Doctoral Dissertation

The Influence of Pulsed Voltage Shape and Period on Active Species Production by Pulsed Discharge

（パルス電圧形状と周期がパルス放電による活性種生成に与える影響）

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ABSTRACT

Pulsed power technology has been being researched and developed from last century. From the theorical concept of pulsed power technology, the same amount of electric power can be released with very short duration when compared with the original duration. There are some ways to generate the pulsed power, such as compressing power, which can generate pulsed power by compressing electric power and releasing the same amount of this electric power in shorter time. There is also a different way to generate pulsed power by adding plenty of low electric power together and releasing the sum of added electric power in the same duration. There are a lot of generators that can generate pulsed power efficiently, such as MARX or Linear Transformer Driver (LTD). Besides with the research of improving the performance of pulsed power generator, a lot of studies have been done on the application of pulsed power technology. At the present, pulsed power technology has been being applied in a lot of fields. As one of the applications of pulsed power technology, water treatment based on pulsed discharge has been studied for decades. As a result, active species (ozone, OH radical) produced by pulsed discharge have been considered as effective element for treating harmful chemical in water. And the study of this dissertation focuses on the effect of pulsed discharge on producing these active species (ozone, OH radical) to investigate the way to improve the production of active species for water treatment application by pulsed discharge.

Firstly, evaluation effect of the characteristic of pulsed discharge on ozone production in atmospheric pressure has been done. Linear Transformer Driver (LTD) system was used to generate electric pulse for all experiments. In order to investigate effect of the voltage in the last half of pulse on production of ozone, pulse with waveform of lowered voltage in the last half of pulse has been applied for evaluate efficiency of ozone production. The average total input energy has been maintained as unchanged when different waveform was applied. As a result of the experiment, the concentration and energy efficiency of ozone production can be improved with lessening voltage at the last half of pulse. The neutralization of ion when the primary streamer head has already gone to the ground lead to the decrease of electric field in this phase and difficulty in ozone production, which may be considered as physical reason for this phenomenon.

Next, interval between two pulse was investigated for its effect on ozone production. Pulse interval with value varied from 1μs to 10ms was applied for evaluating ozone production. As the result, when the first pulse was applied to the electrodes in water treatment system, and the second

pulse was applied after more later time, the concentration and energy efficiency of ozone production can be increased. The reason is more occurrence of ozone decomposing reaction released by shorter pulse interval.

In the next part of this study, pulsed discharge was investigated for its effect on OH radical production. Pulse waveform of lowered voltage at the latter half was applied to evaluate OH radical production. The average input energy of pulse per one unit of time was also maintained as unchanged for all experiments in this part. From the experiments, it can be confirmed that amount and energy efficiency of OH radical production can be improved with elimination voltage at the latter half of pulse. As the reason mentioned above for ozone production, the decrease of electric field in the later half phase affected by ion neutralization may be the reason for difficulty in OH radical production in this phase.

Finally, evaluation of OH radical production of only the last phase of pulsed discharge was done to investigate effect of electric field on OH radical production in only the last phase of pulsed discharge. More voltage was added up to the last half of pulse and the OH radical production in only the last phase of pulsed discharge was analyzed. It can be concluded from the experimental results that when the strength of electric field in the last phase is increased, the amount and energy efficiency of OH radical production in this phase can be improved.

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Chapter I

INTRODUCTION

1.1 PULSED POWER TECHNOLOGY

1.1.1 DEFINITION OF PULSED POWER

From the beginning, concept of generating pulsed power is obtaining certain electrical power and outputting the same electrical power in shorter time by compressing this amount of electrical power. As concept shown in the following Fig. 1, instead of applying lower electric pulse in longer duration to obtain certain electrical power, this certain amount of electrical power can be obtained in much shorter time by compressing power [1][2].

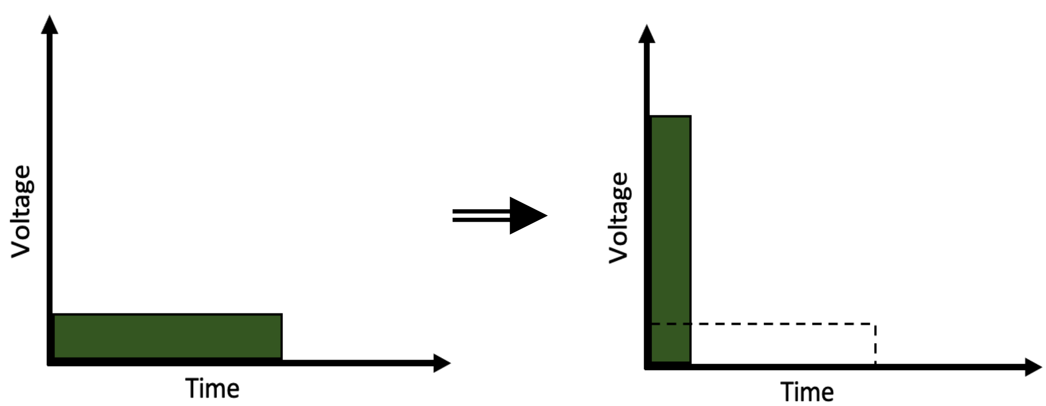


Fig. 1. Concept of generation of pulsed power by compressing electrical power.

Later, concept of adding electrical power has been introduced as a new method of generating pulsed power. As in the Fig.2, to obtain certain electrical power with the same duration, a lot of low pulse with short duration are added together. As a result, higher electric pulse can be obtained at the same time.

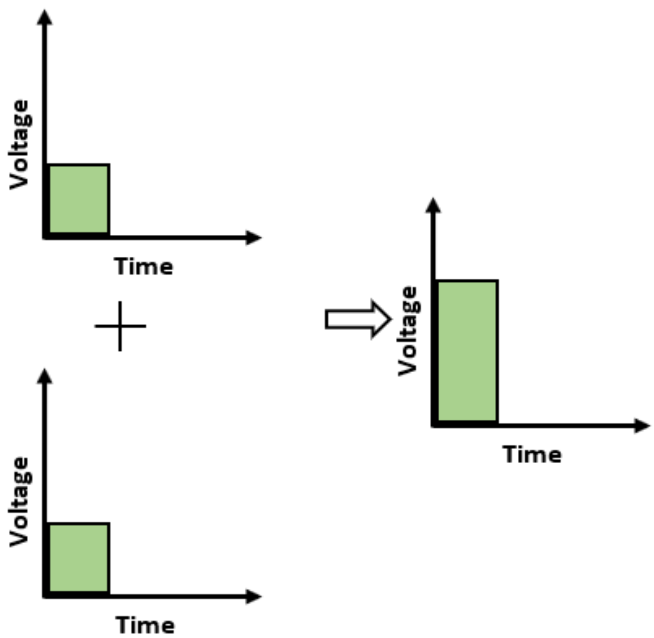


Fig. 2. Concept of generation of pulsed power by adding low pulse together.

