

DECOMPOSITION OF VOLATILE ORGANIC COMPOUNDS BY A MICROWAVE PLASMA DISCHARGE PROCESS AT ATMOSPHERIC PRESSURE¹**Young Sun Mok^{2*}, Moo Hyun Cho[†]**** Department of Chemical Engineering, Cheju National University Ara Jeju 690-756, Korea, Tel: 82-64-754-3682, Fax: 82-64-755-3670**† Department of Physics, Pohang University of Science and Technology Kyungbuk, 790-784, Korea*

Received 3 April 2003, in final form 13 June 2003, accepted 14 July 2003

The application of a microwave plasma process operated at atmospheric pressure to the decomposition of volatile organic compounds has been investigated. This microwave plasma system consists of a 2.45 GHz magnetron detached from a household microwave oven, a directional coupler, a three-stub tuner, a tapered waveguide terminated by a movable plunger, and plasma flame section where a quartz tube with a copper nozzle is located. In this system, the organic compounds such as toluene and trichloroethylene can be decomposed in the way of thermal incineration as well as by the reactions with a variety of active species formed during plasma discharge. The effect of feed gas flow rate on the decomposition was significant because it changed the gas temperature, but the initial concentration of the organic compound in the range of 210-2,100 ppm did not largely affect the decomposition efficiency. The principal byproduct was nitrogen oxides because this system was operated at high temperature. To improve the decomposition of the organic compounds, argon was used as a plasma-assisting gas, together with the air-like feed gas mixture. Large enhancement in the decomposition efficiency of the organic compounds was achieved by the use of argon.

PACS: 52.80.-s, 52.80.Pi

1 Introduction

Many industrial processes including chemical manufacturing, semiconductor and paint industries cause pollution via the emission of various volatile organic compounds (VOCs). Common methods available for VOCs treatment are adsorption process, catalytic oxidation and thermal incineration [1, 2], and lately many studies have reported that non-thermal plasma processes including dielectric barrier discharge and pulsed streamer corona discharge are effective methods for the decomposition of VOCs [3-7]. However,

¹Presented at XIVth Symposium on Application of Plasma Processes, Liptovský Mikuláš (Slovakia), January 2003

²E-mail address: smokie@cheju.ac.kr

incomplete oxidation of organic compounds is typical of non-thermal plasma processes even though oxygen is present in stoichiometric excess [8], i.e., carbon monoxide and various organic fragments with lower molecular weights such as methane, propylene and formaldehyde are produced as byproducts. Moreover, ozone formation in the plasma reactor has also been pointed out as a significant problem that should be resolved [7].

One of the other methods producing plasma at atmospheric pressure is to use microwave. Intense microwave field in waveguide accelerates electrons to give energetic ones that can ionize gas molecules, leading to plasma state [9]. Microwave plasma discharge can readily produce large amounts of a variety of active species such as radicals, ions and excited molecules. As a result, this process can induce effective chemical reactions [10]. At present, the study on the application of microwave plasma discharge to the chemical reactions is in rudimentary stage, and up to now only few studies have been performed for the reforming of natural gas to higher hydrocarbons, the removal of nitric oxide and the decomposition of perfluorocarbons (PFCs) produced in semiconductor industry [10-13].

The decomposition of VOCs using microwave plasma discharge may mitigate the problem related to the production of harmful byproducts of low molecular weights such as formaldehyde and carbon monoxide because the gas temperature can be increased high enough to oxidize VOCs completely. Therefore, the trouble of conventional non-thermal plasma processes can be settled without installing an oxidation catalytic reactor downstream the plasma reactor. In addition, ozone generation can essentially be repressed because the process is operated at high temperature. Magnetron that is the source of microwave is the main part of the microwave plasma process. It is easily available since it is mass-produced for household microwave oven. Such easy availability of magnetron can be a merit of this process from the practical application and economic viewpoints.

There may be several points at issue for further improvement of the microwave plasma process. For practical use, this process should be able to produce plasma at atmospheric pressure. In vacuum, the mean free path of electron is so long that generation and sustenance of plasma may be easy. On the contrary, generation and sustenance of plasma at atmospheric pressure are relatively difficult, which should be solved for easy use of this process. As well, selection and optimization of design parameters are necessary to maximize the conversion efficiency of the electrical energy into the generation of microwave plasma, resulting in the reduction of power consumption. Besides, it is needed to generally examine the effects of several key variables on the decomposition of organic compounds and the formation of byproducts.

