

Destruction of Styrene in an Air Stream by Packed Dielectric Barrier Discharge Reactors

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This study presents the decomposition rates of styrene vapors with non-packed and packed bed dielectric barrier discharge reactors. The concentrations of intermediate byproducts at various plasma operation conditions were evaluated. The results showed that although styrene vapors could be almost completely removed at low styrene inlet concentration of 132 ppm, the selectivity of CO₂ as the major product was rather low in a non-packed bed reactor. It was found that solid carbon containing compound was the major byproduct. An increase in the styrene inlet concentration tended to reduce the styrene removal efficiency, it also led to increase in the solid byproduct. The reactors that packed with glass, Al₂O₃ or Pt-Pd/Al₂O₃ pellets could improve the styrene decomposition efficiency and reduce the formation of intermediate products, of which the best oxidation of styrene to CO₂ could be achieved with a Pt-Pd/Al₂O₃ packed bed reactor. The carbon byproducts could also be reduced if the reactor length was increased. The concentrations of ozone formed during the plasma process were also evaluated for the non-packed and packed bed reactors. The plasma reactor that packed with Pt-Pd/Al₂O₃ pellets was proved to have the lowest O₃ concentration.

KEY WORDS: Dielectric barrier discharge; styrene; catalyst; carbon balance.

1. INTRODUCTION

Styrene is a volatile aromatic compound and widely used as petrification material for many industrial applications such as resin, plastic and medicine. However, styrene will damage human beings and the environment once it is released into the atmosphere. According to the Clean Air Amendment of USEPA in 1990, styrene is listed among the 189 hazardous

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air pollutants. Therefore, the development of an effective control technique is very important to eliminate its detrimental effects. Many techniques have been successfully developed to control the emission of volatile organic compounds (VOCs), for example, thermal decomposition, catalytic oxidation, carbon adsorption, and condensation. Additionally, the application of nonthermal plasma technologies to decompose the hazardous air pollutants in gas streams is extensively investigated in recent years.⁽¹⁻³⁾

Dielectric barrier discharge system is a well-known technique to produce ozone.⁽⁴⁾ In addition, dielectric barrier discharge (DBD) systems were very effectively to decompose many aromatic compounds including benzene,⁽⁵⁾ toluene,⁽¹⁾ *p*-xylene,⁽⁶⁾ and styrene,⁽⁷⁾ etc. The utilization of DBD reactors can decompose VOCs; but, unfortunately, the carbons of VOCs can not be oxidized to CO₂ completely during the plasma decomposition processes. The reported conversion of carbon atoms to CO₂ is only between 30% and 65% and thus a large amount of CO and organic byproducts will be released into the environment.⁽⁸⁾ In addition, the increase of inlet toluene and benzene concentrations can lead to more CO formation. Anderson *et al.* found that higher initial styrene concentration also lead higher probability of polymer formation in a nonpacked DBD reactor.⁽⁷⁾

In order to improve the performance of plasma systems on VOCs decomposition, various materials were packed into the plasma zone of DBD reactors. The packing inside the plasma zone can enhance plasma density because the surface discharge produced at the contact point among packing material would distribute the plasma homogenously inside the chamber. When Al₂O₃,⁽⁹⁾ Ag/TiO₂,⁽¹⁰⁾ Pt/Al₂O₃⁽¹¹⁻¹²⁾ and LaCoO₃ catalysts⁽⁸⁾ were placed in plasma reactors, the catalytic reaction could occur on the surface of these packing materials to enhance VOCs decomposition and, more importantly, to decrease CO formation.

The objective of this study is to evaluate styrene decomposition using wire-tube DBD and packed DBD reactors. Glass, Al₂O₃ and Pt-Pd catalyst were used as packing materials in the packed DBD reactors. In particular, the carbon balance for the gaseous carbon byproducts, solid carbon byproducts, CO and CO₂ formation during styrene decomposition in different DBD reactors were investigated in the present study as well.

2. EXPERIMENTAL

A schematic diagram of the experimental system is illustrated in Fig. 1. The experimental apparatus was composed of three units, including a styrene gas generation unit, a plasma reactor, and a gas analysis

