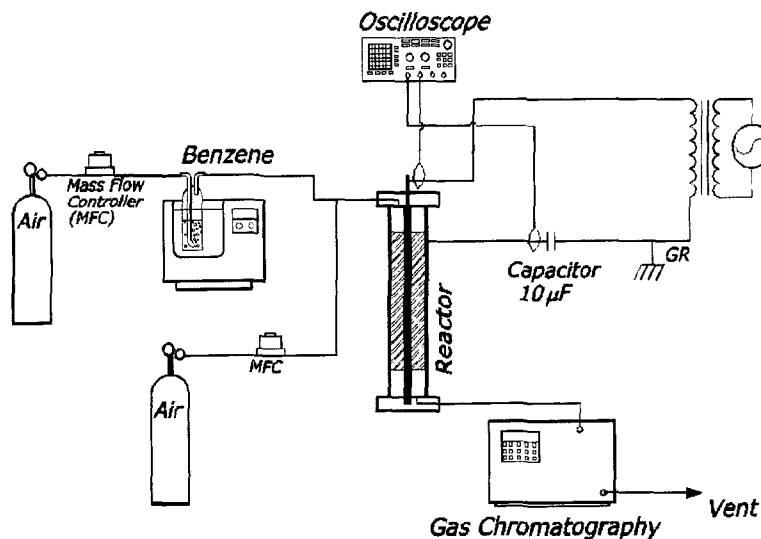


Fig. 1. Schematic diagram of experimental set-up.

The geometry of the DBD reactor is shown in Fig. 2. The reactor was made of a glass tube with the inner diameter of 20 mm, the thickness of 2 mm and the length of 500 mm. The inner electrode was made of a copper rod with the diameter of 12 mm. The gap distance passing the gas streams was 4 mm. A copper film tape was wrapped around the glass tube and served as the outer electrode. The length of the electrically active zone was 400 mm.

The AC power was applied to the DBD reactor. The alternating voltage was produced by a high voltage transformer with a voltage adjuster that varies from 0 to 220 volts. The operating frequency was 60 Hz. We obtained the output voltages from 0 to 27 kV in experiments.

A 1000X high voltage probe (Tektronix, P6015A) and a digital oscilloscope were used to measure the applied voltage to the reactor. A capacitor (10 μ F) was connected to the reactor in series to measure the discharge current and the charge. A 100X divider (Tektronix, P5100) was used to measure the voltage across the capacitor. The benzene concentration in gas streams before and after discharges was measured by a gas chromatograph (Varian, Star 3600CX) with a flame ionization detector (FID). An adsorption test was carried out to observe the adsorption trend of the benzene in the reactor. The outlet benzene concentration was monitored with time in a constant inlet concentration without discharges. We observed that the adsorption of the benzene in the reactor was negligible. However, the gas stream with a specific composition and a flow rate was maintained for 30 minutes for insuring the steady state condition in each test. All experiments were carried out at ambient temperature and pressure.

3. RESULTS AND DISCUSSION

3.1 Electrical properties of DBD

Fig. 3 shows typical voltage (V) and current (I) waveforms before and after the discharge onset. When the applied voltage was lower than the onset value, a

small amount of sinusoidal capacitive current was measured. The phase difference between the current and the voltage is one fourth of a cycle. Once the discharge starts with the applied voltage higher than the onset value, the phase difference is narrowed and much higher discharge current flows.

The discharge energy transferred to the DBD reactor can be calculated from voltage (V)–current (I) waveforms or charge (Q)–voltage (V) plots which are obtained by the electrical circuit shown in Fig. 1. The discharge energy can be obtained by integrating the product of the current and the voltage waveforms with time. The energy can be also obtained by using the Q–V plots. Fig. 4 shows Q–V plots with applied voltages. The energy dissipated per voltage cycle can be calculated by computing the area enclosed by the parallelogram (Carlins and Clark, 1982). Fig. 5 shows the comparison of the electrical energies calculated from

