We are IntechOpen, the world's leading publisher of Open Access books Built by scientists, for scientists

6,900
Open access books available

185,000
International authors and editors

200M
Downloads

Our authors are among the

154
Countries delivered to

TOP 1%
most cited scientists

12.2%
Contributors from top 500 universities

WEB OF SCIENCE™

Selection of our books indexed in the Book Citation Index in Web of Science™ Core Collection (BKCI)

Interested in publishing with us?
Contact book.department@intechopen.com

Numbers displayed above are based on latest data collected.
For more information visit www.intechopen.com

Chapter 9

Non-Thermal Plasma Technic for Air Pollution Control

Takao Matsumoto, Douyan Wang, Takao Namihira and Hidenori Akiyama

Additional information is available at the end of the chapter

http://dx.doi.org/10.5772/50419

1. Introduction

The air pollutions from combustion flue gas and industrial gases became worse and cause the environmental problem. It is difficult for the conventional methods such as selective catalytic reduction method and lime-gypsum method to treat exhaust gases energy efficiently and inexpensively. Its energy efficiency and its initial and running costs are still in negative situation for the backward nations. In recent years, the pollution control techniques using non-thermal plasmas have been widely studied because it is one of the promising technologies for pollution control with higher energy efficiency [1]-[7]. The non-thermal plasma could treat multiple toxic molecules simultaneously, and it can be applied to locations where the conventional catalyst methods are difficult to use. In this chapter, a principle of air pollution control by non-thermal plasma, various methods of non-thermal plasma formation and those current situations are introduced.

2. Non-thermal plasma

Plasma, also referred to as "ionized gas" is mixed state of atoms, molecules, ions, electrons and radicals. Plasma has two general states: equilibrium and non-equilibrium. The equilibrium state indicates the temperatures of electrons, ions and neutrals become almost equal, and the background gas is heated from a few thousands to more than ten thousands Kelvin degrees. Because of this, the plasma getting equilibrium state is called as "thermal plasma". On the other hand, the non-equilibrium state means that the temperatures of electrons, ions and neutrals are quite different, and in general the electron temperature is substantially higher than other particles. Therefore, the rise of background gas temperature is quite low in non-equilibrium state and the plasma being non-equilibrium state is called as "non-thermal plasma". Figure 1 shows a typical example of non-thermal plasma [8]. This figure shows the background gas temperature of non-thermal plasma is enough low to

INTECH
open science | open minds

© 2012 Matsumoto et al., licensee InTech. This is an open access chapter distributed under the terms of the Creative Commons Attribution License (http://creativecommons.org/licenses/by/3.0), which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

touch by a finger. In the non-thermal plasma, the majority of the discharge energy goes into the production of energetic electrons, rather than ion and neutron heating. The energy in the plasma is thus consumed preferentially to the electron impact dissociation and ionization of the background gas for production of radicals that, in turn, decompose the toxic molecules. In short, non-thermal plasma can remove toxic molecules near room temperature without consuming a lot of energy in background gas heating.

For low pressure plasma process such as semiconductor production, the non-equilibrium plasma which is often named "cold plasma" is typically used. Prof. Oda [9] defined that non-thermal plasma is high pressure (typically 1 atmospheric pressure) non-equilibrium plasma. Compared with that cold plasma, the electron temperature and ionization rate are quite lower in non-thermal plasma. Typically, the electron temperature of cold plasma is tens of eV. Meanwhile, in atmospheric pressure, the electron temperature is generally 1 to 10eV and ionization rate is around 0.1%. However, it is important for gas processing in atmospheric pressure because electron and molecular density is overwhelmingly high in comparison to low pressure condition. If the gas processing is done in low pressure condition, the absolute molecule quantity is low. That is, large amount of energetic electron having more than dissociation energy of objective molecules are need in order to generate more radicals and decompose more toxic molecules. Later on, the required value of electron energy for air pollution control is approximately 10eV.

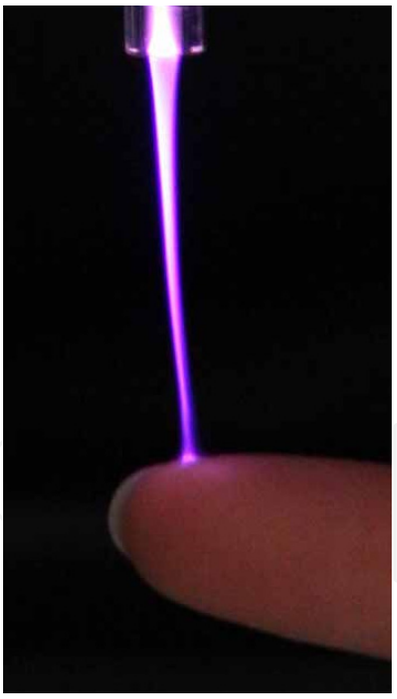


Figure 1. Typical example of non-thermal plasma demonstration [8].

3. Formation methods of non-thermal plasma

Non-thermal plasmas for removal of hazardous pollutants have been produced by an electron beam method and various electrical discharge methods.

3.1. Electron beam

The electron beam irradiation is one of non-thermal plasma formation method. Figure 2 shows schematic representation of electron beam source. In an electron beam method, the electrons are accelerated by high voltage in the vacuum region before being injected into a gas-processing chamber thorough a thin foil window. The energy of electron beam is directly used for dissociation and ionization the background gas. During the ionization by the beam, a shower of ionization electrons is generated, which further produce a large volume of plasma that can be used to initiate the removal of various types of pollutant molecules such as NOx, SOx and VOCs. This exhaust gas treatment technic by an electron beam has a 40 year-old history previously and a lot of pilot plants for air pollution control have been running today [10]-[18].

In particular, an Electron Beam Dry Scrubbing (EBDS) system has been mainly studied at present. Figure 3 shows that the typical principle of EBDS. It is a dry process and does not require an expensive catalyst for NOx removal. In this process, at first, many oxidative radicals such as O, OH and HO_2 were produced by electron beam irradiation into O_2 and H_2O in exhaust gas. Following that, NOx and SOx are oxidized by these radicals to HNO_3 and H_2SO_4. Finally, HNO_3 and H_2SO_4 were converted to ammonium nitrate (NH_4NO_3) and ammonium sulfate $(NH_4)_2SO_4$ by added ammonia (NH_3) into the treated combustion flue gas. These byproducts are collected by the electrostatic precipitator (ESP) and shipped to outside, because NH_4NO_3 and $(NH_4)_2SO_4$ can be used to make fertilizer. Here it should be noted that a part of NO is reduced to N_2 by N radical which produced by electron beam irradiation. This EBDS system has applicability to a high concentration sulfur-containing coal-fired boiler and a treatment of solid waste. In either case, EBDS could treat NOx and SO_2 in high efficiency. According to the literatures [19], over 95% of SO_2 and over 80% of NOx were removed simultaneously when the flue gas of sulfur-containing (at least 2.5%) coal-fired boiler was used as simulate gas.

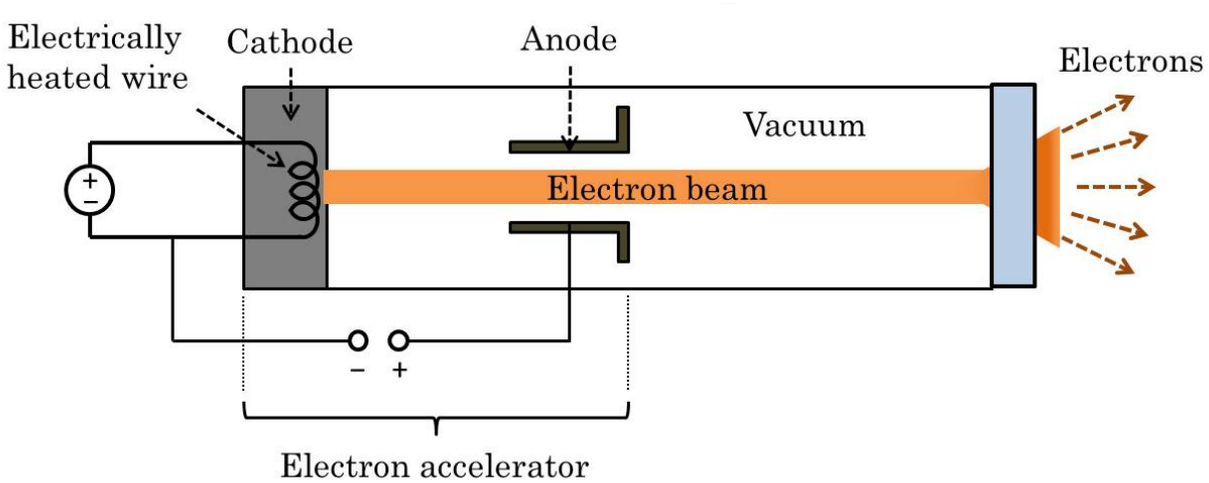


Figure 2. Schematic representation of electron beam source.