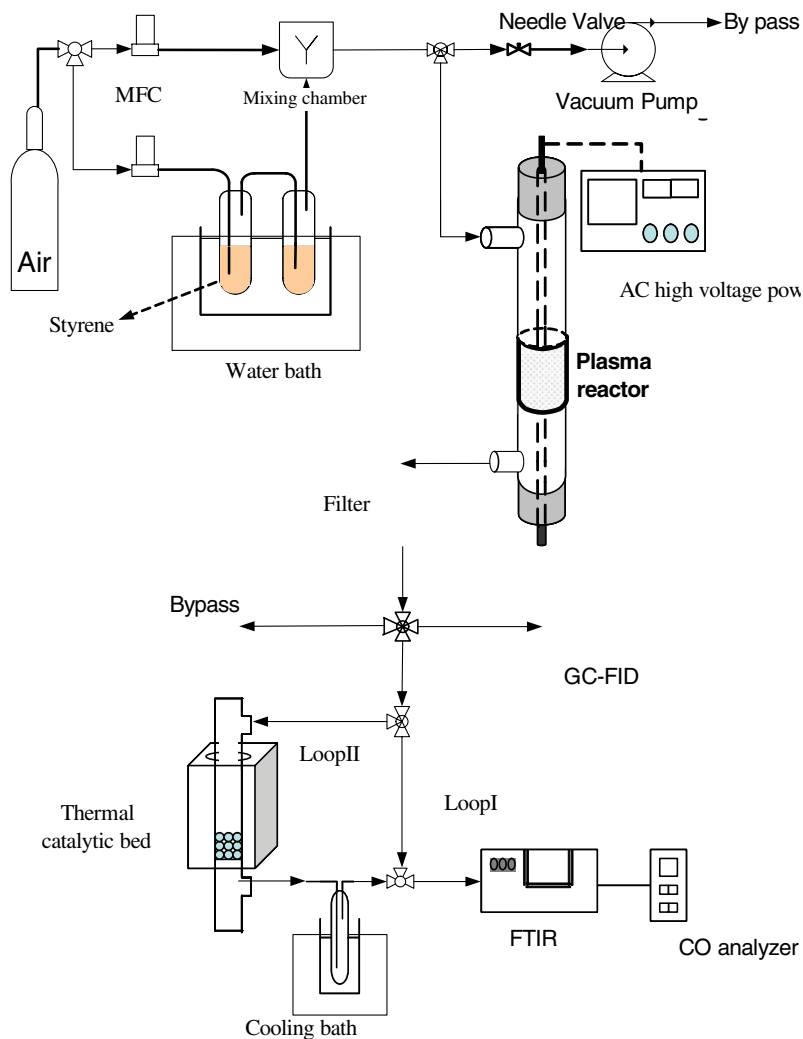


Fig. 1. Schematic of the experimental apparatus.

unit. A dry-grade compressed gas tank was used to supply air streams (80% N_2 and 20% O_2). The styrene stream was prepared by purging pre-purified gas through the aeration cylinders. The concentration of styrene could be controlled by adjusting either the water bath temperature or the diluting airflow rate. The flow rates of all air streams were regulated by

mass flow controllers (Brooks, 5850). And the excess airflow was removed by a bypass device.

The DBD reactor investigated in this study was a typical wire-tube system. A stainless steel wire (5 mm diameter) as an inner electrode was suspended along the axis of a Pyrex-glass tube (inner diameter: 20 mm). The outer electrode (ground electrode) was made of stainless steel wire mesh and was wrapped around the outside of the glass tube. The effective length of plasma zone in the DBD reactor could be adjusted by the length of outer electrode. The 60 Hz AC power was applied to the inner electrode. In addition, the temperature of the reactor was maintained at room temperature (approximately 25°C) by a cooling fan.

The performance of a packed-bed reactor was also evaluated in this study. Glass, Al₂O₃ and Pt-Pd/Al₂O₃ pellets (K.J. Environmental Technology Co. Ltd., Taiwan) with the same diameter of 5 mm were used as the packing materials and they were placed in the plasma region between two electrodes. The contents of Pt and Pd on the Al₂O₃ pellets were quantified by an Atomic absorption spectrometer (Hitachi Z8100) following the dissolving these metals with a hydrofluoric acid solution. The contents of Pt and Pd on Al₂O₃ pellets were 0.081 wt.% and 0.075 wt.%, respectively.

The concentrations of styrene in airflows at the inlet and the outlet of the testing reactors were analyzed by a gas chromatographer coupled with a flame ionization detector (GC-FID) (China Chromatography 79800) and a packed column (SP-1200, 2-m length). The decomposition efficiency of styrene is defined as

$$\eta = \frac{C_{\text{in}} - C_{\text{out}}}{C_{\text{in}}} \times 100\% \quad (1)$$

where C_{in} and C_{out} are the inlet concentration and the outlet concentration of styrene, respectively.

A filter was employed in downstream of the plasma reactor in order to prevent the formed organic aerosol or solid materials entering to the analyzers. A Testo535 on-line analyzer was used to measure the CO concentration in the outlet airflow. CO₂ and ozone concentrations in the outlet gas flows were analyzed by a Fourier transformation infrared spectrometer equipped with a gas cell (FTIR, BRUKER, VECTOR 22, Infrared Analysis Inc., model 2.4-P. A., Front detector: DTGS). The spectrum of FTIR was 2 cm⁻¹ resolution using the Blackman-Harris-3-term with apodization and zerofilling factor of 2. The scanner velocity was 10 kHz. The sampling streamline was passed through the FTIR gas cell and then was introduced to the CO analyzer at a flow rate of 500 ml/min.

In order to understand the total carbon distribution of products in the effluents, the amount of carbon in gaseous phase and solid phase were

also analyzed in this study. The carbon by-products from styrene decomposition could be simplified as five parts:

- $C_8H_8 \rightarrow$ (a) C_8H_8
(b) CO
(c) CO_2
(d) Organic byproducts in gas phase (not including styrene)
(e) Organic byproducts in solid phase

The concentrations of styrene, CO and CO_2 in effluents could be determined by the sampling loop I. When the sampling stream was switched to the loop II, the thermal catalytic-bed reactor was employed to convert total gaseous organic compounds and CO in the stream into CO_2 completely. Figure 2 shows that the only CO_2 was observed in the FTIR spectra as the styrene passed through the thermal catalytic-bed reactor at $400^\circ C$. Thus, the amount of carbon of organic byproducts (not including styrene) in gaseous phase, [C]-gas, and that in solid phase, [C]-solid, can be calculated by Eqs. (2) and (3), respectively.