1.1.2 HISTORY OF DEVELOPING PULSED POWER TECHNOLOGY

Looking back to the first haft of the 20th century, there were a lot of research and developing activities that have mechanism similar to the concept of pulsed power technology [3]. Here, most of research and developing activities is developed in military field. T. A. Mehlhorn et al. in [3] shown that in the United States they had started developing heavy weapons which was used for plenty of purposes such as national security, and in that period pulsed power technology was applied as main key for their researching and developing activities. At the same time plasma physics had been mainly focused together with pulsed power technology.

In the next haft of the 20th century, there were lots of development and research focused on developing modern model of pulsed power generator, as pointed out in [3], the contemporary model of machines that could generate pulsed power had been firstly shown in the 1960s, and the model of machine that can generate pulsed power with short pulse period has also been performed in this time. Also, the development of nanosecond order electrical pulse was performed in this period as I. Smith showed in [4].

The following Fig. 3 is a brief of the development of pulsed power technology related with each timeline, and the information is based on the review of [3].

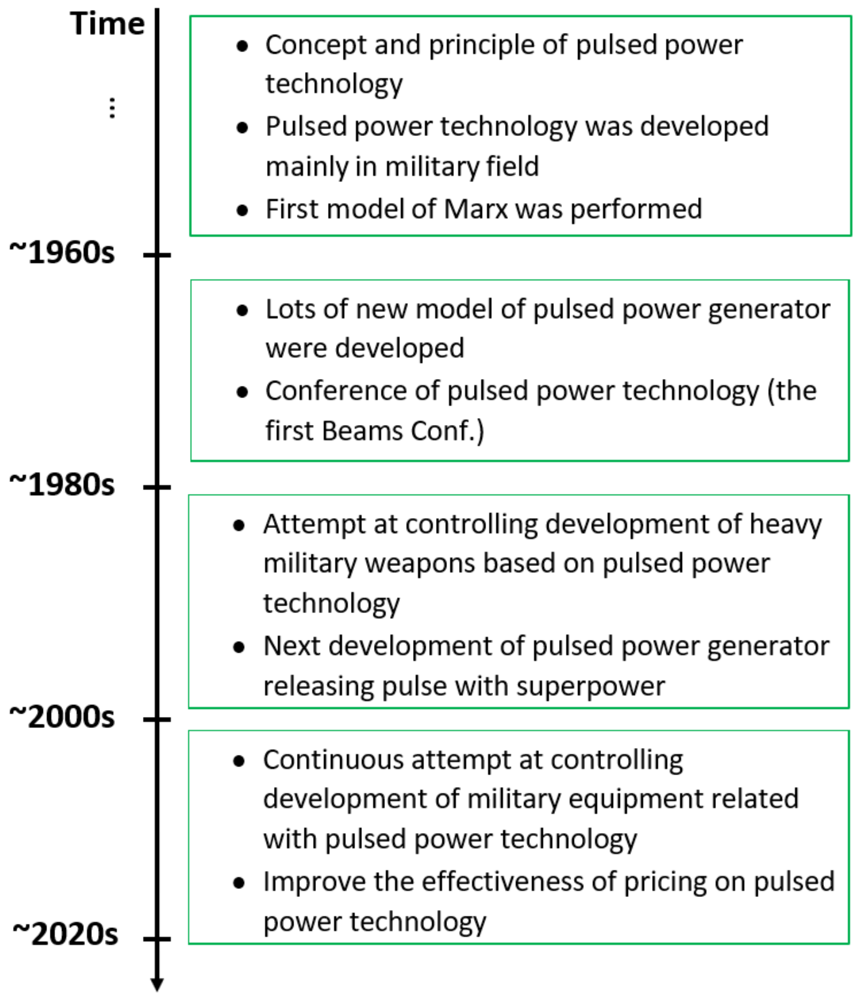


Fig. 3. The brief timeline of developing pulsed power technology.

1.2 APPLICATION OF PULSED POWER TECHNOLOGY

1.2.1 APPLICATION OF PULSED POWER TECHNOLOGY IN BIOLOGICAL FIELD

Pulsed power technology is also being applied in a lot of industrial fields beside with military, such as biology, environmental science [5]. About application of pulse power technology on biological science, J.Kolb et al. [6] has performed experiment showing the effect of electric fields generated by nano-second pulse discharge on biological cells, and these experiment results have shown the investment of the effect and changing in real biological cells realized by applying short nano-second pulse [5] [6]. The pulse generator used to generate these short nano-second pulse was Blumlein line, and fluorescence method was applied to investigate changing in biological cells [6]. J. Kolb et al. [6] also pointed out some secondary effects of nano-second pulse discharge on biology.

H. Akiyama et al. [5] has also shown another application of pulse power technology on biological field, such as eliminating a lot of kind of bacteria to treating the air environment, and the type of pulsed discharge mentioned was gas discharging. There are plenty of studies about applying pulse power technology to treating harmful bacteria in air environment. Typically, the study in [7] has shown the effect of pulsed discharging on treating air and water environment. The main type of pulsed discharge used in these experiments is classified as corona pulsed discharge, microorganisms in air environment treated by pulses discharge was investigated, these experiment results have shown the relation between reduction of microorganisms and corona pulsed discharge [7].

As in [5], there are a lot of another studies of application of pulsed power technology in biological fields, such as treating biological cells in water, improving efficiency of harvesting in agriculture [8][9][10]. These studies have shown the effect of high-voltage pulsed power on biological cell in general or plant in agricultural field clearly, a comparison between some pulsed power generating method and pulsed power supply was also shown in these studies.

Furthermore, H. Akiyama et al. has mentioned that pulsed power technology has also been applied to treating cell in live body, which can open another application of pulsed power technology in medical field [5] [11] [12].

The following Table. 1 shows some typical applications of pulsed power technology in biological field and detail examples for each application.

Table. 1 Typical application of pulsed power in biology and detail example.