The potential of using an electron beam for removal of post combustion toxic gases (NOx, SO_2) was recognized in the early 1970s by the Ebara Corporation (Japan) [20]. Following successful initial batch tests of the Ebara plant, various tests on small pilot plants have been conducted in the Canada [16], Korea [21], Poland [22], and Japan [23], etc. The tests performed in these installations proved that a significant amount of NOx (and SO_2) exhausted from power plants, municipal-waste incinerators, and combustion boilers, etc., could be efficiency removed. In addition, A.G. Ignat'ev [24], B.M. Penetrante [25] and Y. Nakagawa [26] have indicated that using a pulsed electron beam improves the energy efficiency for exhaust gas treatment instead of using a DC electron beam. However, the electron beam methods hasn't put into practical use yet due to the high capital cost of accelerators, X-ray hazard and the unavoidable large energy loss caused by vacuum interface.

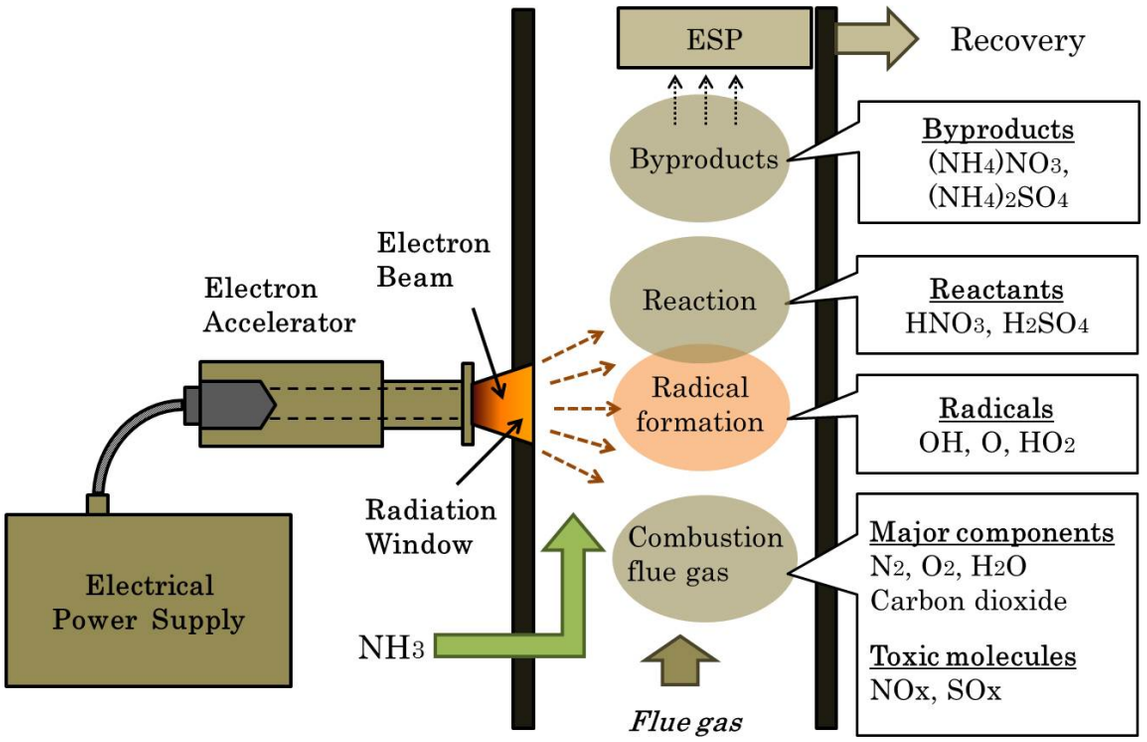


Figure 3. Principle of combustion flue gas treatment by electron beam.

3.2. Electrical discharge

In contrast to the electron beam which produce non-thermal plasma by supplying energetic electrons to objective gas, electrical discharge methods which led objective gas into plasma directly and generate energetic electron and radicals. Electrical discharges could produce non-thermal plasma in atmospheric pressure gases by various power supply such as direct current (DC), alternating current (AC), or pulse power sources. Among them, the dielectric barrier discharge (DBD) method using AC high voltage source and pulsed power discharge method have been developed particularly to this day. In this section, DBD and pulse power discharge are introduced.

3.2.1. Dielectric Barrier Discharge

A schematic representation of typical DBD electrodes is shown in figure 4. The DBD is also called as a silent discharge. In DBD reactor, AC high voltage which is typically 10 to 20 kV and 50 Hz to 2 kHz are applied to electrodes, one or both of which are covered with a thin dielectric layer, such as glass. The gap distance between electrodes is a few hundred of μm to several mm order. The barrier discharge is characterized by millions of small pulsed micro discharge which occur repetitively in gas space. The current density of the micro discharge is approximately 1 kA/cm^2, the diameter is 0.1 mm and the pulse duration is 3ns. Because of energetic electrons are generated in this micro discharge, various radicals and ions are produced by the electron collision with gas molecules. These radicals defuse into the barrier discharge space and react with background gas. As a result, ozone generation and NOx or VOCs removal are realized.

Dielectric barrier discharge processing is very mature technology, first investigated in 1850's for the production of ozone. Ozone has some effects such as sterilization, deodorization, and decolorization, because of its strong oxidization power, placing it second after fluorine. Furthermore, ozone has no residual toxicity due to its spontaneous decomposition feature. Therefore, ozone has already been put to practical use in water purification instead of conventional sterilization by chlorine. Ozone has been used in Europe for water treatment since early in the 20th century. Initial applications were to disinfect relatively clean spring or well water, but they increasingly evolved to also oxidize contaminants common to surface waters. Since 1950's, ozonation has become the primary method to assure clean water in Switzerland, West Germany and France. More recently, major fresh water and waste water treatment facilities using ozone water treatment methods have been constructed throughout the world. Additionally, new industrial applications of ozone such as wastewater treatment, exhaust gas treatment, odor elimination and semiconductor manufacturing have been studied recently [27-35]. However these new ozone applications demand high concentration of ozone, it was become possible for DBD to produce high concentration (100 to 300 g/m^3) of ozone due to the improvement of dielectric materials and electrodes cooling function, and development of ultra-short gap electrodes [27]-[35]. Figure 5 shows a schematic representation of a cylindrical reactor which is modern shape of today's ozonizer. Consequently, the dielectric barrier method is most common way of ozone generation today. In addition, DBD has been studied for flue gas cleaning and toxic gas decomposition. In the literatures, removal of various toxic molecules such as NOx in diesel engine exhaust, greenhouse gas and VOCs such as formaldehyde which causes a sick building syndrome, have been demonstrated.

However, ozone generation by a dielectric barrier discharge is common way at today, it has still some agendas for industrial applications. For example, need of external cooling system for discharge electrodes and its sensitivity narrow gap separation take plenty operation costs. In addition, the narrow gap is sensitive to grit, dust and vibration. Therefore, use of this pollution control by ozone process is limited to a part of well-financed company or state and public institutions. The improvement of energy efficiency for DBD system is strongly demanded in order to spread the ozone processing moreover. In addition, NOx removal and

VOCs treatment using DBD is still in laboratory stage, because DBD could not treat those toxic molecules completely and its energy efficiency is unfavorable at the present stage.

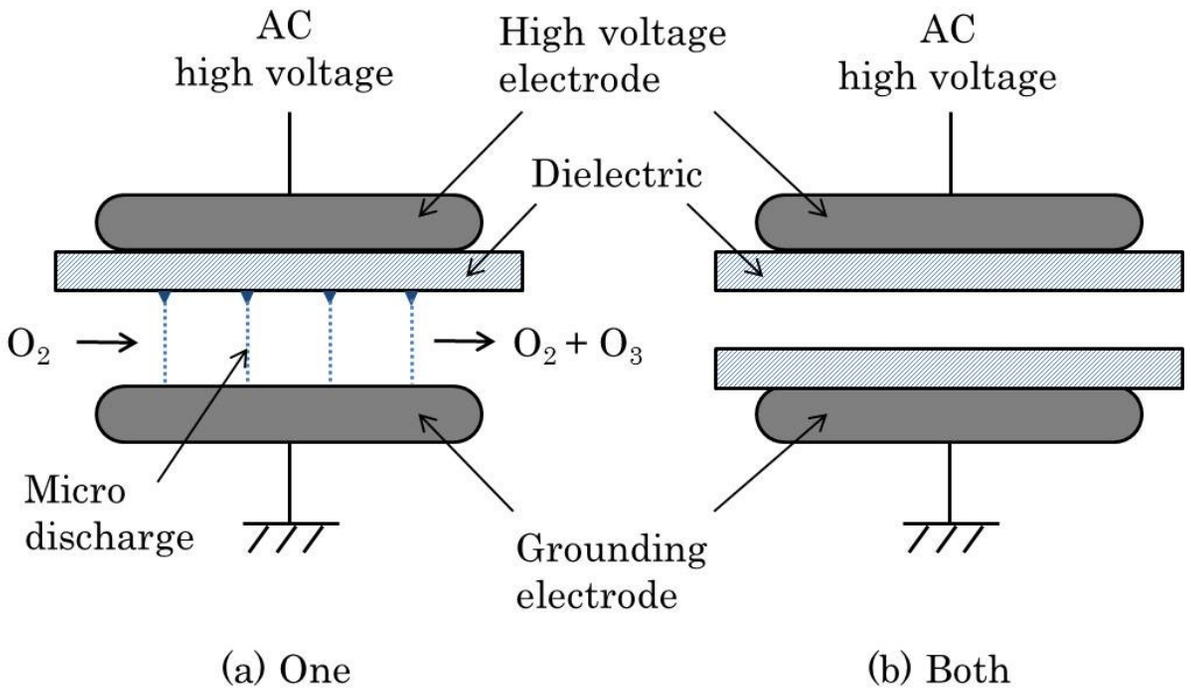


Figure 4. Schematic representation of dielectric barrier discharge electrode.