The present study reports a method for producing and keeping up plasma at atmospheric pressure and some primitive results for the decomposition of organic compounds. The organic compounds chosen in this study were toluene and trichloroethylene. The present microwave plasma system was designed in order that plasma may be produced and sustained at atmospheric pressure. The performance of this process for the decomposition of organic compounds was assessed with the variables such as feed gas flow rate and initial concentration. The major byproducts anticipated in this process are CO_2 , CO and HCHO, and formation of nitrogen oxides is also anticipated because the microwave energy is consumed to heat up the feed gas stream. These byproducts were also analyzed. In air-like gas mixture, vibrational excitation and dissociation of N_2 molecules may consume a large amount of electron energy [14], resulting in lower decomposition

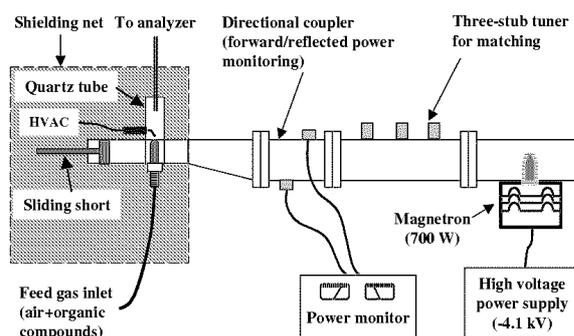


Fig. 1. Schematic of the 2.45 GHz microwave-induced plasma process.

efficiency of organic compound. To prevent such losses of electron energy, the possibility of the use of argon as a plasma-assisting gas was examined.

2 Experimental

The schematic diagram of the experimental setup used in this work is shown in Fig. 1. The apparatus consisted of a plasma reactor, a microwave power system, and a gas flow system. A 2.45 GHz magnetron detached from a household microwave oven (Model OM75S, Samsung Electronics, Korea) was used as the microwave source for the production of plasma. The magnetron used generates averagely 850 ± 50 W RF power. Microwave is transferred to the plasma reactor through the waveguide (W430). A three-stub tuner and a sliding short were installed to minimize the reflected power by tuning them properly. A quartz tube of 38 mm in diameter that passes through the upper surface of the wide waveguide is the plasma reactor. A 2/5" commercially available copper nozzle of the oxygen-fuel torch cutter was vertically inserted from the bottom wall of the wide waveguide. This nozzle has a center hole and several side holes. The feed gas was injected into both the center hole and the side holes of this nozzle. The quartz tube can help one take out the treated gas for analyses although it is in principle unnecessary to produce plasma. The plasma was initiated by generating a spark between the grounded nozzle tip and the needle igniter operated with AC high voltage (16.5 kV). Once the plasma was ignited, it was able to be stably sustained even when the igniter was turned off. Although the temperature of the plasma produced was high, the copper nozzle did not melt since it was continuously cooled down by the gas fed through it. The region around the quartz tube was surrounded by a stainless steel net for electromagnetic shielding.

Since the electric field around the nozzle should be high enough to produce plasma effectively, the waveguide was tapered to half in height, i.e., the height of the waveguide was reduced from 43.18 mm to 21.59 mm with the width kept constant (86.36 mm). The

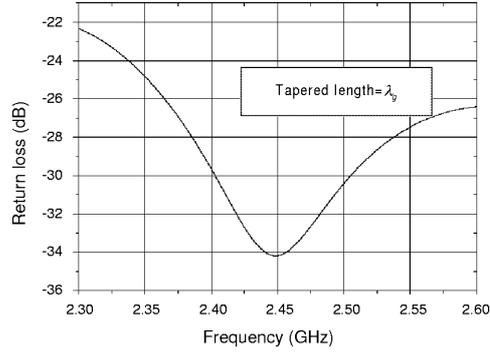


Fig. 2. Relation of the return loss to the frequency of microwave (one E-plane tapered).

optimal tapered length is related to the guide wavelength expressed as below [15]

$$\lambda_g = \frac{\lambda}{\sqrt{1 - (\lambda/2a)^2}}, \quad (1)$$

where λ_g is the guide wavelength, λ is the free space wavelength and a is the width of the waveguide. According to the simulation using HFSS computer code [16], the tapered length should be equal to the guide wavelength to minimize the return loss of RF power. Fig. 2 shows the relation of the return loss to the frequency in case that only one E-plane is tapered [16]. In Fig. 2, the return loss shows a minimum at a frequency of 2.45 GHz when the tapered length is equal to the guide wavelength. In case of 2.45 GHz, the free space wavelength λ is calculated to be 12.24 cm by a simple equation ($c = \lambda f$, c : speed of light; f : frequency). Using this value, the guide wavelength λ_g was found to be 17.3 cm.

The magnetron included in household microwave oven generally uses a half-wave voltage doubling circuit. Since the frequency of AC input is 60 Hz in the present case, the circuit generates 60 Hz high voltage pulse. Fig. 3 shows the waveform of the voltage applied to the cathode of the magnetron. The voltage is almost square wave with the pulse width of 8.3 ms, and its peak value is about -4 kV. In principle, this kind of low frequency pulse voltage makes the RF power intermittent, and installing an igniter is necessary to initiate the production of plasma.