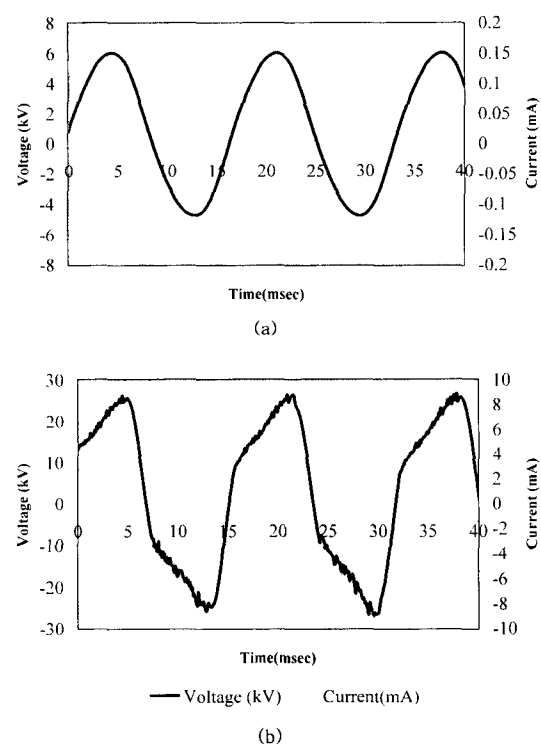


Fig. 3. Voltage and current waveforms before (a) and after (b) discharge onset.

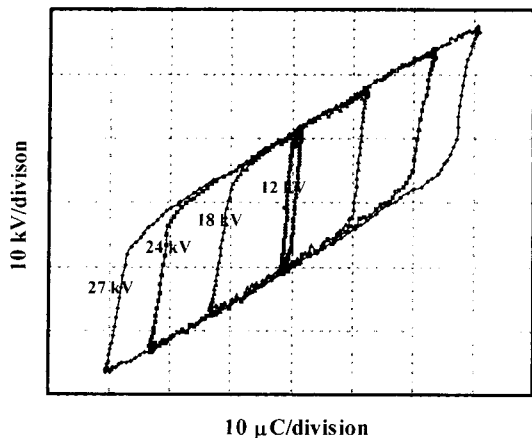


Fig. 4. Charge-voltage plots with applied voltages.

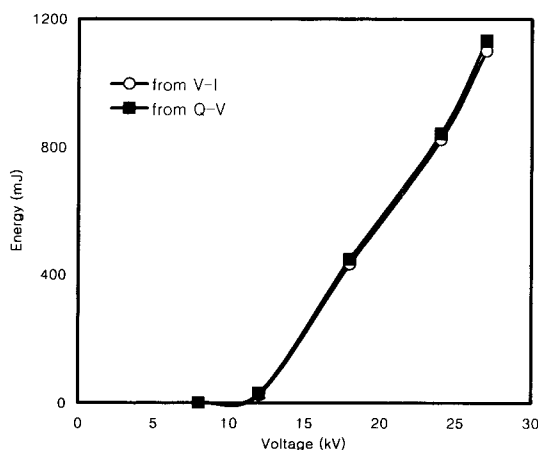


Fig. 5. Comparison of energies calculated from V-I waveforms and Q-V plots.

V-I waveforms and Q-V plots. It is observed that the energies calculated by two methods have nearly the same value.

3.2 Benzene removal characteristics

Fig. 6 shows a typical trend of benzene removal with applied voltages when the gas flow rate containing the 120 ppm of benzene was 2.5 liters per minute. The benzene removal initiated with the application of voltages higher than the discharge onset value. The

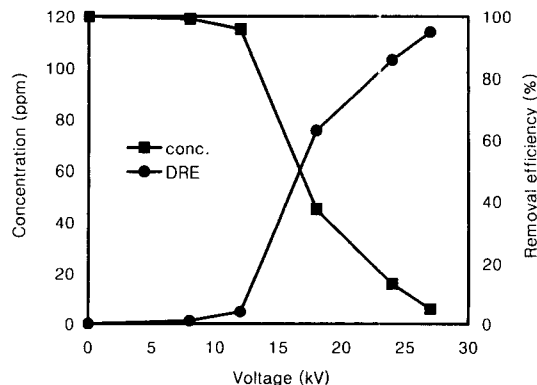


Fig. 6. Removal trend of benzene with applied voltage.