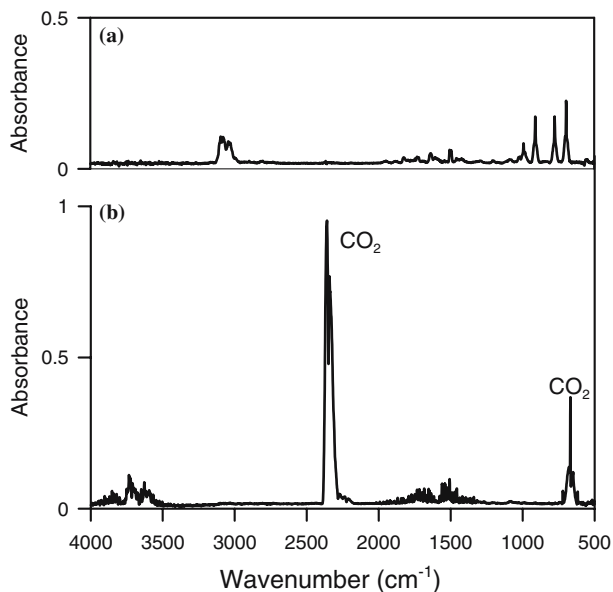


Fig. 2. FTIR spectrum patterns (a) before and (b) after thermal catalytic-bed reactor at $400^\circ C$ with inlet styrene concentration of 408 ppm.

$$[C]\text{-gas} = [\text{CO}_2]_{\text{II}} - [\text{styrene}]_{\text{I}} \times 8 - [\text{CO}_2]_{\text{I}} - [\text{CO}]_{\text{I}} \quad (2)$$

$$[C]\text{-solid} = [\text{styrene}]_{\text{inlet}} \times 8 - [\text{CO}_2]_{\text{II}} \quad (3)$$

where $[\text{styrene}]_{\text{inlet}}$ is the concentration of styrene in the inlet streams before being decomposed by DBD reactors. The symbols of I and II in the suffixes represent the concentration in the loop I and II, respectively.

3. RESULTS AND DISCUSSION

The experiments were carried out to examine the styrene decomposition by the DBD reactors with/without different packing materials at 25°C. The operating flow rate was set at 600 mL/min and the initial styrene concentration was 408 ppm in this case study. The length of plasma zone in the DBD reactors was 2 cm. After the outlet concentrations of styrene equaled to the inlet concentration, 20 kV of AC power was applied to start the plasma process. The styrene decomposition in different DBD systems as a function of operating time is shown in Fig. 3. The concentration of styrene in the effluents of nonpacked and DBD reactor packed with glass pellets (glass/DBD) immediately responded to the power on and the concentration of styrene decreased to a stable level within 3 min. But, the styrene decomposition of the DBD reactor packed with Al₂O₃ pellets (Al₂O₃/DBD) and DBD reactor packed with Pt–Pd/Al₂O₃ pellets (Pt–Pd/DBD) showed a delay response. The delay response of an Al₂O₃/DBD reactor, probably as a result of desorption process, was consistent to others' work in the treatment of VOCs.⁽¹³⁾ In addition, the surface area of Al₂O₃ pellet (293.4 m²/g) was larger than that of Pt–Pd coated pellet (198.6 m²/g). Thus, the amount of styrene absorbed on the Al₂O₃ pellet exceeded that on Pt–Pd coated pellets in that the longer time was required for the decomposition. The decomposition of styrene by an Al₂O₃/DBD reactor needed 100 min to reach a stable level. By contrast, they were only 33 min for Pt–Pd/DBD reactor and 5 min for nonpacked and glass/DBD reactors.

The decomposition efficiency of styrene on the four types of DBD reactors under steady-state are showed in Fig. 4. As shown, the removal of styrene was enhanced by glass pellets packing as well. For example, at applied voltage of 14 kV, the decomposition efficiency of a nonpacked DBD reactor was only 9%. When the glass pellets were packed into the DBD reactor, the decomposition efficiency was elevated up to 49%. The enhancement of the removal of VOCs was a result that the surface discharge produced at each packing glass pellets contact point distributed the plasma homogenously inside the reactors.⁽¹⁾ When the Al₂O₃ pellets were utilized as packed-bed in our testing reactor, the removal efficiency

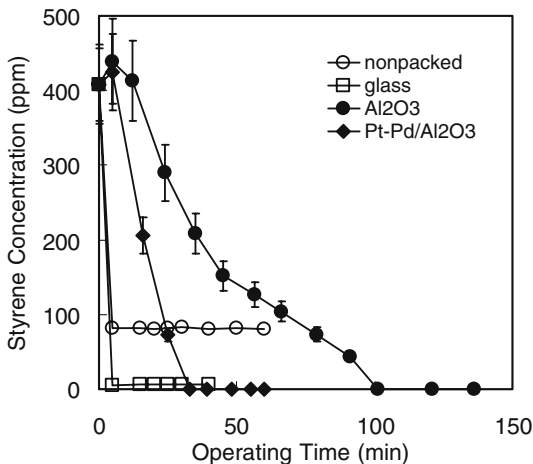


Fig. 3. Styrene decomposition efficiency as a function of operation time over different DBD reactors with plasma zone length of 2 cm at applied voltage of 20 kV (initial styrene concentration = 408 ppm and flow rate = 600 mL/min).