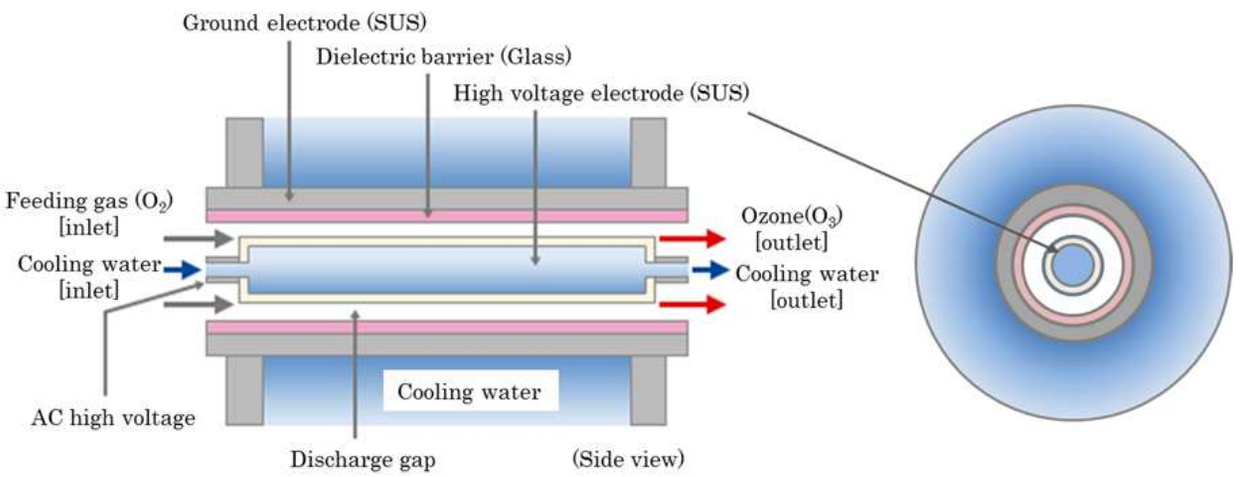


Figure 5. Schematic representation of cylindrical cooled reactor.

3.2.2. Pulse power discharge

Pulsed power discharges have been studied for many years since it is one of the promising technologies for the removal of the hazardous environmental pollutants as well as electron beam and dielectric barrier discharge. Typically, many researchers have reported that the

pulsed discharge performed DeNOx process more effectively comparison with direct current (DC) corona discharge [3], [6, 7], [36]-[39]. Because the pulsed discharge is generated by intermittent pulse voltage, it is difficult to transfer to an arc discharge. Therefore, pulsed discharge is possible to apply an overvoltage between electrodes. Moreover, it is noted that the rate of over voltage (applied voltage / DC breakdown voltage) is depend on the voltage rise time and the pulse duration. In this way, since the pulse discharge could apply an overvoltage, it is possible to generate large amount of energetic electrons which have over 10 eV in atmospheric plasma [7]. Generated high-energy electrons could easily dissociate a nitrogen molecule (N_2) which having 9.8 eV of comparatively-high dissociation energy in gas. Therefore, generation of large amount of radicals that contributes to the gas processing become possible, and effectiveness of pulsed discharge could be obtained.

Pulsed power is a technology that concentrates electrical energy and turns it into short pulses of enormous power. Pulsed power technology had been studied for X-ray generation and gaseous discharge from the beginning of twentieth century. At the time, a capacitor discharge which is output from charged capacitor thorough discharge switch has been used for pulsed power generation. Even now, this capacitor discharge method has been adopted widely because this is most simple and low cost method. However, if the operating voltage is critically high, the simple DC charge for the capacitor is impractical. To fix this problem, the Marx circuit system where parallel charged capacitors are connected in series with spark gap switches was invented. From the latter part of the twentieth century, the way of using a pulse forming line (PFL) began to use widely as an intermediate devise of energy storage. This is because, it is recognized that a discharge from a PFL which having constant impedance could obtain more stable output than direct discharge from a capacitor. Additionally, the output pulse duration is shortened by the introduction of PFL. With this, the peak of available power is significantly increased. Furthermore, during the decades, the development of high power semiconductor switch, magnetic core and etc. have allowed us to manufacture the pulse power source having higher energy transfer efficiency. As a result, the pulsed discharge has been recognized as one of the promised non-thermal plasma to practical use in this day.

In addition, recently, it is reported by many researchers that a shorter-duration pulsed power with higher voltage rise time gives significant improvement of energy efficiency of pollutant gas treatment. In the pulsed discharge, the short duration pulse has an effect to prevent the energy loss due to heating by terminating the voltage before the plasma phase shift to thermal plasma. Additionally, it is reported that the faster rise time of applied voltage provides more energetic electrons and a higher energy [7], [40]-[45]. From these factors, it is considered that the development of a short pulse generator is of paramount importance for practical applications. Consequently, pulse power sources for environmental applications had been shifted from a simple condenser source which generates microseconds of pulse power to a pulse forming line (PFL) source which can output sub-microseconds of pulse duration in 1990's. Furthermore, the nanoseconds pulse source has been developed since 2000's and the nanoseconds pulsed streamer discharge has shown remarkable results in energy efficiencies of pollutant control [6, 7], [46]-[49].

Hereinafter, some differences of general pulsed discharge and the nanoseconds pulsed streamer discharge will be introduced. At first, figure 6 shows images of light emissions from conventional pulsed discharges as a function of time after initiation of the discharge current [7]. The peak voltage was +72 kV with 100 ns of pulse duration and 50 ns of voltage rise time. With regard to the coaxial discharge electrode, a rod electrode made of stainless steel, 0.5 mm in diameter and 10 mm in length was placed concentrically in a copper cylinder, 76 mm in diameter. The bright areas of the framing images show the position of the streamer heads during the exposure time of 5 ns. In a rod-to-cylinder coaxial electrode, the positive streamer discharge propagate straight in the radial direction from the coaxial electrode because the interactions between the electric fields near the neighboring streamer heads are the same at somewhere in the coaxial electrode geometry. The streamer heads are associated with a higher density of ionization due to the high electric field therein, and subsequently enhanced recombination, which is followed by increased light emission [7], [40]-[45] (Fig.6). In conventional pulsed discharge, the emission at the vicinity of the rod electrode is observed 10-15ns after pulsed voltage application. The streamer heads were generated in the vicinity of the central electrode and then propagated toward the ground cylinder electrode. After full development of the streamer heads between the electrodes, the discharge phase transformed to a glow-like discharge with a large flow of current in the plasma channel produced by the streamer propagation. Finally, the glow-like discharge finished at the end of the applied pulsed voltage [7], [40]-[45] (Fig.6, Fig.7). Therefore, two stages of the discharge can be clearly defined during the conventional pulsed discharge. The first one is the 'streamer discharge', which means the phase of streamer heads propagation between electrodes. The other is the 'glow-like discharge' that follows the streamer discharge. Here it should be mentioned that in some publications, aforementioned two discharge phases are collectively called as pulse corona discharge. Additionally, the track of the streamer head which propagates from the central rod electrode to the outer cylinder electrode is called as 'primary streamer', and the subsequent streamer head that started from the central electrode at 30 ~ 35 ns (Fig.6, Fig.7) and disappeared at the middle of the electrodes gap is called a 'secondary streamer'.

By the way, as I mentioned in previous section, formation of ultimate non-thermal plasma where only electron has energy is aspired in non-thermal plasma processing. However, energy loss by background gas (ions and neutral molecules) heating starts when the discharge phase shifts to glow-like as shown in figure 6 and 7. In figure 6 and 7, until 50ns, you can see that the discharge is composed with only streamer phase which is a very high level of non-thermal condition. Therefore, if we are able to use this phenomenon which happened until 50 ns, we could become produce radicals in efficiency by using over 10 eV of energetic electrons which are exist in streamer head without ions and neutron molecules heating. Based on this idea, nanoseconds pulse generator has been developed by a lot of researchers recently.