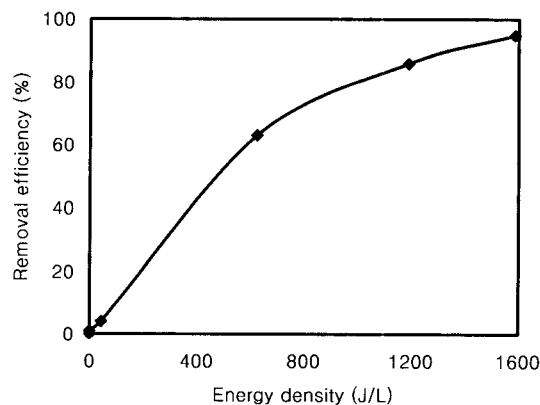


Fig. 7. Effect of electrical energy density on benzene removal.

removal efficiency increased with the applied voltage and approximately 95% of the inlet benzene was removed at the voltage of 27 kV.

Fig. 7 shows the removal efficiency as a function of the electrical energy density, which is defined as the transferred energy to the gas volume, as following,

$$ED = \frac{Ef}{Q} \tag{1}$$

where, ED is the electrical energy density (joules per liter), Q is the gas flow rate (liters per second), E is the discharge energy per voltage cycle (joules) and f is the frequency (hertz). The result shows that approximately

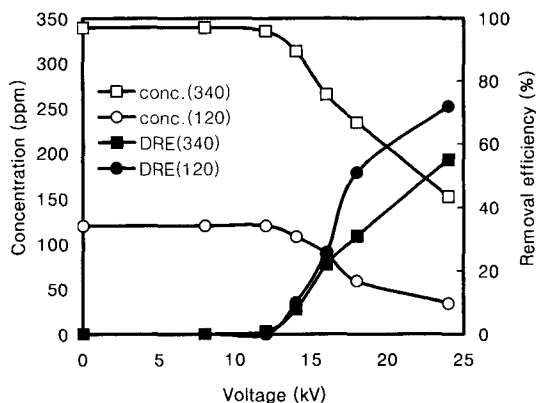


Fig. 8. Effect of inlet concentration on benzene removal.

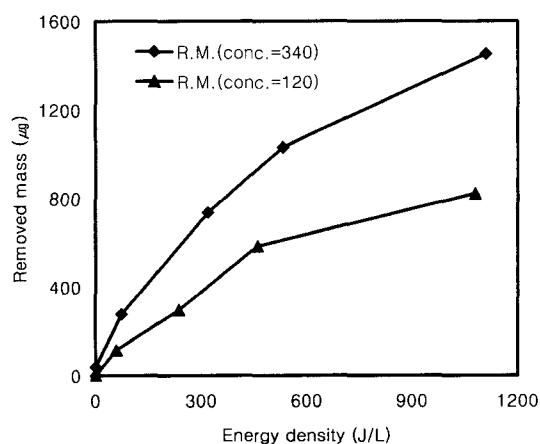
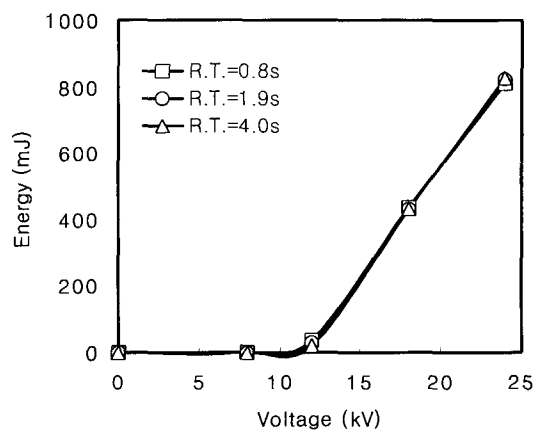