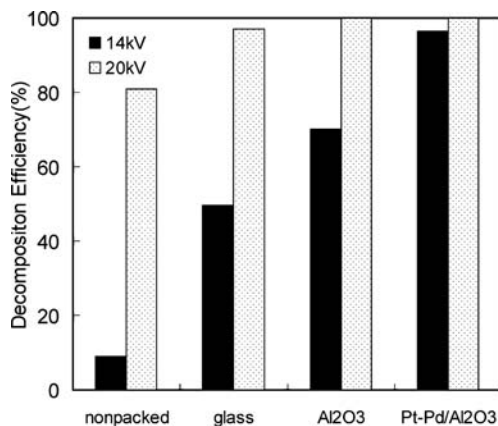


Fig. 4. Styrene decomposition efficiency in different DBD reactors with plasma zone length of 2 cm (initial Styrene concentration = 408 ppm and flow rate = 600 mL/min).

was greatly enhanced. The improvement is because of the produce of short-lived oxidizing species in the inner volume of porous surface of Al₂O₃ pellets.⁽¹³⁾ In addition, the catalytic reaction occurs on the surface of the Pt-Pd coated pellets led more styrene decomposition to achieve a decomposition efficiency of 96% at 14kV. Figure 4 also indicates that the decomposition efficiency increased as the increase of applied voltage.

When the applied voltage was increased to 20 kV, the styrene could be destructed completely by the $\text{Al}_2\text{O}_3/\text{DBD}$ and Pt–Pd/DBD reactors.

The concentrations of CO and CO_2 in the effluents from the decomposition of styrene by the four types of DBD reactors are showed in Fig. 5. The CO_2 concentration in the effluents seems to increase with the increase of the decomposition of styrene. The maximum concentration of CO_2 was observed using the Pt–Pd/DBD reactor. Figure 5 also shows that a higher amount of CO was observed when the DBD reactor equipped with packed-beds. However, the CO concentrations from $\text{Al}_2\text{O}_3/\text{DBD}$ and Pt–Pd/DBD reactors were lower than that from the glass/DBD reactor. The porous and catalytic material in a DBD reactor could change the ratio of CO and CO_2 in the products. The ratios of $\text{CO}_2/(\text{CO} + \text{CO}_2)$ for glass, Al_2O_3 and Pt–Pd coated pellets packed reactors at 20 kV were 41%, 48% and 57%, respectively. This observation suggests that a Pt–Pd coated pellet packed-bed is a potential tool to increase the oxidation of hydrocarbon compounds in a plasma reaction.

The selectivity to CO, CO_2 , C-gas and C-solid with respect to carbon conversion in the decomposition of styrene can be expressed as follows:

$$S_{\text{CO}} = \frac{[\text{CO}]}{[\text{C}_T]} \times 100\% \quad (4)$$

$$S_{\text{CO}_2} = \frac{[\text{CO}_2]}{[\text{C}_T]} \times 100\% \quad (5)$$

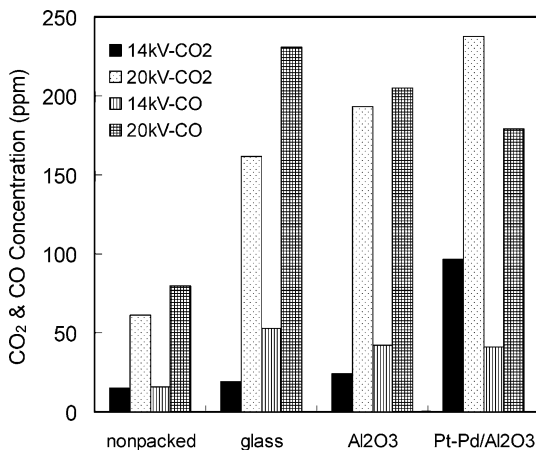


Fig. 5. The formation of CO and CO_2 in the styrene decompositions from different DBD reactors with plasma zone length of 2 cm. (initial styrene concentration = 408 ppm and flow rate = 600 mL/min).

$$S_{\text{CO}_x} = \frac{[\text{CO}] + [\text{CO}_2]}{[\text{C}_T]} \times 100\% \quad (6)$$

$$S_{\text{C-gas}} = \frac{[\text{C}]_{\text{-gas}}}{[\text{C}_T]} \times 100\% \quad (7)$$

$$S_{\text{C-solid}} = \frac{[\text{C}]_{\text{-solid}}}{[\text{C}_T]} \times 100\% \quad (8)$$

where $[\text{CO}]$, $[\text{CO}_2]$, and $[\text{CO}_x]$ are the CO, CO₂, and (CO + CO₂) concentrations after the reactor, $[\text{C}]_{\text{-gas}}$ and $[\text{C}]_{\text{-solid}}$ as defined by Eqs. (2) and (3) with respect to the amount of carbon of organic byproducts (not styrene) in gaseous phase and that in solid phase, respectively. $[\text{C}_T]$ the total carbon reacted during the decomposition of styrene.

$$[\text{C}_T] = ([\text{C}_8\text{H}_8]_{\text{initial}} - [\text{C}_8\text{H}_8]_{\text{final}}) \times 8 \quad (9)$$

where $[\text{C}_8\text{H}_8]_{\text{initial}}$ and $[\text{C}_8\text{H}_8]_{\text{final}}$ are styrene concentrations in the gaseous streams before and after the reactor, respectively. Although Fig. 4 shows that the styrene was completely destructed in the Al₂O₃/DBD and Pt-Pd/DBD reactors at 20 kV. However, the carbon of styrene could not be completely converted to CO or CO₂. As shown in Fig. 5, the maximum S_{CO_x} of the Pt-Pd/DBD reactor for the decomposition of styrene was only 12%. This result indicates that the most of carbon in the decomposition of styrene was converted to other hydrocarbons rather than CO or CO₂. In fact, we did find benzene, toluene, and benzene acetaldehyde by the on-line GC-FID system as well as some brown residues on the effluent gas tube wall.