Application of pulsed power technology in biology	**Type of pulsed power generator**	**Typical example of application**
Reducing biological live organism in polluted environment	Pulse source that can generate corona pulsed discharge	Elimination of bacteria in air or water environment [7]
Resolving problem of water environment	Blumlein PFN	Treatment of cyanobacteria in water environment [8]
Agricultural harvest	MARX and C-W circuit	Improving the development of mushroom in real farm [10]
Medical treatment	PFN	Treatment of some issues related with cell and DNA [12]

1.2.2 PULSED POWER TECHNOLOGY FOR RESOLVING PROBLEM OF AIR ENVIRONMENT

Here, high-voltage pulsed power has been mainly applied to resolve a lot of problem of environment. Based on a lot of chemical reactions that were created by high-voltage pulsed discharge, pulsed power technology has been being applied to treat polluted air environment or water environment. The key point for this application of pulsed power technology is the production of a lot of strong active species such as ozone, hydroxyl radical, oxygen radical. These active species are mainly produced by high-voltage pulsed discharge, and plasmas also play role in these productions of active species.

For the application of pulsed power technology in treatment of polluted gas, there are lots of studies on applying pulsed power technology to removing or reducing harmful pollutants in air environment. Widely usage of fossil production by human for daily life activities and industrial activities can be considered as reason of polluted air environment, so development of air treatment method is very important. Therefore, for decades, there are a lot of research and development focusing on air treatment and applying pulsed power technology on treating polluted air is one of gas treatment method. By applying high-voltage pulsed discharge, non-thermal plasma can be generated in atmospheric pressure, and non-thermal plasma has been considered as key to treat pollutants in the air [5] [13]. Based on phenomenon of dissociation and ionization between lots of production produced by high-voltage pulsed discharge and gas, plenty of strong radical can be formed and these radical can react with a lot of chemical connection in pollutants [13]. As a result, strong radical produced by high-voltage pulsed discharge can remove or reduce the concentration of pollutant in the air. Until now, there are a lot of studies on improving energy efficiency of removing pollutants in the air environment and deeply understanding the mechanism of producing these radicals. For more detail, the following Table. 2 lists some harmful pollutants in the air that can be treated by high-voltage pulsed discharge [5].

Table. 2 Typical examples of applying pulsed power technology for treating harmful pollutants in the air.

Type of pulsed discharge	**Treatable harmful pollutant in the air**	**Typical main treating reaction**
Corona discharge	Nitric oxides (NO_x) [14] [15]	Removing NO/NO_x [14]
Streamer discharge	Nitric oxides (NO_x) [16]	Removing NO_x [16]
Nanosecond pulsed discharge	Carbon dioxide (CO_2) [17]	Conversing CO_2 [17]
Dielectric barrier discharge	Sulfur dioxide (SO_2) [18][19]	$O + SO_2 + M \rightarrow SO_3 + M$ $O_3 + SO_2 + M \rightarrow SO_3 + O_3$ [17]
Pulsed corona discharge	Volatile Organic Compounds (VOCs) [20]	Decomposing VOCs (toluene, butyl acetate, isopropyl alcohol) [20]

1.2.3 APPLICATION OF PULSED POWER TECHNOLOGY IN WATER TREATMENT

Another popular application of pulsed power technology on resolving environmental problem is water treatment based on high-voltage pulsed discharge. For water treatment, characteristic of high-voltage discharge in liquid solution or water is mainly applied [5].

Until now, there are a lot of research on applying high-voltage pulsed discharge in liquid, which lead to widely developing efficient method of treating wastewater. T. Namihira et al. [21] performed experiment to determine the temperature of plasma and the electron density realized by plasma; all these plasma in their experiments was generated by high-voltage pulsed discharge directly in water solution. There are so many other studies about this pulsed discharge in water. For example, plasma generated by high-voltage pulsed discharge in water has been studied for more understanding the ionization potential, the bound state of electron affected by charge interaction and some other elements by X. Lu et al. in [22]. They have created model to confirm the influence of this model on energy of plasma generated by pulsed discharge underwater, and the simulation has been done to confirm their experiment results [22]. In the research of [22], the electrode was put directly into water to create high-voltage pulsed discharge in water, and their results has shown that their model can be applied to study the phenomena of pulsed discharge underwater with different discharging time. Or K. Grosse et al. [23] has conducted research on high-voltage pulsed discharge in water with nano-second pulse width, the voltage of pulse was varied from 14 kV to 26 kV and the bubble realized by plasma underwater was analyzed, then the temperature and density of electron was also analyzed for more understanding about the high-voltage pulsed discharge in water which have a lot of effects on studies of application pulsed power technology in the future.

Not only pulsed discharge directly into water was studied for application of pulsed power technology in water treatment, but there are also many studies about high-voltage pulsed discharge indirectly into water, such as wastewater flows through discharge area in droplet and the electrode was not put into water, or the pulsed discharge contacts only with the surface of water. As studies in [24] [25] [26] [27] [28] [29], the electrodes in these experiments have been designed for high-voltage pulsed discharge not happening underwater. For the experiment in [24] [25] [26], it has been confirmed that when wastewater was sprayed through the discharge area in droplet form, there are a lot of advantage in water treatment by pulsed power technology. Or when the high-voltage pulsed discharge only happens above the surface of water, it also has effect on decomposing pollutants [27] [28] [29].

1.2.4 OTHER APPLICATION OF PULSED POWER TECHNOLOGY

Furthermore, in the Table. 3, another application of pulsed power technology for industry is listed with typical example [5].

Table. 3 Other fields and typical example of application pulsed power technology.

Field applying pulsed power technology	Typical example	Reference
Producing material industry	Treating surface of some materials with complicated detail	[30] [31] [32]
	Recycling the industrial material	[33]
	Microwave heating	[34]
Chemical industry	Conversion of methane and acetylene	[35]
Excimer laser technology	Driving excimer laser source	[36] [37]

1.3 TYPE OF ELECTRIC DISCHARGE

1.3.1 OVERVIEW OF ELECTRICAL DISCHARGE [38]

In this part, characteristic and types of electrical discharge will be introduced more in detail. Electrical discharge is the main of pulsed power technology, and based on characteristic of electrical discharge, electrical discharge is separated to many types and there are some different applications for each type of electrical discharge.

Firstly, in order to make discharge happen, the phenomenon of break down need to be available, as in [38], a voltage of larger than 320 V is needed to make the break down phenomenon happen between the electrodes with an air gap, and this break-down-voltage may be smaller if the air gap is reduced. At the same time, an electric current is required to maintain this phenomenon of break down [38].