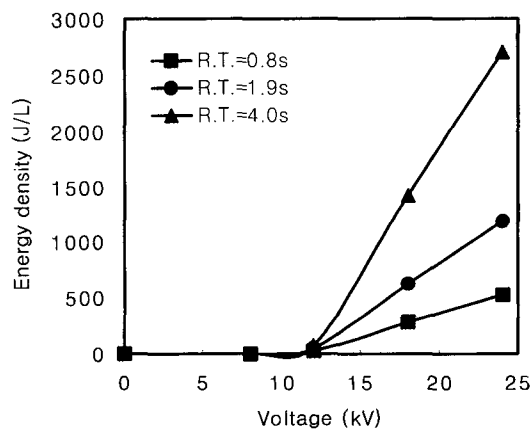
Fig. 9. Effect of inlet concentration on removed mass of benzene.

$1.6 \text{ kJ liter}^{-1}$ of the energy density is required to obtain the removal efficiency over 95%.

The inlet concentration effect on the benzene removal with the applied voltage is shown in Fig. 8. The constant flow rate of air containing 120 ppm and 340 ppm of the benzene was injected to the reactor. It was found that the higher inlet concentration resulted in the lower removal efficiency. The reduction of the removal efficiency at higher inlet concentration was already reported (Ogata *et al.*, 1999). In order to evaluate the inlet concentration effect on the benzene decomposi-



(a)



(b)

Fig. 10. Transferred energy (a) and energy density (b) during a voltage cycle as a function of gas retention time.

tion rate, we plotted the removed mass with the energy density in Fig. 9. We can see that the removed mass at the high concentration is higher than at the low concentration. This result shows that the decomposition rate increases with the concentration. It has been proposed that the VOC removal in electrical discharges be occurred by direct electron impacts, ion reactions and radical reactions (Urashima *et al.*, 1997). The increase of inlet concentration can enhance possibilities of these reactions. Therefore, the decomposition rate has a pro-

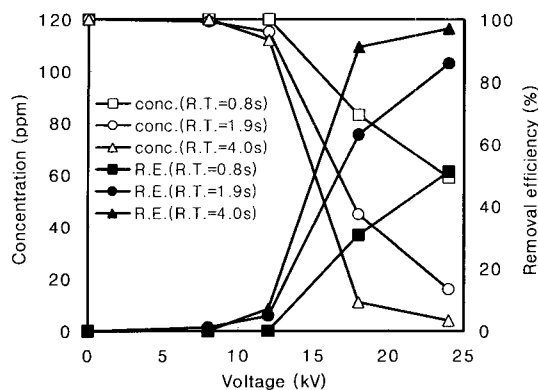


Fig. 11. Dependence of benzene removal on gas retention time.

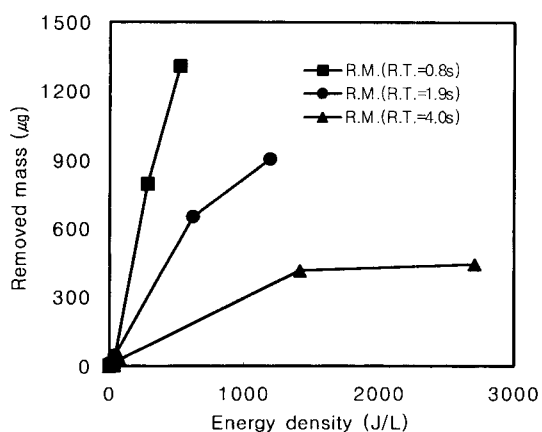


Fig. 12. Effect of gas retention time on removed benzene mass.

portional relationship with the VOC concentration.