Figure 6 shows that the decomposition efficiency of a nonpacked DBD reactor could be increased with the decrease of inlet styrene concentration. The completely decomposition was observed at the inlet styrene concentration of 132 ppm. As seen, the selectivity of CO and CO₂ also increased with the decrease of inlet styrene concentration. However, the maximum selectivity of CO and CO₂ were only about 10% and the selectivity of C-gas was 9%. By contrast, the selectivity of C-solid exceeded 72%. The phenomenon indicates that solid species was the major byproduct of styrene decomposition in a nonpacked reactor. In addition, the higher the initial inlet concentration caused the higher the probability of polymer formation. As shown, the selectivity of C-solid increased from 72% to 91% as the inlet styrene concentration increased from 132 to 1008 ppm.

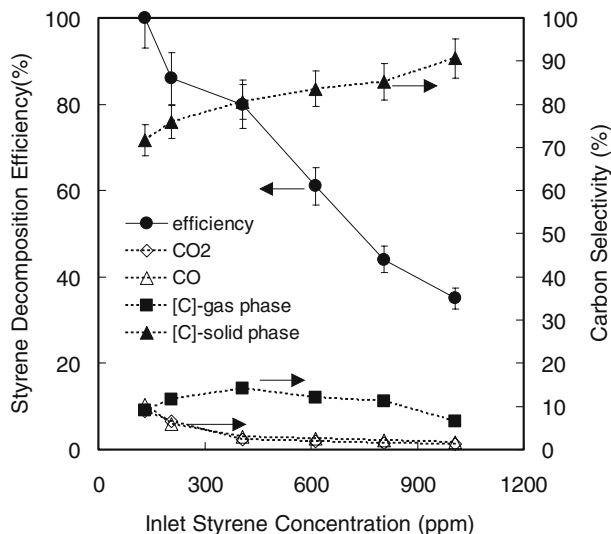


Fig. 6. Styrene decomposition efficiency and carbon selectivity as a function of inlet styrene concentration in a nonpacked DBD reactor with plasma zone length of 2 cm at applied voltage of 20 kV.

Figure 6 indicates that the inlet styrene concentration of 132 ppm could be destructed by DBD systems. However, the carbon conversion during the decomposition of styrene showed that only a few proportion of carbon was fully oxidized to CO₂ by a DBD reactor with plasma length of 2 cm (corresponding retention time of 0.65 s). The flow residence time in the plasma zone of DBD reactors with glass or BaTiO₃ pellets for hydrocarbons decomposition ranged from 2.83 to 30 s in the literature.^(1,14–17) In order to understand the influence of plasma zone length on the formation of hydrocarbon byproducts during plasma oxidation, the plasma zone of the DBD reactor was extended by varying the length of outer electrode. An increase in the lengths of plasma zone tended to increase flow residence time (plasma reaction time), it also increase the total input plasma power into gas streams to led more hydrocarbon destruction.^(14,17) The operating flow rate, applied voltage, and initial concentration of styrene were set at 600 mL/min, 20 kV and 132 ppm, respectively. The plasma lengths employed were from 10 to 60 cm; therefore, the flow residence time ranged from 3.26 to 20.00 s for nonpacked DBD reactors and they would be changed from 1.46 to 8.77 s for packed-bed DBD reactors. Figure 7 shows the relationship between the lengths of plasma zone and input power. As shown, the input power of reactors increased linearly with the

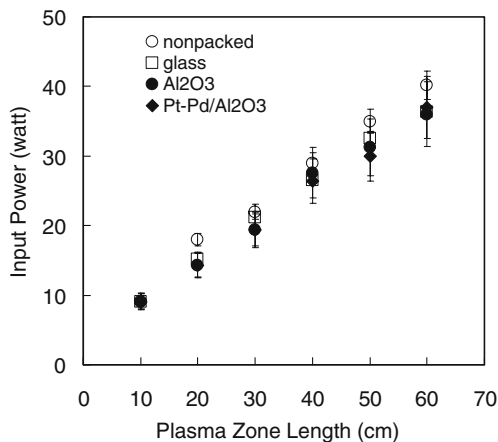


Fig. 7. Discharge power as a function of plasma zone length in different DBD reactors at applied voltage of 20 kV (initial styrene concentration = 132 ppm and flow rate = 600, mL/min).

lengths of plasma zone from 9 to 40 W. The input power of nonpacked bed reactor was slightly higher than that of packed-bed reactors at the same applied voltage. Higher space charge effect between the packing pellets may retard current and power output at the same applied voltage. The packing pellets in the reactor retarded input power was also observed in our previous studies.^(1,17) However, the results indicate that the influences of packing material on plasma characteristics among packed DBD reactors are not obvious.

The selectivity of C-solid and C-gas compounds decreased and the selectivity of CO and CO₂ increased as the increase of the length of plasma zone in all the DBD reactors (Fig. 8). Notably, the decomposition efficiency of styrene in all DBD reactors achieved 100%. However, these selectivities were quite different for these four DBD systems. As shown in Fig. 8(a), the solid compound was the major byproduct for the nonpacked DBD reactor with a plasma zone length of 10 cm (corresponding flow residence time: 3.2 s) where its selectivity was as high as 72%. In addition, the solid compounds were difficultly removed by the nonpacked DBD reactor even the plasma zone length was increased to 60 cm (corresponding flow residence time: 20 s). Additionally, the maximum selectivity of CO and CO₂ were only 40% and 31%, respectively.