Framing images and streak image of the discharge phenomena caused by a nanoseconds pulsed power generator (NS-PG) having a pulse duration of 5 ns and maximum applied voltage of 100 kV was developed by Prof. Namihira et al. in early 2000s [6, 7], [46]-[49] are

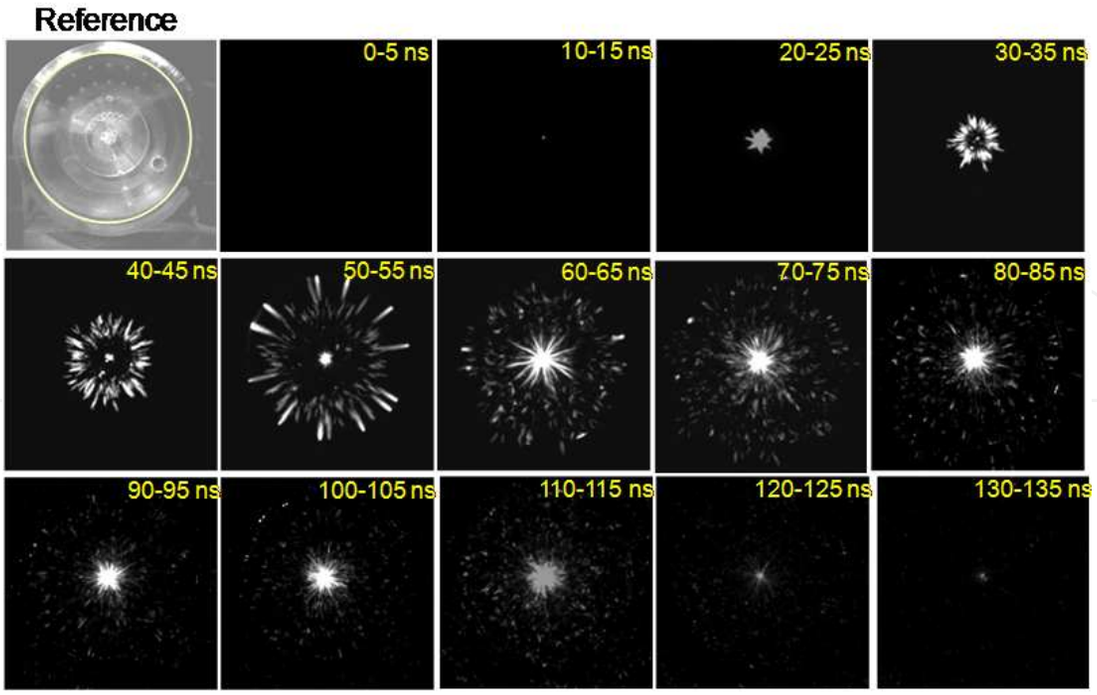


Figure 6. Images of light emissions from positive pulsed streamer discharges as a function of time after initiation of the discharge current.

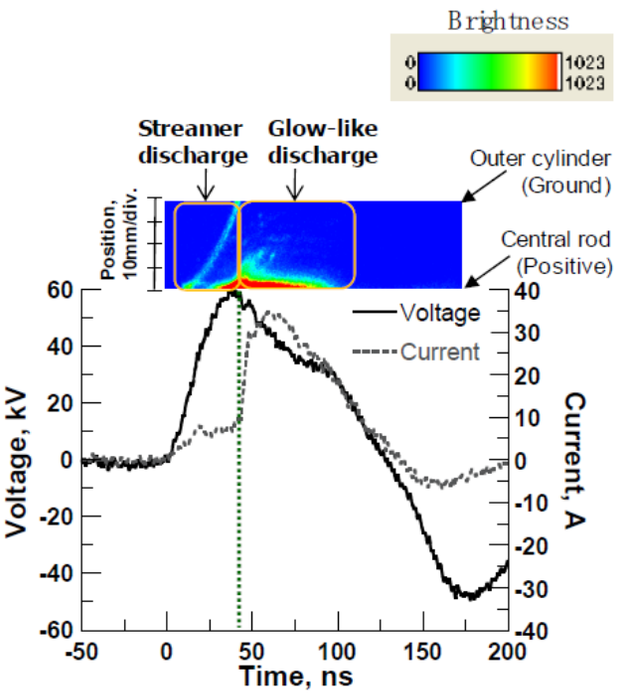
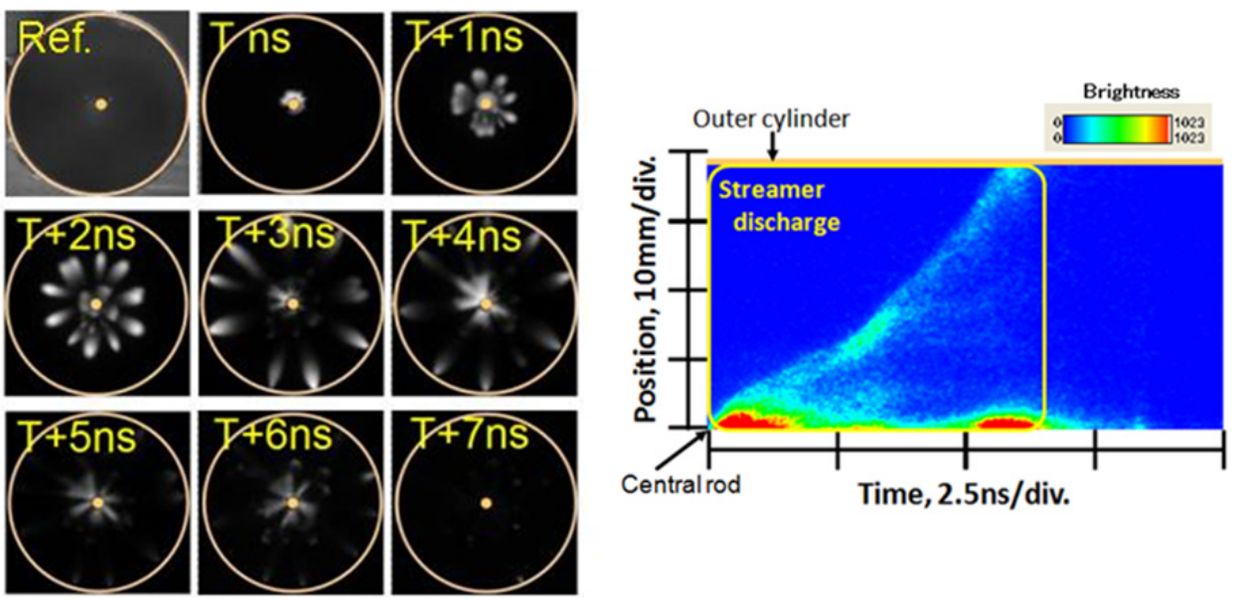


Figure 7. Typical applied voltage and discharge current in the electrode gap, and streak image for the generator with 100 ns of pulse duration. Applied voltage to electrode and discharge current through the electrodes were measured using a voltage divider and a current transformer, respectively. The vertical direction of the streak image corresponds to the position within the electrode gap. The bottom and top ends of the streak image correspond to the central rod and the surface of the grounded cylinder, respectively. The horizontal direction indicates time progression. The sweep time for one frame of exposure was fixed at 200 ns.

shown in Fig. 8, respectively [7]. In case of nanoseconds pulsed streamer discharge, the streamer heads were generated near the central rod electrode and then propagated toward the grounded cylinder electrode in all radial direction of the coaxial electrode as is the case in the conventional pulsed discharge shown in figure 6. The time duration of the streamer discharge was within 6 ns. At around 5 ns, emission from a secondary streamer discharge was observed in the vicinity of the central rod electrode. This is attributed to the strong electric field at the rod. Finally, emission from the pulsed discharge disappeared at around 7ns, and the glow-like discharge phase was not observed. As a result, as can be seen in fig. 8(b), energy loss by background gas heating caused by glow-like phase is suppressed extremely small comparison with 100ns pulsed discharge case (Fig. 7). The average propagation velocity of the streamer heads reported as 6.1 ~ 7.0 mm/ns for a positive peak applied voltage of 67 ~ 93 kV. The average velocity of the streamer heads slightly increased at higher applied voltages. Since the propagation velocity of the streamer heads is 0.1 ~ 1.2mm/ns for a 100 ns pulsed discharge, five times faster velocity is observed with the NS-PG. These characteristics comparison of pulsed discharges is summarized in Table 1. The streamer head always has the largest electric field in the electrode gap, and it is known streamer heads with higher value electric fields have a faster propagation velocity [7]. Therefore, it is understood that the faster propagation velocity of the streamer head means that the streamer head has more energetic electrons and higher energy. Consequently, the electron energy generated by nanoseconds pulsed discharge is higher than that of a general pulsed discharge. Here it should be mentioned that the voltage rise time (defined between 10 to 90%) was 25 ns for a 100 ns general pulsed discharge and 2.5 ns for the 5 ns nanoseconds pulsed discharge. Therefore, the faster propagation velocity of streamer head might be affected by the faster voltage rise time.