The effect of the gas retention time on the benzene removal was investigated. The gas retention time was adjusted by controlling the gas flow rate with the same inlet composition. Fig. 10 shows the effect of gas retention time on the energy per voltage cycle and energy density. The energy transferred to the discharge was not remarkably changed by the variation of the gas retention time. The result shows that the gas flow rate has a negligible influence on discharge properties. However, the gas retention time significantly affected

on the energy density because the gas flow rate varied at nearly the same discharge condition. The energy density did rapidly increase with the retention time. Fig. 11 shows the dependence of the benzene removal on the retention time. The removal efficiency increased with the retention time. Fig. 12 shows the removed benzene mass with the energy density. The removed mass increased with the decrease of the retention time at the same energy density. From this result, it might be noted that the benzene decomposition rate increased with the decrease of the retention time.

4. CONCLUSIONS

In this study, the electrical and chemical characteristics of the DBD process for the benzene removal were investigated. The benzene removal initiated with the discharge by the application of voltages higher than the onset value. Electrical energies calculated from V-I waveforms and Q-V plots have the same value. The removal efficiency over 95% was obtained by applying approximately 1.6 kJ per liter of energy density. The removal characteristics of the benzene were affected by the energy density, the inlet concentration and the gas retention time. The increase of the inlet concentration decreased the removal efficiency. However, the benzene decomposition rate increased with the concentration. The variation of the gas retention time did rarely affect discharge characteristics. However, the benzene removal was significantly influenced by the retention time. The increase of the retention time enhanced the removal efficiency. On the contrary, the decomposition rate decreased with the gas retention time. The identification of the optimum condition between the decomposition rate and the removal efficiency has to be followed.

REFERENCES

Carlins, J.J. and R.G. Clark (1982) Ozone generation by

- corona discharge. In *Handbook of Ozone Technology and Applications* (R.G. Rice and A. Netzer Eds), Ann Arbor Science, Michigan, pp. 41–75.
- Chang, M.B. and C.C. Chang (1997) Destruction and removal of toluene and MEK from gas streams with silent discharge plasmas. *AiChE Journal* 43(5), 1325–1330.
- Eliasson, B. and U. Kogelschatz (1991) Nonequilibrium volume plasma chemical processing. *IEEE Transactions on Plasma Science* 19(6), 1063–1077.
- Futamura, S., A.H. Zhang, and T. Yamamoto (1997) The dependence of nonthermal plasma behavior of VOC's on their chemical structure. *Journal of Electrostatics* 42, 51–62.
- Heo, K.W., S.B. Yang, S.H. Lee, Y.K. Hong, S.Y. Shin, and J.H. Kang (2000) Study on the decomposition of some volatile organic compounds by photocatalyst plasma reaction. *Journal of Korean Society for Atmospheric Environment* 16(4), 373–380.
- Lee, B.K. and K.R. Jung (2000) Removal of volatile organic compounds by photo-catalytic oxidation. *Journal of Korean Society for Atmospheric Environment* 16(E), 39–46.
- Oda, T., A. Kumada, K. Tanaka, T. Takahashi, and S. Masuda (1995) Low temperature atmospheric discharge plasma processing for volatile organic compounds. *Journal of Electrostatics* 35, 93–101.
- Ogata, A., N. Shintani, K. Mizuno, S. Kushiya, and T. Yamamoto (1999) Decomposition of benzene using a nonthermal plasma reactor packed with ferroelectric pellets. *IEEE Transactions on Industry Applications* 35(6), 753–759.
- Won, Y.S., D.H. Han, W.S. Park, T. Stuchinskaya, and H.S. Lee (2001) Decomposition of trichloroethylene/air mixture by electron beam irradiation in a flow reactor. *Journal of Korean Society for Atmospheric Environment* 17(1), 97–104.
- Urashima, K., J.S. Chang, and T. Ito (1997) Destruction of volatile organic compounds in air by a superimposed barrier discharge plasma reactor and activated carbon filter hybrid system. In *Proceedings of the 1997 IEEE Industry Applications Society Annual Meeting, New Orleans*, pp. 1969–1974.
- Urashima, K. and J.S. Chang (2000) Removal of volatile organic compounds from air streams and industrial flue gases by nonthermal plasma technology. *IEEE Transactions on Dielectrics and Electrical Insulation* 7(5), 602–614.