The selectivities of carbon byproducts, for the glass and Al₂O₃ pellet packed DBD reactors, as a function of plasma lengths are displayed in Fig. 8(b) and (c), respectively. As displayed, the solid compounds

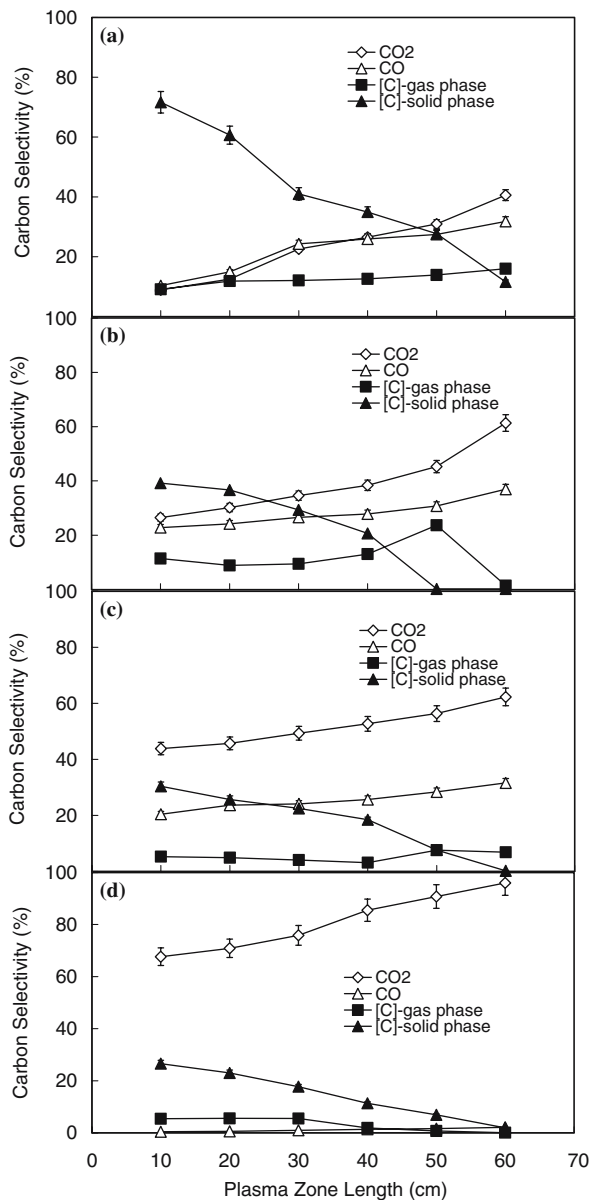


Fig. 8. Compare the differences in carbon selectivity between the different plasma zone lengths of reactors of (a) nonpacked DBD, (b) glass/DBD, (c) Al₂O₃/DBD, (d) Pt-Pd/DBD at applied voltage of 20 kV. (initial styrene concentration = 132 ppm and flow rate = 600 mL/min).

were reduced and the selectivity of CO_2 was enhanced by the glass/DBD and Al_2O_3 /DBD reactors as compared with the nonpacked DBD reactors. For example, the selectivity of solid compounds for the glass/DBD and Al_2O_3 /DBD reactors were reduced to 39% and 30% at a plasma zone length of 10 cm (corresponding flow residence time: 1.5 s), respectively. Remarkably, when the plasma zone length was increased to 60 cm (corresponding flow residence time: 8.7 s) for glass/DBD and Al_2O_3 /DBD reactors, the solid byproducts were completely removed and the selectivity of CO_2 was increased to 62%. However, Fig. 8(a) to (c) show that the CO could not be removed from gas streams by simply passing them through DBD reactors. The reason for the observed phenomenon is probably that CO is more difficultly ionized than hydrocarbon byproducts in the glass/DBD reactors owing to its high ionization potential of 14 eV, which exceeds that of some hydrocarbons (styrene: 8.46 eV, benzene: 9.24 eV, phenol: 8.49 eV, benzene acetaldehyde: 9.3 eV, toluene: 8.8 eV).⁽¹⁸⁾ Thus, plasma energy of glass/DBD reactors would react with hydrocarbons rather than CO. This finding also suggests that when CO is once produced during VOCs decomposition process, it would be difficultly removed. Thus, the nonpacked DBD, glass/DBD and Al_2O_3 /DBD reactors could not properly conquer this CO problem.

When the Pt–Pd coated pellets were utilized as packed-bed placed in the plasma zone, the solid compounds were significantly reduced as shown in Fig. 8(d). The solid compound was no more a major byproduct and styrene was more effectively oxidized to form CO_2 in short retention time. For example, the selectivity of CO_2 (67%) was larger than that of solid compound at a plasma zone length of 10 cm (corresponding flow residence time: 1.5 s). In addition, the incompletely oxidized byproduct (CO) was suppressed by Pt–Pd/DBD reactors and the selectivity of CO was kept within 2%. The hydrocarbon byproduct was completely removed within 60 cm (corresponding flow residence time: 8.7 s) and the conversion of CO_2 was as high as 96%. It indicates that Pt–Pd catalyst can effectively prevent the formation of CO, gaseous hydrocarbon and solid residues during the decomposition of styrene and has catalytic activity to oxidize the intermediated CO and hydrocarbon byproducts to CO_2 . Therefore, a Pt–Pd packed-bed is a potential tool to enhance the selectivity of CO_2 to solve the problem of carbon deposition and incompletely oxidized byproduct emission during the decomposition of styrene inside a DBD reactor.