As mentioned above, with the phenomenon of break down, an electric discharge can be generated and there are a lot of types of the electric discharge. For example, as listed in [38], the electric discharge can be classified as the following type: glow discharge, arc discharge, corona discharge, and spark discharge. Phenomenon of breakdown has been known as a complicated phenomenon. Depended on the value of the voltage that applied to the electrodes, there are a lot of different types of the electric discharge, such as when the value of voltage applied to the electrodes is larger than the value of the break-down-voltage and the current which has value of the range from mA to A appears here, a glow discharge will happen between these electrodes [38]. When this electric current become larger than the value of the glow discharge, an arc discharge (long arc discharge) will happen, and because the required voltage applied to the electrodes for the happen of arc discharge can be smaller than the required voltage for the glow discharge so the arc discharge can happen individually without the happen of glow discharge [38].

The following Fig. 4 is the summarized condition of the voltage and current needed for each type of the glow discharge and arc discharge, this information was collected from [38].

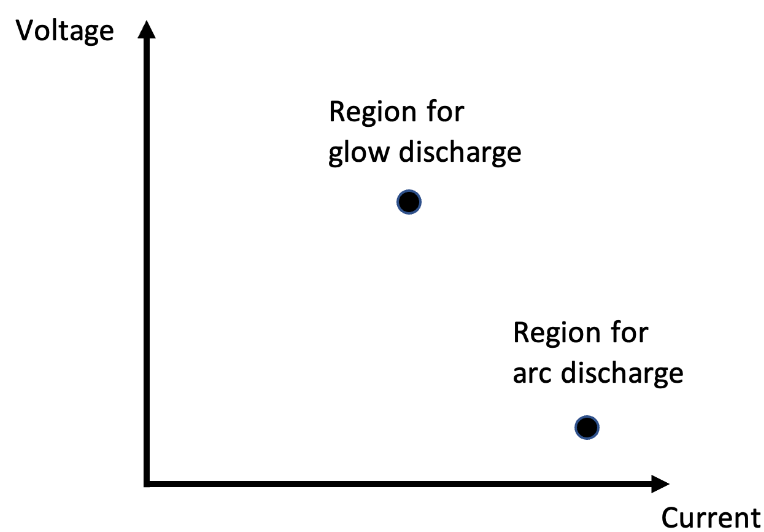


Fig. 4 Condition of value of the voltage and current for the glow discharge and arc discharge.

About the corona discharge, as mentioned in [38], corona discharge will happen near the surface that has curvy shape such as round corner or wire, because the electric field close to these surfaces can be greater. For the condition for the corona discharge happening between the electrodes, the required voltage applied to the electrode for the happen of corona discharge is smaller than the required voltage for the glow discharge or arc discharge [38]. Furthermore, corona discharge was described in [39] as a gas discharge happened between the electrodes, and the difference of the corona discharge with the other type of discharge is the difference of the low electric field region and high electric field region in the area between the electrodes. The field of charge plays main role in formation of voltage, current and distribution of current density in the gap [39]. The phenomenon of corona discharge can be considered as the popular discharge that has been being applied widely for many fields such as material industry, chemical industry, or environmental solution.

The spark discharge is mentioned in [38] as a discharge that just last for a short time and this phenomenon of discharge can be seem like short-lived event. And the spark discharge also happens between the electrodes when the electric field becomes strong enough, at the time of the enough electric field appearing between the electrodes, the ion concentration will become higher and the characteristic of the object between these electrodes will become as conductor in a short while [40].

1.3.2 DIELECTRIC BARRIER DISCHARGE

For the non-thermal electrical discharge, there are some different types of generation, and the dielectric barrier discharge is one of these types. Until now, the dielectric barrier discharge has been well known as the electrical discharge happening between two electrodes and the available of insulator between these electrodes is required to generating the dielectric barrier discharge [41] [42] [43] [44].

The following Fig. 5 to Fig .7 show the simple view of different typical types of dielectric barrier discharge, the information in Fig. 5 to Fig .7 was referred from review of [41] [42].

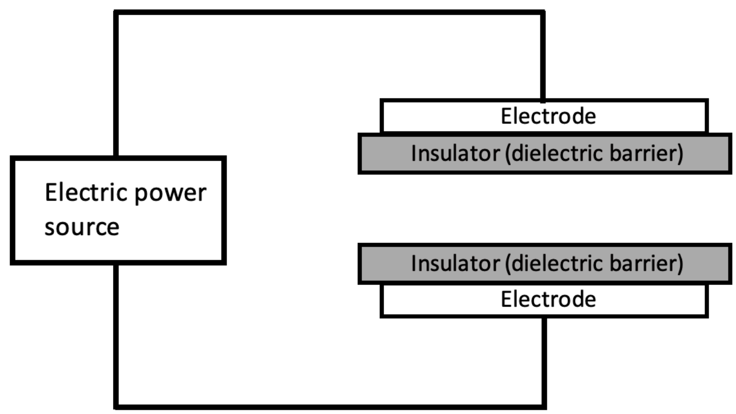


Fig. 5 The typical view of dielectric barrier discharge with two layers of insulator between electrodes.

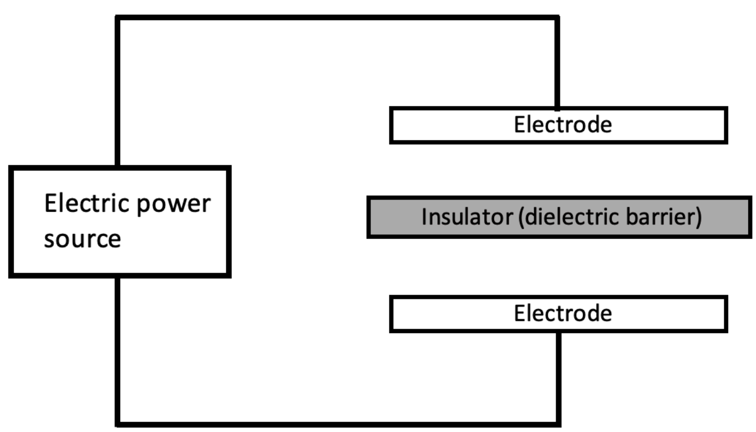


Fig. 6 The typical view of dielectric barrier discharge with one layer of insulator in the middle of two electrodes.