One of the volatile organic compounds chosen in this study is toluene that is of the greatest importance in terms of the amount of emission. Toluene is classified as an aromatic compound, and it has been reported that the decomposition of toluene using non-thermal plasma is very difficult due to its chemical stability attributed to the resonance hybrid and requires a large amount of energy [17]. If the microwave plasma process can successfully treat such chemically stable compound, the other organic compounds are expected to be readily decomposed by this process. That is why toluene was chosen as a target compound in this study. The major component of the feed

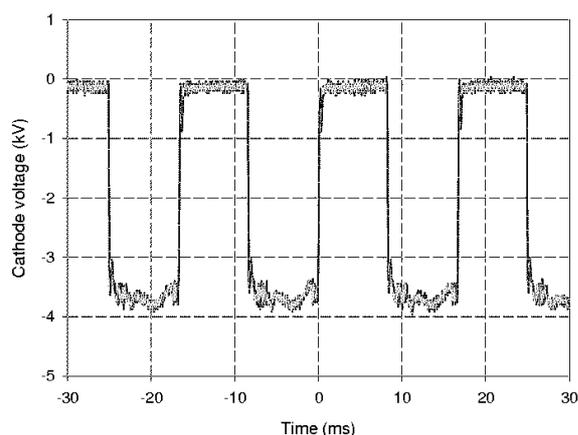


Fig. 3. Waveform of the cathode voltage applied to the magnetron.

gas stream was air (N_2 : 80%(v/v); O_2 : 20%(v/v)). The concentration of toluene was adjusted by using its vapor pressure. A mass flask containing toluene was immersed in a refrigerating circulator kept at a constant temperature of 20°C. At this temperature, the vapor pressure of toluene is 2.59 kPa. Nitrogen whose flow rate was regulated by a mass flow controller (Model 1179, MKS Instruments, Inc., USA) was saturated with toluene as it passed through the flask. When nitrogen was saturated at this temperature, the concentration of toluene corresponds to 25,570 ppm (parts per million, volumetric). This nitrogen saturated with toluene was later mixed with air, thereby diluted to a desired concentration. The concentration of toluene was changed from 210 to 2,100 ppm. The other organic compound chosen in this study is trichloroethylene, and its concentration was controlled to 1,000 ppm by the same method with that adopted for toluene. The flow rate of the feed gas stream was typically $1.67 \times 10^{-4} \text{ m}^3/\text{s}$, and it was varied in the range of 8.33×10^{-5} to $2.5 \times 10^{-4} \text{ m}^3/\text{s}$. So as to examine the effect of plasma-assisting gas on the decomposition of organic compounds, argon was used together with the air-like feed gas. The flow rates of argon and the feed gas were regulated by flow meters.

The net microwave power used for the production of plasma is the forward power minus the reflected power. The forward and reflected powers were measured by a directional coupler and a power monitor (MSTA Co., Korea). The AC input power was also measured by a digital power meter (WT200, Yokogawa, Japan). For the analysis of the organic compounds, a gas chromatograph equipped with a flame ionization detector (GC-14B, Shimadzu, Japan) was used. The decomposition efficiency of the organic compounds was obtained by measuring the difference in concentrations before and after plasma discharge. The byproducts such as CO and CO_2 were analyzed by a flue gas analyzer (GreenLine, Eurotron, USA) and a CO_2 monitor (Model 8762, TSI, Inc., USA), respectively, and for the measurement of formaldehyde, a chemical detector (Product number 91L, Gastec Co., Japan) was used. The concentration of NO_x formed in the reactor was analyzed by a chemiluminescence $\text{NO-NO}_2\text{-NO}_x$ analyzer (Model 42C, Thermo

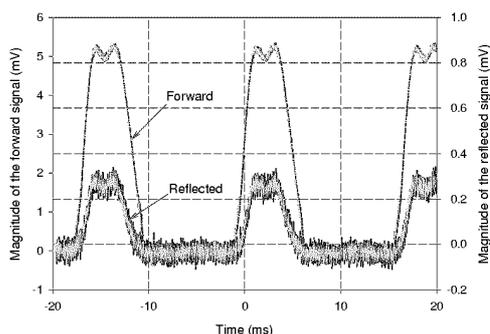


Fig. 4. Signals of the forward and reflected power converted into voltage.

Environmental Instruments, Inc., USA).

3 Results and discussion

The AC input power delivered to the magnetron system was measured to be 1.05 kW by a digital power meter. About 200 W was dissipated in the microwave generation circuit including cathode filament and high voltage transformer, and 850 ± 50 W was converted into RF power. Fig. 4 shows an example of the forward and reflected power when the offset of the short (distance from the center of the nozzle to the sliding short) was 22 mm. In this case, about 6% of the forward power was reflected, and thus 94% was used for the production of plasma. By adjusting the position of the sliding short, the reflected power can be minimized. The ratio of the reflected power (PR) to the forward power (PF) as a function of the offset of the short is shown in Fig. 5. When the offset of the short was longer than 30 mm, the reflection ratio came to about 50%. However, the reflection ratio was minimized as the distance from the nozzle center to the short decreased to around 20 mm. Further experiments were carried out at this offset position of the short.