Figure 9 displays the FTIR spectra of the emissions from the different DBD reactors at 20 kV. The retention times of treated gas in plasma zone of different reactors were similar to each other and they were 9.7 s (corresponding plasma zone length of 30 cm) for a nonpacked reactor and 8.7 s (corresponding plasma zone length of 60 cm) for packed reactors,

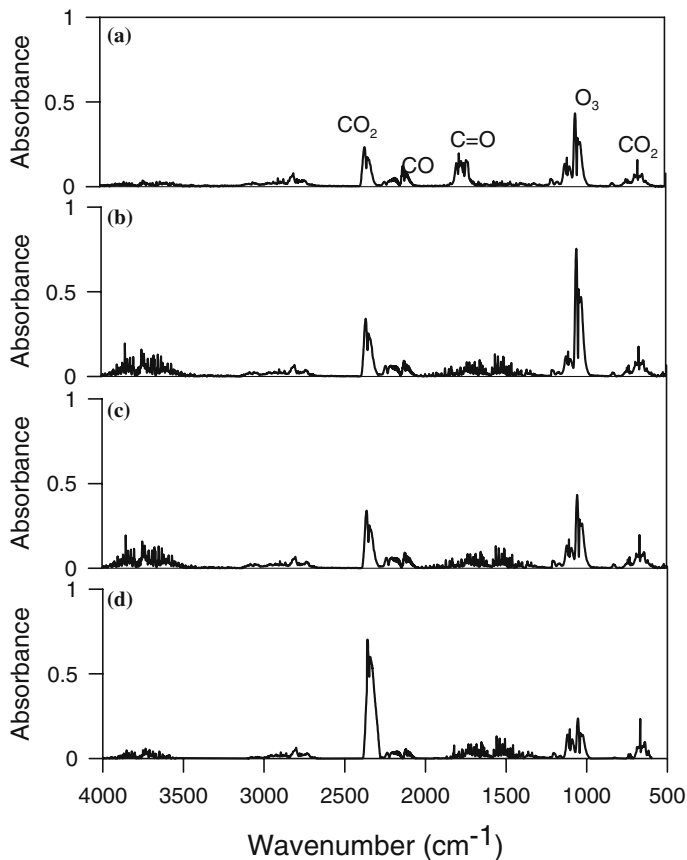


Fig. 9. FTIR spectrum patterns of effluent from (a) nonpacked DBD reactor; (b) glass/DBD reactor; (c) Al_2O_3 /DBD reactor; and (d) Pt-Pd/DBD reactor at 25°C with inlet styrene concentration of 132 ppm at 20 kV (Flow residence time for (a): 9.7 s and (b-d): 8.7 s).

respectively. As seen, the peaks at the wavenumber of 2358 and 1052 cm^{-1} indicate the existence of CO_2 and ozone, respectively. In addition, the wavenumber between 1600 and 1800 cm^{-1} is C=O group band. The C=O group only appeared in the emission of the nonpacked reactor that suggests that gaseous hydrocarbons in the nonpacked reactor were difficultly removed as compared with the packed reactors. Although the conversion of CO_2 in a glass/DBD reactor was larger than that in a nonpacked reactor, the amount of ozone production was also enormous. When the Al_2O_3 and Pt-Pd coated pellets were utilized as packing materials in DBD reactors, the ozone emission could be reduced.

In the plasma process, ozone is an unavoidable byproduct produced by the reactions among various radicals that are the products of the dissociation of excited O_2 . These side-completing reactions also affect the complete destruction and oxidation of styrene and byproducts to CO_2 . The ozone concentrations in the air streams containing zero or 132 ppm styrene from different packed reactors as a function of the length of plasma zone are shown in Fig. 10(a) and (b), receptively. As seen, the ozone concentration increased with the increase of the length of plasma zone. The maximum concentration of ozone was observed in the Glass/DBD reactor. Figure 8(a) shows that the ozone concentration in the effluents from varying DBD reactors followed the order Glass/DBD > Al_2O_3 /DBD > Pt-Pd/DBD. This result indicates that packing glass pellets could enhance the space plasma density in the DBD reactors. This resulted in an increase in the hydrocarbon removal but it also led to more

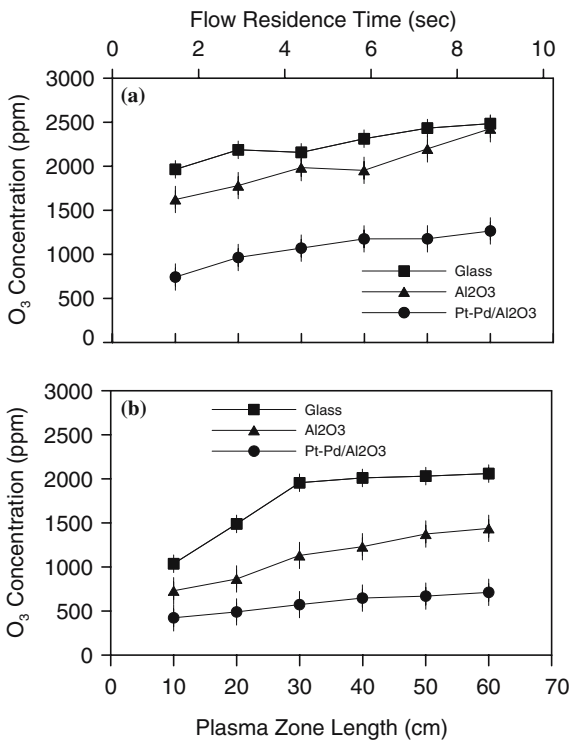


Fig. 10. Ozone production as a function of plasma zone length in packed DBD reactors in (a) only air stream and (b) styrene of 132 ppm at applied voltage of 20 kV.

ozone formation. The ozone formation in $\text{Al}_2\text{O}_3/\text{DBD}$ reactors was less than that in Glass/DBD reactors, this result supports that ozone could be adsorb on the porous surface of Al_2O_3 pellets. The Pt and Pd were catalysts with a high activity in the decomposition of ozone.⁽¹⁹⁾ Thus, the ozone concentration in the effluents from Pt-Pd/DBD reactor could be significantly reduced.