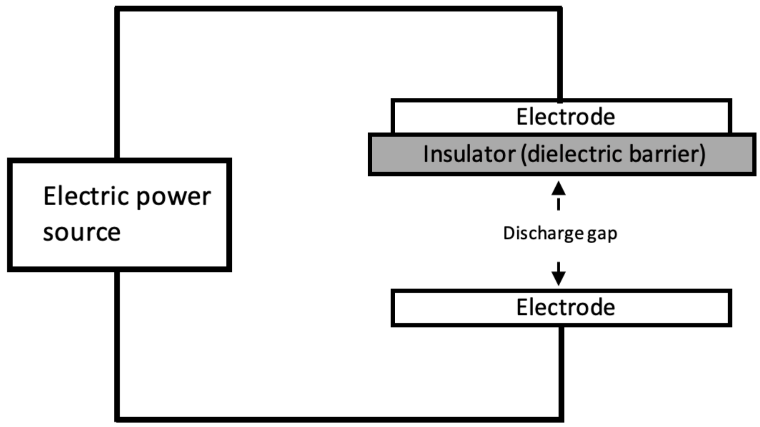


Fig. 7 The typical view of dielectric barrier discharge with one layer of insulator and discharge gap between two electrodes.

The phenomenon of break down needs to be appeared to allow the current flow through the discharge gap when the suitable electric field is generated [41]. U. Kogelschatz in [41] also pointed out that the material of insulator preferred for the layer of dielectric barrier should be silica, ceramic, enamel or polymer and a repetitive rate should be high for the current can flow through the dielectric barrier. When the 10^5Pa-order pressure is applied and a high electric field available in the gap, micro discharge may be appeared and this condition is suitable for the application of gas treatment for resolving environmental problem, generation of ozone or generating the formation of excimer [41]. For some cases, as the experiments in study of [45], P. Vanraes et al. shown that when the layer of water is available between gap of the dielectric barrier discharge with one layer of insulator (as Fig. 7), this water layer may have function as a dielectric barrier for few nanoseconds. The following Fig. 8 is the simple view of experimental setup for experiment of P. Vanraes et al. and this outline is sketched based on the information of [45].

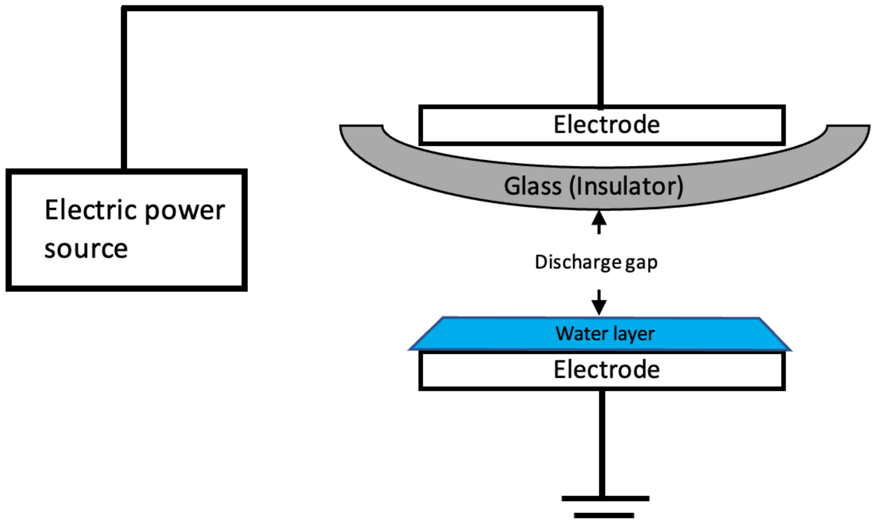


Fig. 8 Outline of the setup and design of the electrodes for dielectric barrier discharge of the experiment in [45].

1.3.3 STREAMER DISCHARGE [46] [47]

The streamer discharge is one of the most popular pulsed discharge that is being applied in many fields. In order to generate streamer discharge, high electric field is required for contacting with the gas, or in other word, high voltage need to be applied to electrode in a very short time [46]. And the main factor for generation of streamer discharge has been pointed out by S. Nijdam et al. in [46] that the electrons need to be accelerated where the electric field is available, and the electrons need to collide with other molecules such as gas molecules in the discharge area. The following Fig. 9 performs the process that the electrons collide with other molecules in the discharge area when streamer discharge is generated, the information in Fig. 9 was referred from the review of [46]. About this process, firstly, the energy conversation of kinetic energy of electron will be happened. Next, electron in the discharge area with high voltage applied to the electrodes will excite other molecules in this area by the conversed kinetic energy, and the ionization of these molecules will happen in the next step. Finally, a lot of negative ions will be produced in the discharge area as a result of the connection between electron and other molecules [46].

Until now, pulsed streamer discharge has been well known as one of the most efficient way to generate non-thermal plasma, so streamer discharge has some interest for generating non-thermal plasma when compared to generating plasma with thermal; such as it will allow some reactions which cannot happen in thermal condition happen or it can improve the energy efficiency because there are not any requirement of heating which consumes lots of energy [46].

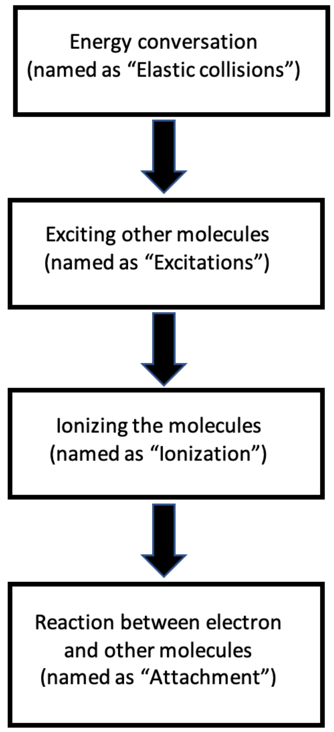


Fig. 9 The process that electron collide with other molecules in the discharge area.

The following Fig .10 is the process that the energy from the electric source is conversed to the final target of application for many fields by pulsed discharge in atmospheric pressure, and this graph was drawn based on the information in [46]. Therefore, pulsed electrical discharge can be classified as one of the steps of conveying energy from the original electric source to the applications for many fields in the final step.