(a) Flaming images (b) Streak image

Figure 8. Framing and streak images of nanoseconds pulsed discharge in a coaxial electrode..

This nanoseconds pulsed power has demonstrated extremely high NO removal efficiency and ozone yield [6, 7], [46], [49]. The performances of nanoseconds pulsed discharge is summarized in Table 2. These energy efficiencies of nanoseconds pulsed power are highest value in the recent literatures about pollution control by non-thermal plasma. For the future, nanoseconds pulse power will be expected to verify its practical effectiveness by more practical experiments.

	General pulsed discharge		Nanoseconds pulsed discharge
Voltage rise time	25ns		2.5ns
Voltage fall time	25ns		2.5ns
Pulse duration	100ns		5ns
Discharge phase	Streamer	Glow-like	Streamer
Propagation velocity of streamer heads (Vapplied-peak)	0.1~1.2mm/ns (10~60kV)	-	6.1~7.0mm/ns (67~93kV)
Electrode impedance	5-17kΩ (L=10mm)	2kΩ (L=10mm)	0.3kΩ (L=200mm)

Table 1. Characteristics comparison of pulsed discharges.

	General pulsed discharge	Nanoseconds pulsed discharges	
Pulse duration	50ns	5ns	2ns
Voltage rise time	25ns	2ns	1ns
Voltage fall time	25ns	2ns	1ns
NO removal efficiency Simulated gas: NO (200ppm)/N_2 (at 60% of removal ratio)	0.37 mol/kWh	0.52 mol/kWh	0.89 mol/kWh
Ozone yield Feeding gas: Oxygen [at 10g/m^3 of O_3 concentration]	30 g/kWh	400 g/kWh	470 g/kWh

Table 2. Gas treatment characteristics comparison of pulsed discharges.

4. Present situation of non-thermal plasma on practical use

As I've discussed, non-thermal plasma has been attracted attention as a new technology of flue gas treatment for the next generation in recent years. Among the many air pollution control of non-thermal plasma, NOx removal and VOCs treatment have been particularly considered as a promising technology. Acid rain is partly produced by emissions of nitrogen

oxides such as nitric oxide (NO) and nitrogen dioxide (NO_2) originating from fossil fuels burning in thermal power stations, motor vehicles, and other industrial processes such as steel production and chemical plants. Non-thermal plasmas for removal of NOx have been produced using an electron beam, a dielectric barrier discharge, and a pulsed corona discharge at various energy effectiveness. As explained in previous section, a lot of pilot plant employing electron beam has been running. Also, various electrical discharge methods have been evolved for practical use and some examples of pilot plant using discharge methods is reported at present situation.

However, the energy efficiencies and its performances of air pollution control technique using non-thermal plasma are still unfavorable regrettably. Therefore, a plasma-catalytic hybrid system is currently employed in a practical sense. The complex of a non-thermal plasma and catalyst can be utilized these characteristics of high responsiveness to persistent substance of non-thermal plasma and high reaction selectivity of catalyst. Additionally, there are many merits of the this hybrid system from the point of view of catalyst such as reduction of precious metal catalyst use, regeneration effect of catalyst by plasma irradiation and durability improvement of catalyst by inhibition of reaction temperature etc. [50]-[60]. This hybrid system is commonly combined in one of two ways. The first is the introduction of a catalyst in the plasma discharge (in plasma catalysis, IPC), the second by placing the catalyst after the discharge zone (post plasma catalysis, PPC). Figure 9 shows typical process flow diagrams and description of main functions of IPC and PPC systems. In IPC system, catalyst is activated by plasma exposure. IPC system is a method to improve reaction efficiency and a reaction characteristic by plasma activation of catalyst. In fact, many researchers have reported composite effects such as improvement of decomposition efficiency and reduction of byproduct production by using IPC system. Moreover, it is well known that the catalyst become activated by plasma irradiation in low-temperature region where the catalyst doesn't exhibit catalytic activity. A reactor utilizing these composite effects is named as Plasma-Driven Catalysis (PDC) [58, 59]. For example, however it is considered that the plasma methods have difficulty in treating NOx by reduction, the PDC can run NOx removal by reduction process. In addition, the PDC have a stimulating effect on VOCs decomposition and conversion of VOCs to favorable product of CO_2.

The effect of IPC system differs depending on a combination of the electrical discharge method and the type of catalyst. Therefore, it is considered that the combinatorial optimization is important for IPS system. In addition, it is reported that the influence of reaction field where the catalyst is placed is quite large. Typically, catalyst should be placed on a location where the plasma density is higher in pulse corona discharge reactor or dielectric barrier discharge reactor. Because, more radials and energetic electrons are exist in there. A packed-bed reactor is a typical example of IPC system in common with PDC. A typical schematic diagram and its appearance of packed-bed reactor are shown in figure 10. In this type of reactor, surface discharge and DBD methods is generally adopted as shown in figure 10. Additionally, catalyst or ferroelectric or both are employed as packing material between electrodes. The reason why the ferroelectric is packed is extremely high energetic electrons are produced near the contact points of ferroelectric pellets packed-in the plasma

reactor, because of a huge electric field generated near the contact points [53]. As explained in the previous section, the energetic electrons are employed directly to dissociate and ionize the pollutants as well as carrier gas molecules to produce various radicals to react with and convert a part of pollutants. Fundamental characteristics of a dielectric barrier discharge (DBD) in a ferro-electric packed bed reactor have been studied for the Barium Titanate (BaTiO3) based spherical-shaped pellets for the specific dielectric constant from 660 to 104 from the viewpoint of reactor performance improvement [53]. The dielectric constant of pellet packed in the reactor affects discharge characteristics such as power consumption of the reactor, micro discharge onset voltage, number of micro discharge. As the results, the performance of packed bed plasma reactor depends on the dielectric constant and/or material of the pellet packed in the reactor.