By comparing with Fig. 10(a) with Fig. 10(b), one can see that the existence of styrene in the air stream could reduce the ozone production. The reduction of ozone concentration in Glass/DBD reactors is owing to that the formation of ozone requires active oxygen species, while the presence of hydrocarbons would consume them.

The $\text{Al}_2\text{O}_3/\text{DBD}$ and Pt-Pd/DBD reactors led the 28% and 65% reduction of ozone emission at 8.7 s (corresponding plasma zone length of 60 cm) as compared with Glass/DBD reactor. The more ozone reduction in both packed reactors reflects the more oxygen active species react with hydrocarbons. It could support that the performance of $\text{Al}_2\text{O}_3/\text{DBD}$ and Pt-Pd/DBD reactors for styrene decomposition are better than that of Glass/DBD reactor.

The more ozone reduction in the Pt-Pd/DBD reactor suggests the "ozone catalytic oxidation" effect was observed in this study.^(20,21) This help to explain the significant reduction of CO and styrene removals as shown previously in Fig. 4 and Fig. 8(d). As the ozone was decomposed in the presence of Pt-Pd catalyst, more O active species were formed and they could oxidize more CO and hydrocarbons.

4. CONCLUSION

This study investigated the effects of glass, Al_2O_3 and Pt-Pd coated pellets packed inside DBD reactors on the decomposition of styrene and her byproducts. The results indicate that the decomposition efficiency increased as applied voltage increased as well as inlet styrene concentration decreased. The packed-bed DBD reactor can effectively enhance the composition of styrene as compared with that from nonpacked reactors. The utilization of glass and Al_2O_3 pellets can improve the decomposition efficiency and reduce the selectivity of gaseous and solid hydrocarbons, but Glass/DBD and $\text{Al}_2\text{O}_3/\text{DBD}$ reactors would not inhibit the formation of CO. The utilization of a plasma catalytic reactor with Pt-Pd coated pellets is the best method because they can drive the oxidation of carbon towards CO_2 rather than CO and therefore prevent the negative effect of the carbons deposited and gaseous byproducts on the packed-bed reactor.

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REFERENCES

1. C. H. Lin and H. Bai, *J. Environ. Eng.-ASCE* **127**, 648 (2001).
2. M. B. Chang and C. C. Chang, *AICHE J.* **43**, 1325 (1997).
3. H. M. Lee and M. B. Chang, *Plasma Chem. Plasma Process* **23**, 541 (2003).
4. U. Kogelschatz, *Plasma Chem. Plasma Process.* **23**, 1 (2003).
5. A. Ogata, K. Miyamae, K. Mizuno, S. Kushiyama, and M. Tezuka, *Plasma Chem. Plasma Process.* **22**, 537 (2002).
6. H. M. Lee and M. B. Chang, *Plasma Chem. Plasma Process* **21**, 329 (2001).
7. G. K. Anderson, H. Snyder, and J. Coogan, *Plasma Chem. Plasma Process* **19**, 131 (1999).
8. U. Roland, F. Holzer, and F. D. Kopinke, *Catal. Today* **73**, 315 (2002).
9. M. A. Malik, and X. Z. Jiang, *J. Environ. Sci. (China)* **10**, 276 (1998).
10. H. H. Kim, Y. H. Lee, A. Ogata, and S. Futamura, *Catal. Commun.* **4**, 347 (2003).
11. V. Demidiouk, S. I. Moon, and J. O. Chae, *Catal. Commun.* **4**, 51 (2003).
12. C. Ayrault, J. Barrault, N. Blin-Simiand, F. Jorand, S. Pasquiers, A. Rousseau, and J. M. Tatibouet, *Catal. Today* **89**, 75 (2004).
13. Y.-H. Song, S. J. Kim, K. I. Choi, and T. Yamamoto, *J. Electrostatics* **55**, 189 (2002).
14. A. Ogata, K. Mizuno, S. Kushiyama, and T. Yamamoto, *Plasma Chem. Plasma Process.* **18**, 363 (1998).
15. A. Ogata, D. Ito, K. Mizuno, S. Kushiyama, A. Gal, and T. Yamamoto, *Appl. Catal. A.* **236**, 9 (2002).
16. S. Futamura, A. Zang, G. Prieto, and T. Yamamoto, *IEEE Trans. Ind. Appl.* **34**, 967 (1998).
17. C. L. Chang and T. S. Lin, *Plasma Chem. Plasma Process.* **25(3)**, 227 (2005).
18. <http://webbook.nist.gov/chemistry/>. NIST Standard Reference Database Number 69. Visited on January 31, 2005.
19. R. M. Heck, R. J. Farrauto, and H. C. Lee: *Catal. Today* **13**, 43 (1992).
20. C. L. Chang and T. S. Lin, *Plasma Chem. Plasma Process.* **25(4)**, (2005).
21. P. Hunter and S. T. Oyama, *Control of Volatile Organic Compound Emissions: Conventional and Emerging Technologies*, John Wiley & Sons, New York, 2000, p. 224.