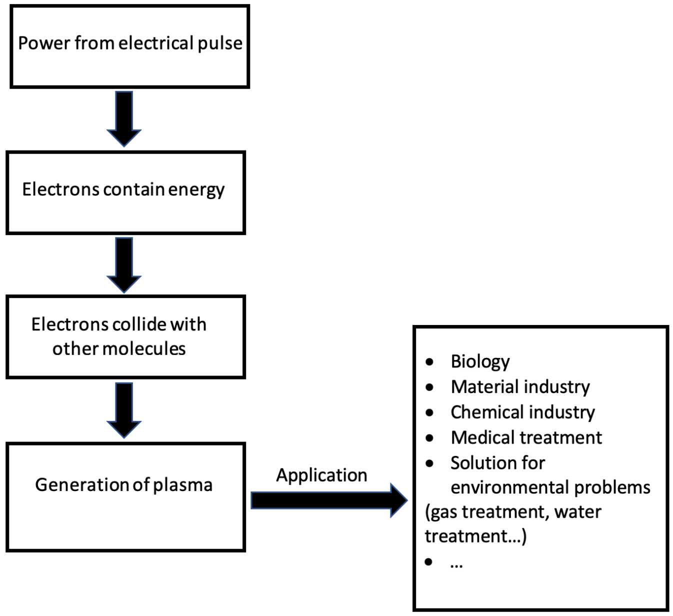


Fig. 10 The simplified process of conversing energy from the original pulsed power source to the final application.

All experiments of the research in this dissertation, pulsed streamer discharge in atmospheric pressure is mainly applied for evaluation. And the following Fig. 11 is the photo of the pulsed streamer discharge with generation of plasma.

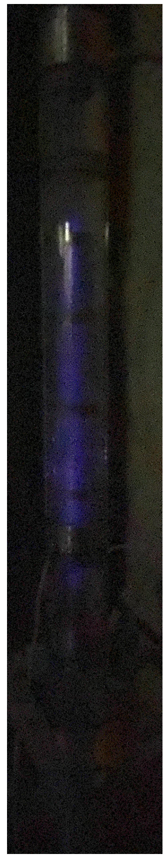


Fig. 11 Photo of pulsed streamer discharge in atmospheric pressure with generation of plasma.

1.4 INTRODUCTION DIFFERENT LINE OF PULSE GENERATOR

1.4.1 OVERVIEW OF PULSE GENERATOR

Until now, for many decades, there are a lot of research and development of pulse generators. Beside with improving the energy efficiency of pulsed power generator, there are plenty of studies and development of miniaturing size of pulse power generator or improving the performance of these pulsed power generators. Due to a lot of different application of these pulsed power generators, lots of models of pulsed power generator have been shown for many decades from huge size to compact size. For example, pulsed power generator named as "ETIGO-IV" which can be classified as huge size of pulsed power generator [48]. A. Tokuchi et al. [48] has introduced a new generation of pulsed power generator which can generates high-power electrical pulse with repetitive rate, this pulsed power generator has ability of generating electrical pulse with peak voltage and peak current of 400 kV and 13 kA, respectively. This generation- "ETIGO-IV" of pulsed power generator can be applied for many applications such as processing material, generating high-power microwave [48]. There are a lot of further studies about developing and improving this generation of pulsed power generator- "ETIGO-IV", as in [49] [50] [51].

About compact size of pulsed power generator, beside with developing huge size of repetitive pulsed power generators, there are also a lot of studies and development of compact size of repetitive pulsed power generators. As shown in [52], due to great development of semiconductor switch with thyristor such as Insulated-gate Bipolar Transistors (IGBTs) or Metal-Oxide-Semiconductor Field-Effect Transistors (MOS-FETs), a lot of generations of pulsed power generators with compact size have been being performed and developed, and as a result this development of pulsed power generator in miniatured size has ability of being applied in many other fields. These generation of pulsed power generator can be applied in many fields, such as modifying material for material industry, treating some environmental problems for maintaining safe environment or developing some treatment method in medical field [52]. Despite of having small size, these pulsed power generators can generate electrical pulse with impressive characteristic, W. Jiang et al. [52] pointed out that 11 kV-voltage, 5 kA-current, 100 ns-pulse-width, 6 kHz-frequency electrical pulse can be generated by pulsed power generator constructed by IGBTs. Or by applying pulsed power generator developed based on switch of MOS-FETs, 5 kV-voltage, 75 A-current, 240 ns-pulse-width, 2.1 MHz-frequency electrical pulse can be generated [52]. Some of typical pulsed power generator with compact size will be introduced as following.

1.4.2 SEMICONDUCTOR OPENING SWITCH (SOS)-PULSED POWER GENERATOR [53] [54]

For last few decades, a lot of researches of developing pulsed power generator using semiconductor switch have been conducted, as S.K.Lyubutin et al. [53] announced a concept of generating repetitive pulse based on Semiconductor Opening Switch (SOS). Based on their realization the formation of silicon affected by the Semiconductor Opening Switch (SOS) which has an approximately 60 kA/cm^2-current-density [53], this SOS was applied for developing as a method of inductive storage. The development of SOS in [53] led to a new technology of generating pulsed power by switching system that can realize repetitive electrical pulse with the parameter listed in Table. 4. The following Table. 4 shows the parameter of the electrical pulse, which was generated by the system based on the development of SOS; and all these information and value is collected from the study in [53].

Table. 4 The parameter of electrical pulse generated by the system applying SOS [53].

Parameter of the output of electrical pulse	Value
Voltage	from 10 kV to 1000 kV
Current	from 0.5 kA to 50 kA
Pulse width	from 10 ns to 100 ns
Repetitive rate	from 0.01 kHz to 10 kHz
Energy of pulse	from 0.1 J to 10 kJ
Average power of pulse	from 10 kW to 500 kW

With the rapidly expanded application of pulsed power technology for pulsed discharging in atmospheric pressure, the parameter of high-voltage electrical pulse was required more severely, for example the pulse width need to be shortened further [54]. Based on the concept of applying semiconductor opening switch for generating electrical pulse, T. Yokoo et al. [54] has done experiments for developing pulsed power generator with the technology - "Inductive Energy Storage (IES)". As the result from the experiments by T. Yokoo et al., the following parameter of high voltage pulse, which was generated by pulsed power generator applying SOS, can be confirmed. The following Table. 5 performs the parameters of electrical pulsed discharge with application of SOS, and all information was gathered from the study in [54].