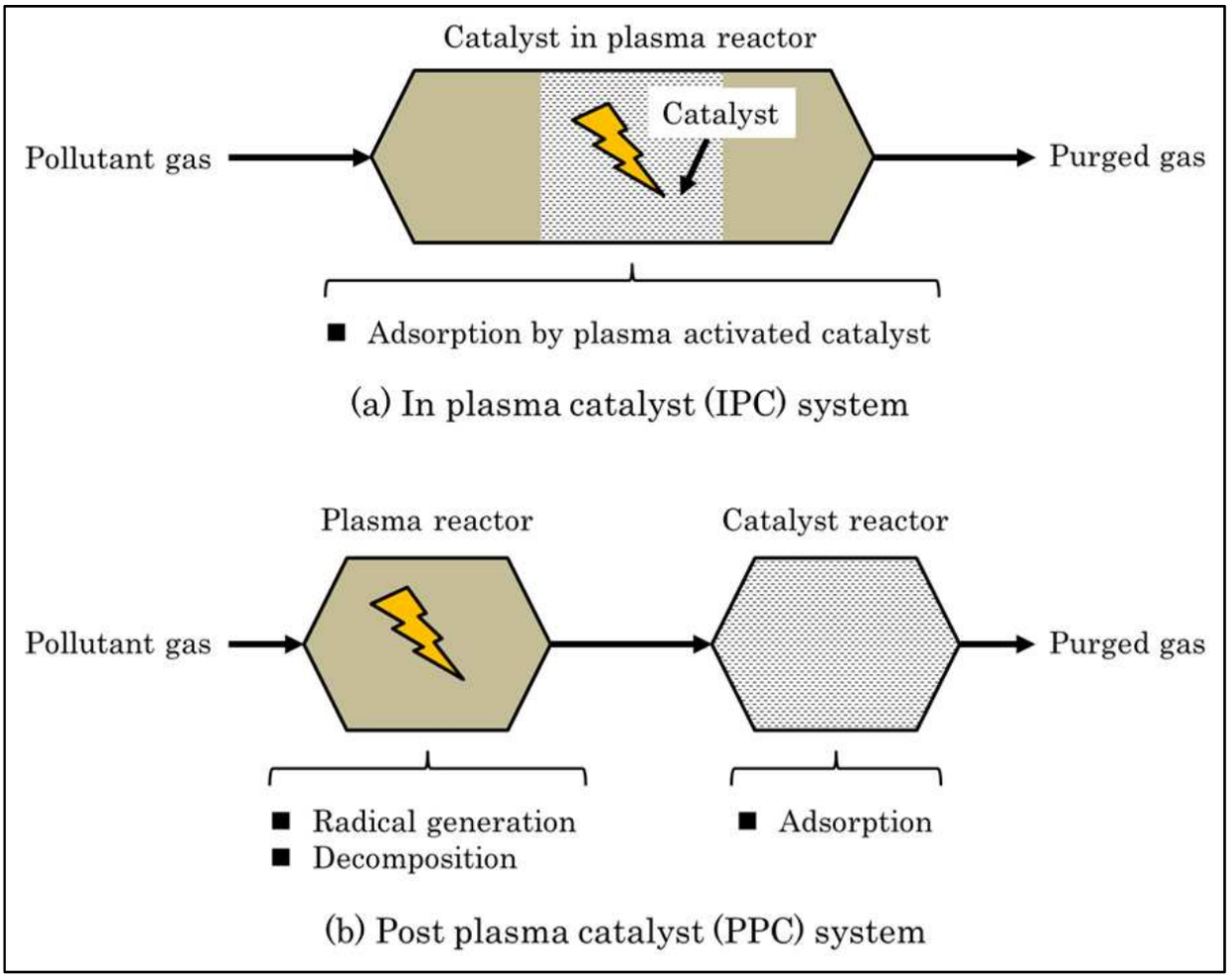
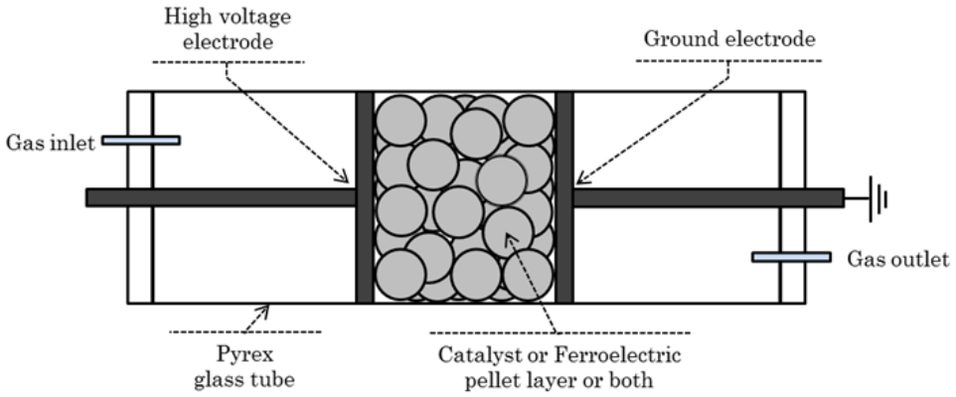
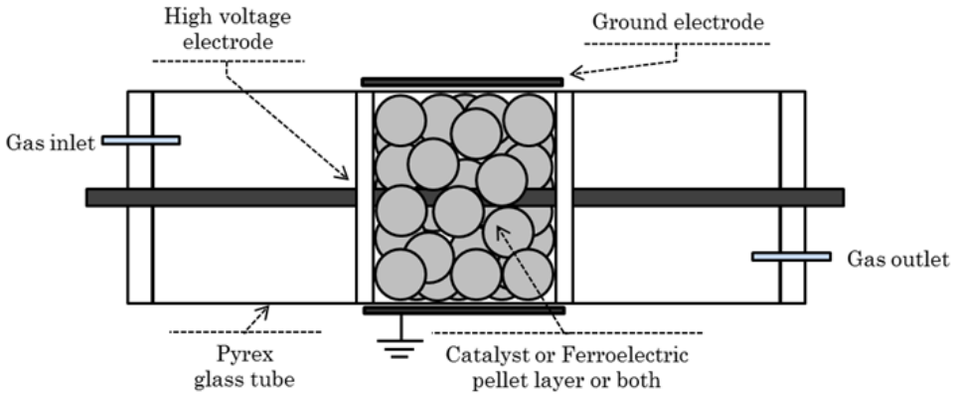


Figure 9. Typical process flow diagrams and description of main functions of Post Plasma Catalyst (PPC) and In Plasma Catalyst (IPC) system [55, 56].



(a) Surface discharge type



(b) Dielectric barrier discharge type

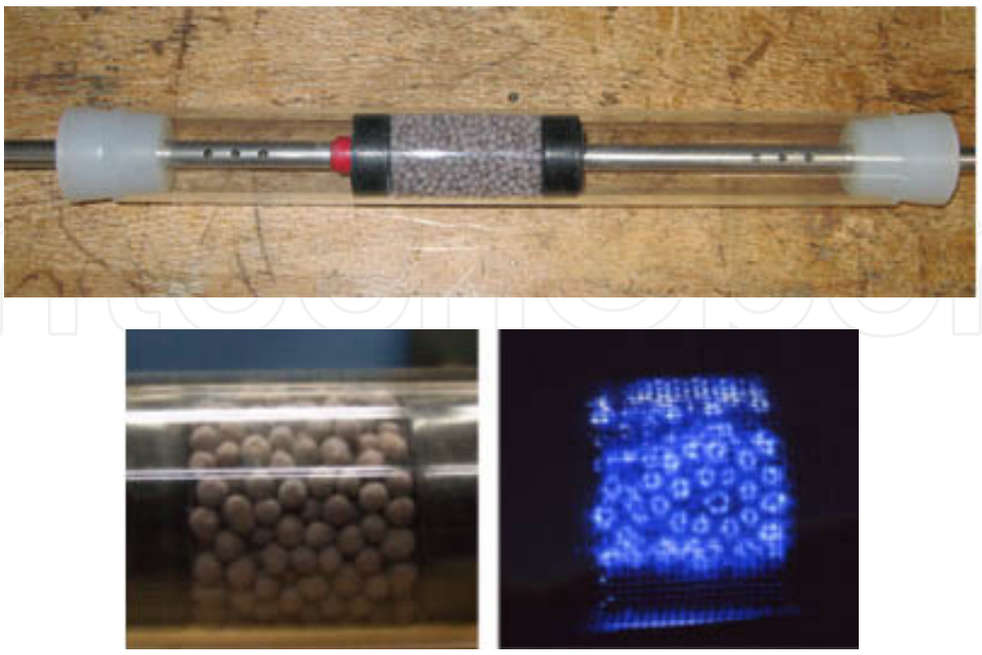


Figure 10. Schematic diagram and its appearance of packed-bed reactor (Photograph: Prof. Takaki group, Iwate University, Japan)[50], [53].

On the other hand, in the PPC system, the two functions of plasma and catalyst is completely-separated. Therefore, the configuration of reactor and system configuration are nearly independent on each other. As can be seen in figure 9(b), pollutant gas is induced into plasma reactor at first and the toxic molecules are decomposed or oxidized by energetic electrons or radicals which are generated in plasma. After that, residual contaminants that plasma couldn't treat and byproducts are removed by catalyst. Instead, this PPC system is sometimes used so as to extend time for replacement of catalyst. On another front, plasma reactor is sometimes incorporated to generate long-lived radicals such as ozone which work with catalyst as shown on figure 11. In figure 11, ozone was generated in plasma reactor, and then O radical which has stronger oxidative activity than ozone is generated by a reaction of ozone with catalyst. In consequence, VOC is decomposed by O radical to H_2O and CO_2.

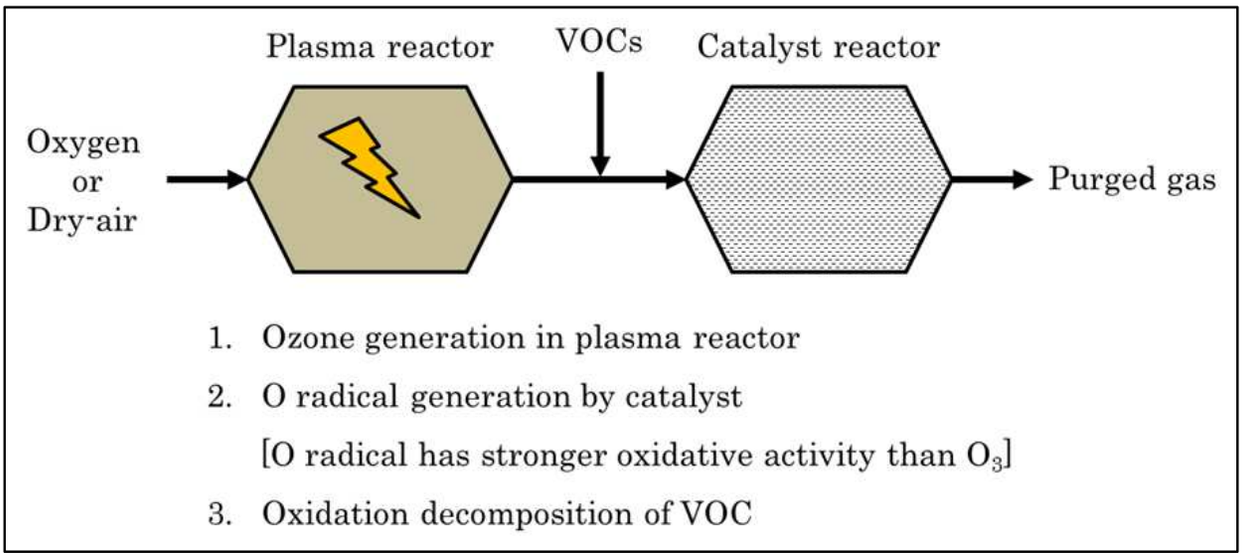


Figure 11. Example of VOC decomposition mechanism using a PPC system [62]-[64].