Fig. 6 presents a photograph of the microwave plasma discharge when the feed gas flow rate was 1.67×10^{-4} m³/s. The initiation of the plasma was done by generating a spark between the nozzle tip and the needle igniter. The plasma flame started from the nozzle tip and propagated upwards. Once the plasma was ignited, it was able to be stably sustained even when the igniter was turned off. Due to the high collision rate with surrounding gas molecules, the plasma flame appeared to be the same as the flame of the oxygen-fuel torch. The length and width of the plasma flame are related to the RF power dissipated for the production of plasma. In this experimental condition, the plasma flame was extended up to 4.7 cm at 20 mm offset of the short. As can be seen, the width of the plasma flame was very narrow, implying that the feed gas flow should be concentrated into the flame center for efficient treatment.

In case of non-thermal plasma processes, the principle reactions for toluene decomposition are known to be the charge transfer reactions of toluene with ions including N_2^+ , O_2^+ , N^+ and O^+ [4, 18]. The charge transfer reactions and the relevant rate constants

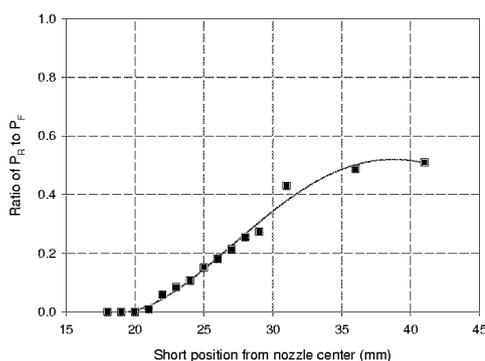


Fig. 5. Ratio of the reflected power to the forward power as a function of short position from the nozzle center (nozzle diameter: 10.2 mm; no air flow).

are summarized in the literature [18]. As well, the energetic electron impact dissociations may provide additional decomposition channels [4]. Besides such charge transfer reactions and energetic electron impact dissociations, thermal decomposition is believed to play an important role in the microwave plasma process because of high gas temperature. The effect of feed gas flow rate on the decomposition efficiency is depicted in Fig. 7. The decomposition efficiency was lowered much when the flow rate increased. The decrease in the decomposition efficiency with the increase in the flow rate may partly be explained by the decreased gas temperature. The adiabatic temperature increase can be calculated by using a simple equation as follows:

$$q = m \int_{T_1}^{T_2} C_p dT, \quad C_p = \alpha + \beta T + \gamma T^2 \text{ (cal/mol K)}. \quad (2)$$

Here, T_1 is the temperature of the feed gas entering the nozzle, i.e., 25°C, and T_2 is the final temperature. In Eq. (2), m is the molar flow rate of the feed gas and equal to the product of the molar gas density (40.9 mol/m³ at 25°C) and the volumetric flow rate. In case of air, the values of α , β and γ are 6.90, 9.2×10^{-4} and -1.76×10^{-4} , respectively. So as to find out T_2 using this equation, energy delivery rate (power) should be known. In the absence of feed gas flow, the reflection ratio was reduced to 0.03 at the offset position of 20 mm (see Fig. 5). When the feed gas was injected to the nozzle, however, the reflection ratio considerably increased. As observed in Fig. 8, the reflection ratio was about 0.2 at flow rates in the range of 8.33×10^{-5} to 2.5×10^{-4} m³/s. Although it was not largely dependent on the flow rate, the reflection ratio in the presence of gas flow was relatively higher, compared to that in the absence of it. Taking this reflection loss into account, the net microwave power used for the production of plasma is calculated to be 700 W (167 cal/s). With this energy delivery rate (power), Eq. (2) can be solved in terms of T_2 . When the gas flow rate at 25°C was 1.67×10^{-4} m³/s, T_2 was found to be 2,900°C. Similarly, the respective adiabatic temperatures at flow rates of 8.33×10^{-5} and 2.5×10^{-4} m³/s were found to be 5,170 and 2,050°C. Since the reactor system

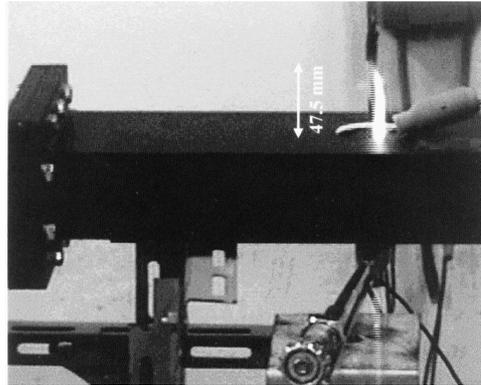


Fig. 6. Photograph of the plasma flame (air flow rate: $1.67 \times 10^{-4} \text{ m}^3/\text{s}$).