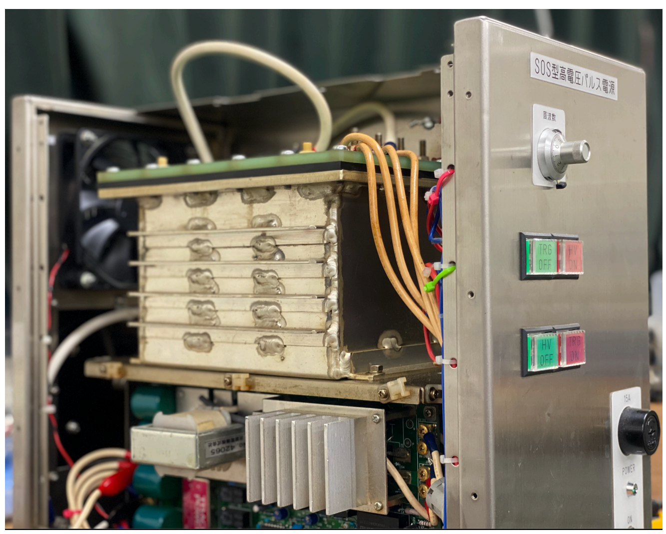
Table. 5 Parameter of the pulse outputted by the pulsed power generator applying SOS of the experimental results in [54].

Parameter of the output of electrical pulse	Value of the experimental result
Peak voltage	approximately 50 kV
Pulse width	approximately 10 ns
Repetitive rate	250 Hz

Recently, the research about application of the semiconductor opening switch diode for generate nanosecond-order electrical pulse has been being developed continuously, as study in [55], T. Sugai et al. has designed and confirmed the amplification of the circuit constructed by IES and SOS. From the experimental results in [55], an electric circuit constructed by two IES circuits and SOS diode was confirmed for the amplification by their synchronization, which has opened a new achieving of generation high-power electrical pulse. This new method of generating electrical pulse with high power is meaningful for improving the efficiency of generating non-thermal plasma, which has been well known for its application in plenty of fields, and high-power pulsed discharge in atmospheric pressure can be seen as one of the most effective ways for generating non-thermal plasma [55].

The following Fig. 12 is photograph of SOS pulse generator, which has been being applied for generating high-voltage pulse for many studies of pulsed power application.

(a)



(b)

Fig. 12 The photo of SOS pulse generator

(a) The front view of SOS

(b) The inside view of SOS generator

1.4.3 MARX GENERATOR

Marx generator has been well known in field of pulsed power technology, for a lot of decades there are lots of research on the application and development of this electrical pulse generating circuit [56] [57] [58] [59] [60] [61] [62] [63]. Marx generator has been known for its function of generating voltage electric pulse with a large range of pulse width from some microsecond to picosecond as in [62]. Marx generator can generate pulse with different waveform but due to its structure, Marx generator have some disadvantage in generating high-voltage pulse with short duration when compared with Linear Transformer Driver (LTD) system, which will be introduced in detail in the next part. And Marx generator has been performed as one of the most popular ways of conversing energy and applied in plenty of fields, such as X. Ren et al. [59] has researched on the development of Marx generator for improving energy efficiency of dielectric barrier discharge which was well known for wide application in a lot of fields based on pulsed power technology.

1.4.4 Linear Transformer Driver (LTD)

Linear Transformer Driver (LTD) has been developed for many decades, and it has been well known for its function of power adding and pulse shaping [2] [64] [65] [66] [67]. Based on technical function of power adding and pulse shaping, LTD generator has been applied to generate high-voltage electric pulse with complex pulse shape and parameter of output can be adjusted flexibly. Besides that, there are also some studies on improving performance of LTD generator and cost efficiency as Y. Feng has done in [68] [69].

The following Fig. 13 to Fig. 15 is the photo of 24-stack LTD generator, one module of LTD and the equivalent electric circuit of stacked LTD generator, respectively. In Fig. 15, the red line circle is the equivalent circuit of one module of LTD generator.

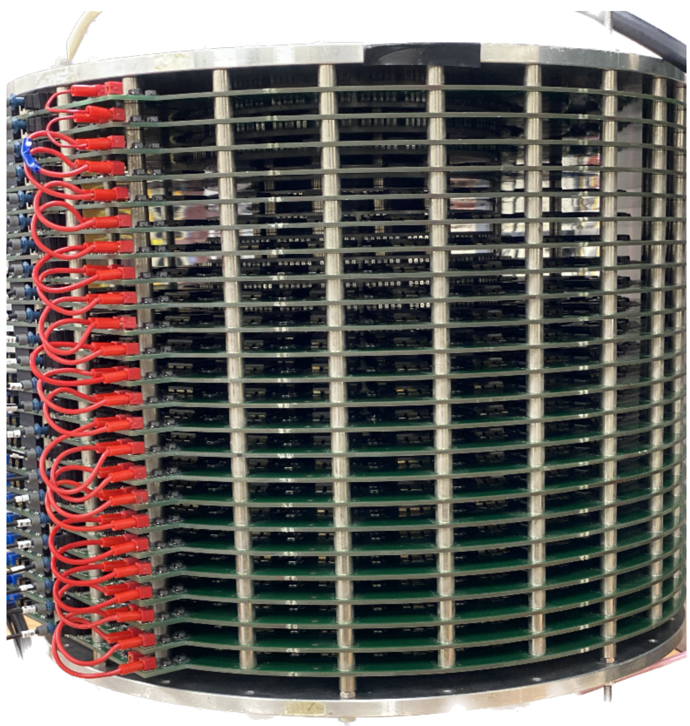


Fig. 13 Photo of 24-stack LTD system.

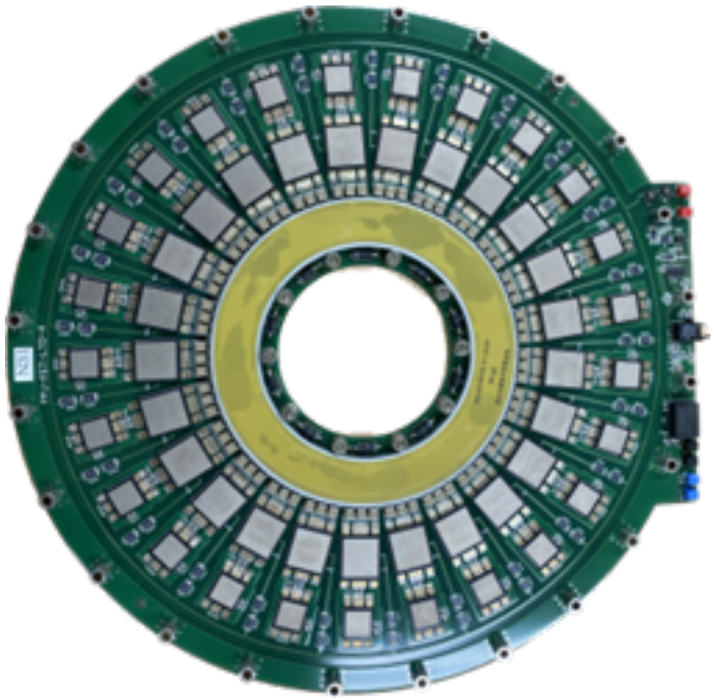


Fig. 14 Photo of one module of LTD generator.