5. Summary

Previous prodigious studies by esteemed researchers from all ages and cultures have proven that the non-thermal plasma makes pollution control more efficient and effective. In consequence, it is recognized that the non-thermal plasma is one of the promising technologies for pollution control. The advantages of non-thermal plasma process were summarized as follows.

- Unlike conventional processes which need external combustion device or gas enrichment device, non-thermal plasma could treat industrial gas at ambient temperature and atmospheric pressure. Therefore, non-thermal plasma methods have a great advantage in energy efficiency.
- Non-thermal plasma is available for various harmful substances due to its great flexibility for the chemical reaction process (it is mainly depends on ambient gas composition). Therefore, in some case, non-thermal plasma could treat multiple toxic

molecules simultaneously. Table 3 shows that typical harmful substances in various exhaust gases. It has been proved that the non-thermal plasma could treat these toxic molecules in the literatures.

- Catalyst performance is highly improved by the concurrent use of the non-thermal plasma. The typical combined effects are increase of the reaction rate, extension of the catalyst lifetime and decrease of the activating onset temperature. Moreover, the agendas of non-thermal plasma process such as byproducts treatment could be solved by the combined use.

At the present day, a lot of practical trials are being conducted by using pilot plant of plasma reactor and more efficient and effective plasma source is developed by researchers from around the world. Air pollution control using non-thermal plasma has been edge closer to practical use. There are great hopes that air pollution control by non-thermal plasma reduce the environmental cost and make environmental effort accessible to companies and nations.

Type of Exhaust Gas	Containing Harmful Substances
Combustion flue gas	NOx, SOx, CO_2
Diesel gas	NOx, SOx, CO_2, Suspended Particulate Matter (SPM)
Industrial gas	VOCs (Aromatic series [Toluene, Benzene, acetone], Halogenated organics, HCHO), Dioxin, CFC-113, TCE

Table 3. Typical harmful substances in each exhaust gas.

Author details

Takao Matsumoto, Douyan Wang, Takao Namihira and Hidenori Akiyama
Kumamoto University, Japan

6. References

[1] Penetrante, B. M. & Schultheis, S. E., (1993). Nonthermal plasma techniques for pollution control, *Fundamentals and Supporting Technologies*, pt. A, 1-393, Springer-Verlag, New York

[2] Penetrante, B. M. & Schultheis, S. E., (1993). Nonthermal plasma techniques for pollution control, *Electron Beam and Electrical Discharge Processing*, pt. B, 1-397, SpringerVerlag, New York

[3] Penetrante, B. M. (1993). Pollution control applications of pulsed power technology, *Proceedings of 9th IEEE Int. Pulsed Power Conf.*, pp. 1-5, Albuquerque, NM, USA

[4] Müller, S.; Zahn, R. J. (2007). Air pollution control by non-thermal plasma, *Special Issue of 13th Topical Conference on Plasma Technology*, Vol. 47, Issue 7, 520–529

[5] Hackam, R.; Akiyama. H. (2000). Air pollution control by electrical discharge, *IEEE Trans. Dielect. Elect. Insulation*, Vol7, No.5, 654-683.

[6] Namhira, T.; Wang, D.; Akiyama, H. (2009). Pulsed power technology for pollution control", *Acta Physica Polonica A*, Vol. 115, No. 6, 953-955

[7] Wang, D.; Namihira, T.; Akiyama, H. (2010). Pulsed discharge plasma for pollution control, *Air Pollution*, Vanda Villanyi (Ed.), ISBN: 978-953-307-143-5, InTech

[8] Takamura, N., Wang, D., Seki, D., Namihira, T., Yano, K., Saitoh, H., Akiyama, H. (2012). *International Journal of Plasma Environmental Science & Technology*, Vol.6, No.1, 59-62.

[9] Oda, T. (2006). Atmospheric pressure nonthermal plasma decomposition of gaseous air contaminants and that diagnosis, *ICESP X – Australia*, Paper 1A1.

[10] Frank, N. R. (1995). Introduction and historical review of electron beam processing for environmental pollution control, *Radiation Physics and Chemistry*, Vol. 45, 989-1002

[11] Chmielewski, A. G.; Licki. J.; Pawelec, A.; Tyminski, B.; Zimek, Z. (2004). Operational experience of the industrial plant for electron beam flue gas treatment, *Radiation Physics and Chemistry*, Vol. 71, 439–442

[12] Duarte, C. L.; Sampa, M. H. O.; Rela, P. R.; Oikawa, H.; Silveira, C.G.; Azevedo, A.L. (2002). Advanced oxidation process by electron-beam-irradiation-induced decomposition of pollutants in industrial effluents, *Radiation Physics and Chemistry*, Vol. 63, 647–651

[13] Licki, J.; Chmielewski, A. G.; Iller, E.; Zimek, Z.; Mazurek, J.; Sobolewski, L. (2003). Electron-beam flue-gas treatment for multicomponent air-pollution control, *Applied Energy*, Vol. 75, issue 3-4, 145-154

[14] Osuda, Y. (1995). Pilot scale test on electron beam treatment of municipal waste flue gas with spraying slaked-lime slurry, *Radiation Physics and Chemistry*, Vol. 45, 1013-1015

[15] Bhasavanich, D.; Ashby, S.; Deeney, C. & Schlitt, L. (1993). Flue gas irradiation using pulsed corona and pulsed electron beam technology, *Proceedings of 9th IEEE Int. Pulsed Power Conf.*, 441-444, Albuquerque, NM, USA

[16] Chang, J.-S.; Looy, P. C.; Nagai, K.; Yoshioka, T.; Aoki, S. & Maezawa, A. (1996). Preliminary pilot plant tests of a corona discharge-electron beam hybrid combustion flue gas cleaning system, *IEEE Trans. Indust. Appl.*, Vol. 32, 131-137

[17] Penetrante, B. M.; Hsiao, M. C.; Bardsley, J. N.; Merritt, B. T.; Vogtlin, G. E.; Wallman, P. H.; Kuthi, A.; Burkhart, C. P. & Bayless, R. J. (1995). Electron beam and pulsed corona processing of volatile organic compounds and nitrogen oxides, *Proceedings of 10th IEEE Int. Pulsed Power Conf.*, 144-149, Albuquerque, NM, USA

[18] Penetrante, B. M. (1997). Removal of NO from diesel generator exhaust by pulsed electron beam, *11th IEEE International Pulsed Power Confernece*, 91-96

[19] *FEATURES, IAEA BULLETIN*, (1994). Electron beam processing of flue gases: Clearing the air, 7-10

[20] Hirano, S.; Aoki, S.; Izutsu, M.; Yuki, Y. (2000). Ebara electron-beam simultaneous SOx/NOx removal process, *Proc. 25th Int. Tech Conf. Coal Util. Fuel Syst.*, 593-604, Florida, USA

[21] Han, B.; Ko, J.; Kim, J.; Kim, Y.; Chung, W.; Makarov, I.E.; Ponomarev, A.V.; Pikaev, A.K. (2001). Combined electron-beam and biological treatment of dyeing complex wastewater. Pilot plant experiments, *Radiation Physics and Chemistry*, Vol. 64, 53–59

[22] Andrzej G. C.; Anna O. (2010). Electron beam technology for multipollutant emissions control from heavy fuel oil-fired boiler, *Journal of the Air & Waste Management Association*, Vol. 60, 932-938

[23] Doi, T.; Osada, Y.; Morishige, A.; Tokunaga, O.; Miyata, T. (1993). Pilot-plant for NOx, SO2 and HCL removal from flue-gas of municipal waste incinerator by electron beam irradiation, *Radiation Physics and Chemistry*, Vol. 42, 679-682

[24] Ignat'ev, A.V.; Kuznetsov, D. L.; Masyats, G. A.; Novoselov, Y. N. (1992). Cleaning flue gas by pulsed electron beams, *Sov. Tech. Phys. Left.*, Vol. 18, No. 11, 745-746

[25] Penetrante, B. M. (1997). Removal of NOx from diesel generator exhaust by pulsed electron beams, 11th IEEE International Pulsed Power Confernece, 91-96

[26] Nakagawa, Y.; Kawauchi, H. (1998). NOx removal in N2 by pulsed intence electron beam irradiation", Jpa. J. Appl. Phys., Vol.37, L91-L93

[27] Kuzumoto, M. (1998). Extremely narrow discharge gap ozone generator, *Journal of plasma and fusion research/the Japan Society of Plasma Science and Nuclear Fusion Research*, Vol. 74, Issue 10, 1144-1150

[28] Toyofuku, M.; Ohtsu, Y.; Fujita, H. (2004). High ozone generation with a high-dielectric-constant material, *Japanese Journal of Applied Physics*, Vol.43, No.7A, 4368-4372

[29] Kuzumoto, M.; Tabata, Y.; Shiono, S. (1997). Development of a very narrow discharge gap ozone generator -Generation of high concentration ozone up to 20 wt% for pulp bleaching-, *Kami Pa Gikyoushi/Japan Tappi Journal*, Vol. 51(2), 345-350