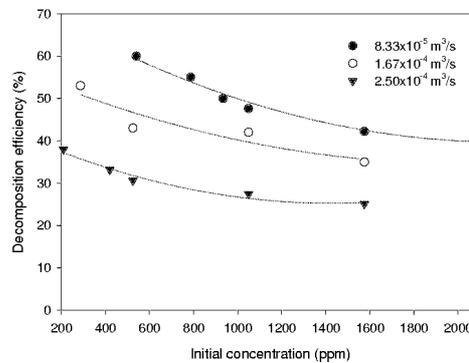


Fig. 7. Effect of the feed gas flow rate on the decomposition efficiency (background: air).

was not isolated from natural cooling, the real gas temperature would be much lower than the adiabatic temperature. But, it is obvious that the increase in the flow rate should cause the decrease in the decomposition efficiency. In addition, the decrease in the decomposition efficiency with the increase in the flow rate can also be explained with the decreases in relative amounts of active species such as N_2^+ , O_2^+ , N^+ and O^+ available for the decomposition. The number of active species is a function of power dissipated for the production of plasma. Thus, the concentration of the active species decreases with the increase in the flow rate when the power is identical.

One general trend appearing in Fig. 7 is that the decomposition efficiency did not largely depend on the initial concentration. For example, when the initial concentration was 250 ppm, about 55% of toluene was removed at a flow rate of $1.67 \times 10^{-4} \text{ m}^3/\text{s}$. When the initial value was increased to 1,600 ppm at the same flow rate, the decomposition efficiency was about 40%. This decomposition efficiency is rather high, compared

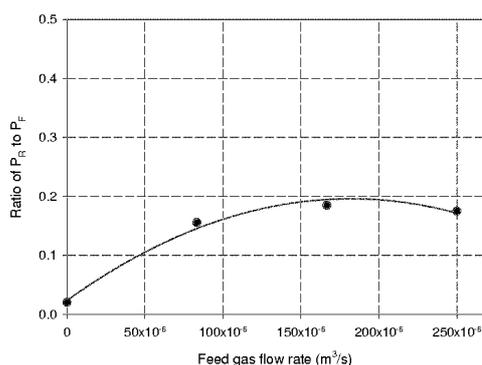


Fig. 8. Ratio of the reflected power to the forward power as a function of feed gas flow rate (offset of the short: 20 mm).

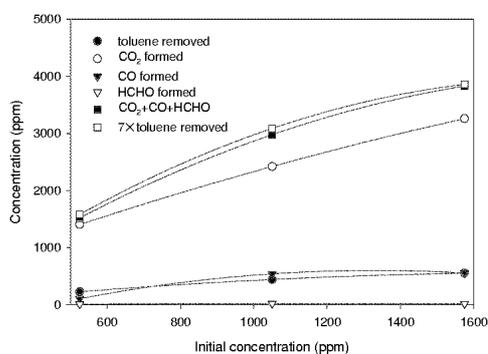
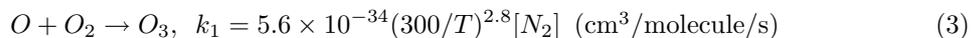


Fig. 9. Byproducts formation in the plasma reactor (feed gas: $1.67 \times 10^{-4} \text{ m}^3/\text{s}$).

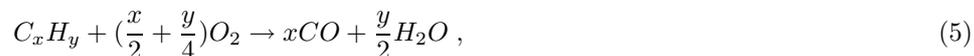
to the high initial concentration. In thermal incineration process, the rate of decomposition is well described by first order reaction [2]. First order reaction means that the decomposition efficiency or the ratio of final to initial concentration is not a function of initial concentration. The weak dependency of the decomposition efficiency on the initial concentration implies that the thermal reaction takes an important part in the decomposition of toluene.

Fig. 9 presents the evolution of byproducts in the plasma reactor. Referring to the previous results, common non-thermal plasma processes utilizing high voltage do not completely decompose VOCs into CO_2 , and generate CO and various organic fragments with lower molecular weights as well as ozone [3, 4, 7, 19, 20]. On the other hand, no other hydrocarbon compounds besides toluene and formaldehyde and no ozone were detected in this process. One molecule of toluene can form seven carbon-containing molecules. The black square symbol in Fig. 9 stands for the summation of CO, CO_2 and HCHO

concentrations, and the white square symbol indicates seven times the amount of toluene decomposed. As observed, the two results were balanced quite well. In this process, the reason that ozone was not formed can be explained by the following equations [21]:



According to Eqs. (3) and (4), the formation of ozone is inversely proportion to the temperature and the temperature dependence of the destruction of ozone is rather strong. Therefore, it is natural that the formation of ozone should not occur in this process operated at high temperature. As presented in Fig. 9, negligible amount of formaldehyde was observed, regardless of the initial toluene concentration. This result may also be interpreted by the high gas temperature. In fact, formaldehyde can be generated as an intermediate compound during the decomposition process, but it is oxidized to carbon oxides fast at high temperature. The other carbon-containing byproducts observed were carbon monoxide and carbon dioxide. In case of thermal incineration, the following two-stage reaction scheme were suggested to express the decomposition of organic compounds although the real mechanism in detail is more complicated [2]:



Equations (5) and (6) indicate that organic compound is first converted into CO, and then CO is further oxidized to CO₂. In case of non-thermal plasma process, however, it has been reported that the generation of CO goes via different pathways from that of CO₂ [3, 22]. As mentioned above, this microwave plasma system can decompose organic compound by both thermal and ionic reactions. Whatever the generation pathways are, CO can be oxidized to CO₂ at high temperature in the presence of oxygen. This can account for the reason that the resulting concentration of CO₂ was much higher than that of CO. In this process, the ratio of CO to CO₂ was very small in the range of 0.06 to 0.19. On the contrary, in typical non-thermal plasma reactor, this ratio is about 0.6 at 20%(v/v) oxygen content [19].

This microwave plasma process is operated at high temperature, so that the formation of nitrogen oxides (NO_x) is possible. Fig. 10 represents the concentration of NO_x generated in this process. As can be seen, about 10,000 ppm of NO_x (NO+NO₂) was generated at a flow rate of $8.33 \times 10^{-5} \text{ m}^3/\text{s}$, and it decreased to 1,500~2,000 ppm as the flow rate increased to $1.67 \times 10^{-4} \sim 3.33 \times 10^{-4} \text{ m}^3/\text{s}$. The high concentration of NO_x formed is easily understood by the high gas temperature. Therefore, this process can be valuable only when it is used to treat more harmful compounds such as perfluorocarbons than nitrogen oxides, although this study investigated the decomposition of common volatile organic compound.

As shown above, the decomposition efficiency was not so high when air was used as the background gas. Nitrogen in air shifts the electron energy density distribution to

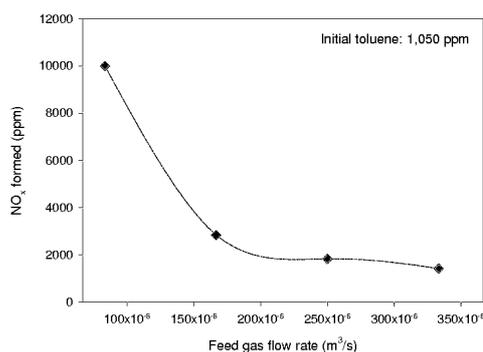


Fig. 10. Formation of nitrogen oxides as a function of feed gas flow rate.

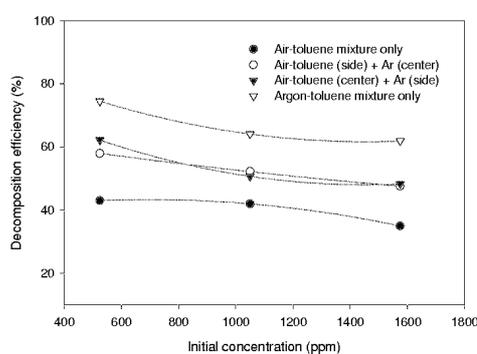


Fig. 11. Effect of argon on the decomposition of toluene (total gas flow rate: $1.67 \times 10^{-4} \text{ m}^3/\text{s}$).

less favorable conditions for the formation of active species [8]. In nitrogen mixtures, a large fraction of electron energy is lost due to vibrational excitation and dissociation of N_2 molecules [14], giving less ionic species to participate in the decomposition. To improve the decomposition efficiency, argon was used to assist plasma generation. When argon is used as the background gas, such losses of electron energy as mentioned above are lowered, and thus it is suitable for the purpose of assisting the generation of intense plasma. First, the experiment was conducted by using argon only as the background gas. After that, both air and argon were used as the background gas. One experiment was performed by injecting $8.33 \times 10^{-5} \text{ m}^3/\text{s}$ of argon into the side holes of the nozzle while $8.33 \times 10^{-5} \text{ m}^3/\text{s}$ of air containing toluene was fed to the center hole of the nozzle. The other experiment was carried out by injecting the gases reversely. Fig. 11 presents the effect of argon on the decomposition efficiency of toluene. The initial concentration in the horizontal axis is based on the total flow rate (argon+air). As expected, the decomposition efficiency was largely enhanced by the use of argon. When argon is used

as the background gas, a part of toluene can be decomposed by the energetic electron impact dissociations:



where e denotes the energetic electron and the rate constant is of the order of 10^{-6} $\text{cm}^3/\text{molecules/s}$ [4]. The additional probable decomposition channels may be the excitation transfer reactions with the excited argon molecules and the charge transfer reactions with argon ions:



where Ar^* and Ar^+ stand for the excited molecule and ion, respectively. The energy yield (energy consumption/toluene molecule decomposed) is dependent on the initial concentration and the microwave power. When the initial concentration was 1,575 ppm and the microwave power was 700 W, for instance, 62 % decomposition efficiency was obtained with argon-toluene mixture. This decomposition efficiency is tantamount to an energy yield of 1,100 eV per toluene molecule decomposed. Meanwhile, Krasnoperov *et al.* [23] have reported that conventional non-thermal plasma process using dielectric barrier discharge requires electrical energy in the range of 505-1980 eV to decompose one molecule of toluene. When argon was injected into the side holes of the nozzle, almost similar decomposition efficiency to the case that argon was injected to the center hole of the nozzle was obtained. In case of trichloroethylene (1,000 ppm), about 80 % decomposition efficiency was achieved by feeding 8.33×10^{-5} m^3/s of argon to the side nozzles with the total flow rate kept 1.67×10^{-4} m^3/s (see Fig. 12). This decomposition efficiency is much higher than 53 % obtained by using air alone as the background gas. Rosocha *et al.* [24] presents the rate constant data for the electron impact dissociation of molecular oxygen to form atomic oxygen for the argon and air combination. The rate of atomic oxygen produced in the air-like mixture is approximately an order of magnitude less than the rate of atomic oxygen produced in the argon mixture. Since atomic oxygen is one of the active species utilized for the decomposition of trichloroethylene [24], the decomposition efficiency in air alone differs from that in air-argon mixture. Argon can also affect the rate of generation of other active species related to the decomposition of trichloroethylene in a similar way. Collectively, the presence of argon enhances the decomposition efficiency of organic compounds. Argon in the treated gas stream can be separated and recycled upstream for repeated use.

Fig. 13 presents the decomposition results of trichloroethylene and toluene conducted in a dielectric barrier discharge reactor. The details of this reactor structure are given in the previous paper [17]. In this system, trichloroethylene and toluene were almost completely decomposed at 140 W and 250 W of input power, respectively. On the other hand, the present microwave plasma system gave much lower decomposition efficiencies despite much higher input power, as observed in Figs. 11 and 12. The decomposition of trichloroethylene in a dielectric barrier discharge reactor largely decreases with the increase in the reaction temperature [25]. In case of the microwave plasma system, the

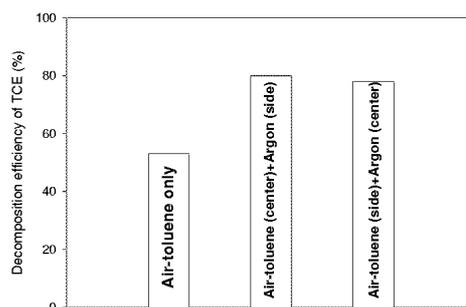


Fig. 12. Comparison between the decomposition efficiencies of trichloroethylene obtained in the presence and in the absence of argon.

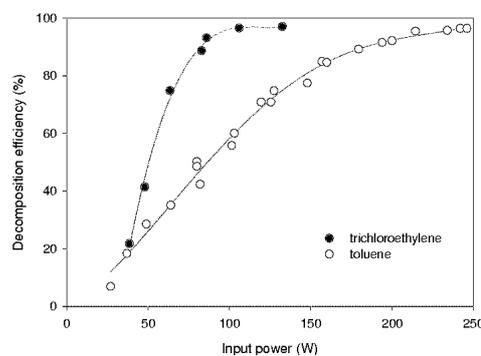


Fig. 13. Decomposition efficiencies of toluene and trichloroethylene as a function of input power in a dielectric barrier discharge reactor (background: air; initial trichloroethylene: 660 ppm; initial toluene: 300 ppm; temperature: room temperature).

temperature of the feed gas rapidly rises up to a few thousand degrees as it comes out from the nozzle, which can lower the decomposition rate. In addition, the vibrational excitation and dissociation of nitrogen molecules dissipate a considerable amount of the microwave energy whereas a dielectric barrier discharge process consumes a large fraction of the electrical energy for the production of reactive species available for the decomposition of the organic compounds. So as to decompose organic compounds effectively in the microwave plasma system, the feed gas flow should be concentrated into the narrow plasma flame. In the present microwave plasma system, the plasma flame quivered a little and frequently leaned, probably because of the presence of the spark igniter. This may also have caused the decrease in the decomposition rate. For the practical application, therefore, the microwave plasma system should be able to produce and sustain plasma without the spark igniter. One method to produce plasma without using spark igniter is to apply dc high voltage to the cathode of magnetron instead of half-wave rectified volt-

age [16], which make it possible to obtain continuous microwave power. The application of the continuous microwave power to the decomposition of organic compounds is left as a topic for further study.

4 Concluding remarks

A microwave-sustained atmospheric pressure plasma system based on waveguide was used to decompose volatile organic compounds. A magnetron detached from a household microwave oven was used as the microwave source. In case of the nozzle tip diameter of 10.2 mm and the hole diameter of 38.0 mm, the reflection of microwave was minimized when the position of the nozzle center was 20 mm apart from the short. The decomposition of toluene largely decreased with the increase in the feed gas flow rate due to the decreased gas temperature and the decreased concentrations of active species available while it was not a strong function of the initial concentration. In the application of this plasma system to the decomposition of organic compounds, argon may be used as a gas assisting plasma generation. When argon and the feed gas were injected to the side holes and the center hole of the nozzle and vice versa, large enhancement in the decomposition of organic compounds such as toluene and trichloroethylene was observed. The principal byproduct was nitrogen oxides because this system was operated at high temperature, but the operation at high temperature resulted in much higher CO₂/CO ratio, compared to the non-thermal plasma process using high voltage.

Acknowledgment: This work was supported by Korea Research Foundation Grant (KRF-2002-003-D00085).

References

- [1] K. Yan, H. Hui, M. Cui, J. Miao, X. Wu, C. Bao, R. Li: *J. Electrostatics* **44** (1998) 17
- [2] C. D. Cooper, F. C. Alley: *Air Pollution Control: A Design Approach*, Waveland Press, Inc. (1994)
- [3] S. Futamura, A. Zhang, T. Yamamoto: *IEEE Trans. Ind. Appl.* **35** (1999) 760
- [4] H. Kohno, A. A. Berezin, J. S. Chang, M. Tamura, T. Yamamoto, A. Shibuya, S. Honda: *IEEE Trans. Ind. Appl.* **34** (1998) 953
- [5] T. Oda, R. Yamashita, I. Haga, T. Takahashi, S. Masuda: *IEEE Trans. Ind. Appl.* **32** (1996) 118
- [6] Y. S. Choi, Y. H. Song, S. J. Kim, B. U. Kim: *HWAHAK KONGHAK* **38** (2000) 423
- [7] S. Moon, J. Chae: *J. Korean Soc. Environ. Eng.* **23** (2001) 243
- [8] H. R. Snyder, G. K. Anderson: *IEEE Trans. Plasma Sci.* **26** (1998) 1695
- [9] H. S. Uhm: Contamination Control of Emission Discharge, US Patent 5,830,328 (1998)
- [10] M. Baeva, H. Gier, A. Pott, J. Uhlenbusch: *Plasma Chem. Plasma Proc.* **21** (2001) 225
- [11] W. Cho, Y. Baek, H. Pang, Y. C. Kim, S. Moon: *J. Korean Ind. Eng. Chem.* **9** (1998) 94
- [12] M. A. Wojtowicz, F. P. Miknis, R. W. Grimes, W. W. Smith, M. A. Serio: *J. Hazardous Materials* **74** (2000) 81
- [13] J. von Hagen, Y. Venot, Y. Zhang, W. Wiesbeck: *IEEE Trans. Plasma Sci.* **29** (2001) 604
- [14] H. Kucukarpaci, J. Lucas: *J. Phys. D* **12** (1979) 2123

- [15] H. K. Park: *Microwave Engineering*, Hee Jung Dang Press, Korea 1991
- [16] Y. S. Bae, E. M. Choi, W. S. Chung, Y. H. Lee, W. Namkung, M. H. Cho: *Joint Int. Plasma Symp. 6th APCPST, 15th SPSM, OS2002 and 11th KAPRA*, Jeju, Korea, 2002
- [17] Y. S. Mok, C. M. Nam, M. H. Cho, I. Nam: *IEEE Trans. Plasma Sci.* **30** (2002) 408
- [18] L. W. Sieck, T. J. Buckley, J. T. Herron, D. S. Green: *Plasma Chem. Plasma Proc.* **21** (2001) 441
- [19] A. Ogata, N. Shintani, A. Mizuno, S. Kushiya, T. Yamamoto: *IEEE Trans. Ind. Appl.* **35** (1999) 753
- [20] Y. S. Mok, J. H. Kim, S. W. Ham, I. Nam: *Ind. Eng. Chem., Res.* **39** (2000) 3938
- [21] R. Atkinson, D. L. Baulch, R. A. Cox, R. F. Hampson Jr., J. A. Kerr, J. Troe: *J. Phys. Chem. Ref. Data* **21** (1992)
- [22] Y. Song, D. Shin, W. Shin, K. Kim, Y. Choi, Y. S. Choi, W. Lee, S. Kim: *J. Korean Soc. Atmospheric Environ.* **16** (2000) 247
- [23] L. N. Krasnoperov, L. G. Krishtopa, J. W. Bozzelli: *J. Adv. Oxid. Technol.* **2** (1997) 248
- [24] L. A. Rosocha, G. K. Anderson, L. A. Bechtold, J. J. Coogan, H. G. Heck, M. Kang, W. H. McCulla, R. A. Tennant, P. J. Wantuck: in *Non-Thermal Plasma Techniques for Pollution Control: Part B* (Eds. B. M. Penetrante, S. E. Schultheis). Springer-Verlag, Berlin, Germany (1993) 281
- [25] B. M. Penetrante, M. C. Hsiao, B. T. Merritt, G. E. Vogtlin, P. H. Wallman: *Workshop on the Treatment of Gaseous Emissions via Plasma Technology*, Gaithersburg, Maryland, 1995