[30] Zhang, Z.; Bai, X.; Bai, M.; Yang, B.; Zhou, X. (2003). An ozone generator of miniaturization and modularization with the narrow discharge gap, *Plasma Chemistry and Plasma Processing*, Vol. 23(3), 559-568

[31] Takayama, M.; Ebihara, K.; Strycwewska, H.; Ikegami, T.; Gyoutoku, Y.; Kubo, K.; Tachibana, M. (2006). Ozone generation by dielectric barrier discharge for soil sterilization, *Thin Solid Films*, 506-507, 396-399

[32] Park, S. L.; Moon, J. D.; Lee, S. H.; Shin, S. Y. (2006). Effective ozone generation utilizing a meshed-plate electrode in a dielectric-barrier discharge type ozone generator, *Journal of Electrostatics*, Vol. 64, 275-282

[33] Jung, J. S.; Moon, J. D. (2008). Corona discharge and ozone generation charactristics of a wire-plate discharge system with glass-fiber layer, *Journal of Electrostatics*, Vol. 66, 335-341

[34] Sung, Y. M.; Sakoda, T. (2005). Optimum conditions for ozone formation in a micro dielectric barrier discharge, *Surface & Coatings Technology*, Vol. 197, 148-153

[35] Catalog of ozonizer, Mitsubishi Electric Corp., Japan. "http://www.mitsubishielectric.co.jp/service/mizukankyo/catalog/pdf/ozonizer.pdf".

[36] Akiyama, H. (1995). Pollution control by pulsed power, *International Power Electrics Conference*, 1937-1400

[37] Mizuno, A.; Shimizu, K.; Chakrabarti, A.; Dascalescu, L.; Furuta, S. (1995). NOX removal process using pulsed discharge plasma, *IEEE Trans. Indust. Appl.*, Vol. 31, 957-963

[38] Penetrante, B. M.; Hsiao, M. C.; Merritt, B. T.; Vogtlin, G. E.; Wallman, P. H. (1995). Comparison of electrical discharge techniques for nonthermal plasma processing of NO in N_2, *IEEE Transactions on Plasma Science*, Vol. 23, No. 4, 679-687

[39] Yoshida, T.; Tochikubo, F.; Watanabe, T. (1995). Diagnostics of pulsed corona discharge for DeNOX process, *11th International Conference on Gas Discharges and Their Applications*, Vol. 2, 410-413

[40] Wang, D.; Okada, S.; Matsumoto, T.; Namihira, T.; Akiyama, H. (2010). Pulsed discharge induced by nano-seconds pulsed power in atmospheric air", Vol. 38, No. 10, 2746-2751

[41] Namihira, T.; Wang, D.; Katsuki, S.; Hackam, R.; Akiyama, H. (2003). Propagation velocity of pulsed streamer discharges in atmospheric air, *IEEE Transactions on Plasma Science*, Vol. 31, No. 5, 1091-1094

[42] Tochikubo, F.; Teich, T. H. (2000). Optical emission from a pulsed corona discharge and its associated reactions, *Jpn. J. Appl. Phys*, Vol. 39, No. 3A, 1343-1350

[43] Tochikubo, F.; Watanabe, T. (2000). Two - dimensional measurement of emission intensity and NO density in pulsed corona discharge, *International Symposium on High Pressure Low Temperature Plasma Chemistry*, Vol. 1, 219-223

[44] Tochikubo, F.; Miyamoto, A.; Watanabe, T. (1995). Simulation of streamer propagation and chemical reaction in pulsed corona discharge, *11th International Conference on Gas Discharges and Their Applications*, Vol. 1, 168-171

[45] Ono, R.; Oda, T. (2007). Optical diagnosis of pulsed streamer discharge under atmospheric pressure, *International Journal of Plasma Environmental Science & Technology*, Vol. 1, No. 2, 123-129

[46] Namihira, T et al. (2000). Improvement of NOx removal efficiency using short width pulsed power, *IEEE Transactions on Plasma Science*, Vol. 28, No. 2, 434-442

[47] Namihira, T et al. (2007). Characterization of nano-seconds pulsed streamer discharges, *2007 IEEE Pulsed Power and Plasma Science Conference*, Albuquerque, USA, 572-575

[48] Tamaribuchi, H et al. (2007). Effect of pulse width on ozone generation by pulsed streamer discharge, *2007 IEEE Pulsed Power and Plasma Science Conference*, Albuquerque, USA, 407-410

[49] Matsumoto, T.; Wang, D.; Namihira, T.; Akiyama, H. (2011). Performances of 2 nanoseconds pulsed discharge plasma, *Japanese Journal of Applied Physics*, Vol. 50, No. 8, 08JF14-1-5

[50] Takaki, K.; Chang, J.-S.; Kostov, K.G. (2004). Atmospheric pressure of nitrogen plasmas in a ferroelectric packed bed barrier discharge reactor. Part I. Modeling, *IEEE Trans. Diel. Elect. Insul.*, Vol. 11, No. 3, 481-490

[51] Uchida, Y.; Takaki, K.; Urashima, K.; Chang, J.-S. (2004). Atmospheric pressure of nitrogen plasmas in a ferroelectric packed-bed barrier discharge reactor. Part II. Spectroscopic measurements of excited nitrogen molecule density and its vibrational temperature, *IEEE Trans. Diel. Elect. Insul.*, Vol. 11, No. 3, 491-497

[52] Rajanikanth, B. S.; Srinivasan, A. D. (2007). Pulsed plasma promoted adsorption/catalysis for NOx removal from stationary diesel engine exhaust, *IEEE Transactions on Dielectrics and Electrical Insulation*, Vol. 14 (2). 302-311.

[53] Takaki, K.; Takahashi, S.; Mukaigawa, S.; Fujiwara, T.; Sugawara, K.; Sugawara, T. (2009). Influence of pellet shape of ferro-electric packed-bed plasma reactor on ozone generation and NO removal, *International Journal of Plasma Environmental Science and Technology*, Vol. 3, No. 1, 28-34

[54] Yamamoto, T.; Yang, C. L.; Beltran, M. R.; Kravets, Z. (2000). Plasma assisted chemical process for NOx control, *IEEE transactions on industry applications*, Vol. 36, No. 3, 923-927

[55] Chena, L.; Zhanga, X.; Huanga,L.; Le, L. (2010). Application of in-plasma catalysis and post-plasma catalysis for methane partial oxidation to methanol over a Fe2O3-CuO/γ-Al_2O_3 catalyst, *Journal of Natural Gas Chemistry*, Vol. 19, issue 6, 628-637

[56] Durme, J. V.; Dewulf, J.; Leys, C.; Langenhove, H. V. (2008). Combining non-thermal plasma with heterogeneous catalysis in waste gas treatment, *Applied Catalysis B: Environmental*, Vol. 78, 324–333

[57] Vandenbroucke, A.; Morent, R.; Geyter, N. D.; Dinh, M. T. N.; Giraudon, J. M.; Lamonier, J. F.; Leys, C. (2010). Plasma-catalytic decomposition of TCE, *International Journal of Plasma Environmental Science & Technology*, Vol. 4, No. 2, 135-138

[58] Durme, J. V.; et al. (2008). Combining non-thermal plasma with heterogeneous catalysis in waste gas treatment, *Applied Catalysis B*, Vol. 78, 324-333

[59] Fan, H. Y.; et al. (2009). High-efficiency plasma catalytic removal of dilute benzene from air, *J. Phys. D: Appl. Phys.*, Vol.42, 225105

[60] Kim, H. H.; Oh, S. M.; Lee, Y. H.; Ogata, A.; Futamura, S. (2005). Decomposition of gas-phase benzene using plasma-driven catalyst (PDC) reactor packed with Ag/TiO_2 catalyst, *Applied Catalysis B: Environmental*, Vol. 56, 213-220

[61] Kim, H. H.; Lee, Y. H.; Ogata, A.; Futamura, S. (2006). Effect of different catalysts on the decomposition of VOCs using flow-type plasma-driven catalysis, *IEEE Transactions on Plasma Science*, Issue 3, Vol. 3, 984-995

[62] Ogata, A.; Saito, K.; Kim, H. H.; Sugasawa, M.; Aritani, H.; Einaga, H. (2010). Performance of an ozone decomposition catalyst in hybrid plasma reactors for VOC removal, *Plasma Chem. Plasma Process*, Vol. 30, 33-42

[63] Kim, H. H.; Ogata, A.; Futamura, S. (2007). Complete oxidation of volatile organic compounds (VOCs) using plasma-driven catalysis and oxygen plasma, *International Journal of Plasma Environmental Science & Technology*, Vol.1, No.1, 46-51

[64] Demidiouk, V.; Chae, J. O. (2005). Decomposition of volatile organic compounds in plasma-catalytic system, *IEEE Transactions on Plasma Science*, Vol. 33, No. 1, 157-161