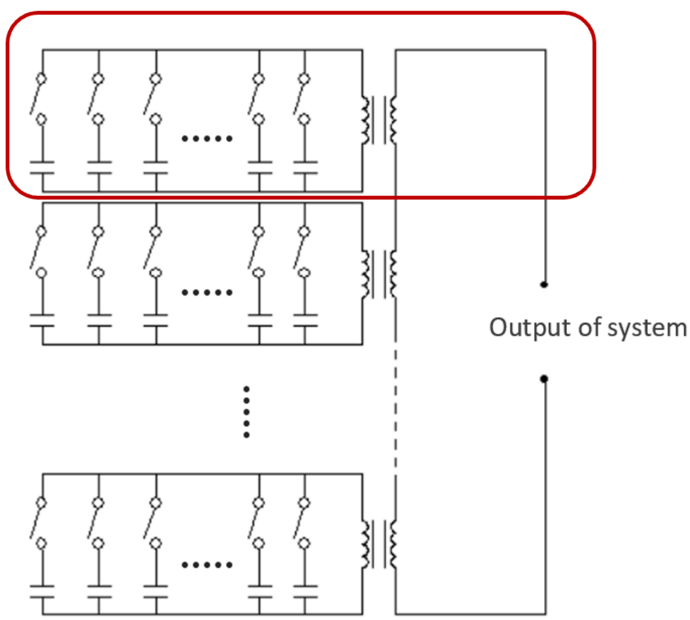


Fig. 15 Equivalent electric circuit of stacked LTD system.

Chapter II

EVALUATION OF OZONE GENERATION

2.1 INTRODUCTION OF RESEARCH

In this section, the main work of evaluation on ozone production in this study will be explained and analyzed [70]. Application of pulsed power technology on resolving a lot of environmental problem was studied and analyzed in a lot of works as [71] [72]. As we have known, ozone is one of the strongest oxidants, so ozone has been being applied in many fields for decomposing a lot of connection of harmful chemical which have significant effect on vary environment and cause plenty of environment problem, such as dye chemical which can be considered as harmful chemical for water environment. And ozone was confirmed as efficient way for decomposing the chemical connection of dye chemical (such as Indigo Carmine) as Y. Minamitani et al. have shown in [24]. And T. Sugai et al. [73] have performed experiments to evaluate the effect of pulse shape on decomposing Indigo Carmine but the direct evaluation of active species production has not been done. Therefore, in order to understand the effect of these pulse shape on production of ozone, a direct evaluation of the effect of pulse shape on production of ozone was done in this section of this study.

2.2 THE SETUP OF EXPERIMENT AND EXPERIMENTAL METHOD

In this part, method of evaluating ozone production by atmospheric pulsed discharge control will be introduced. The following Fig. 16 and Fig. 17 is the real photo of water treatment equipment and equivalent sketch of water treatment attached with Linear Transformer Driver (LTD) pulse generator system. And this LTD system has been used for all experiments in this study as main high- voltage pulse generator, which a lot of unique technical function such as power adding and pulse shaping, this information of LTD system has been introduced in the previous section. The LTD system was selected for these experiments due to its advantage of individual outputting energy; and all modules of LTD system can be connected with ground so this LTD system can generate large output in shorter time compared with other generator as shown in [67]. Therefore, LTD system is more suitable for generation high-voltage pulse with shorter duration [67]. The Fig. 18 shows the concept of pulse shaping, which can be generated by this LTD system.

In the water treatment equipment, the electrodes were constructed by the stainless-steel wire and stainless-steel mesh. The diameter of the stainless-steel wire and curled up stainless-steel mesh is 0.28 mm and 38 mm, respectively. For the connection between these electrodes in this water treatment equipment and the LTD system, a coaxial cable was used. As in Fig. 17, the wire electrode was connected to the positive output of the LTD system and the mesh electrode was connected to the ground, this setup is as performed in [73] [70].

In order to measure the concentration of produced ozone, air has been pumped outside to the water treatment equipment with the flow rate of ~16.5 L/min, and a UV Ozone monitor

(HARE, model 620) was applied to catch the produced zone concentration for evaluation as in Fig. 19.

Fig. 16 Photograph of water treatment equipment in system.

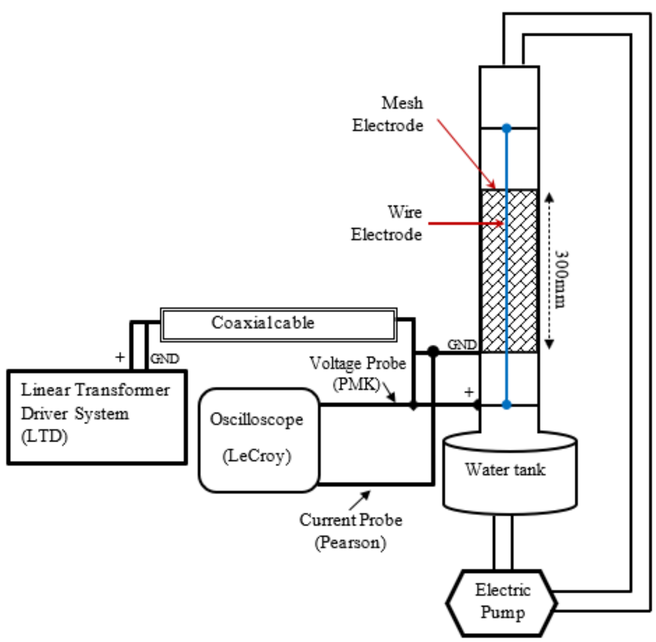


Fig. 17 Equivalent sketch of water treatment system in total.

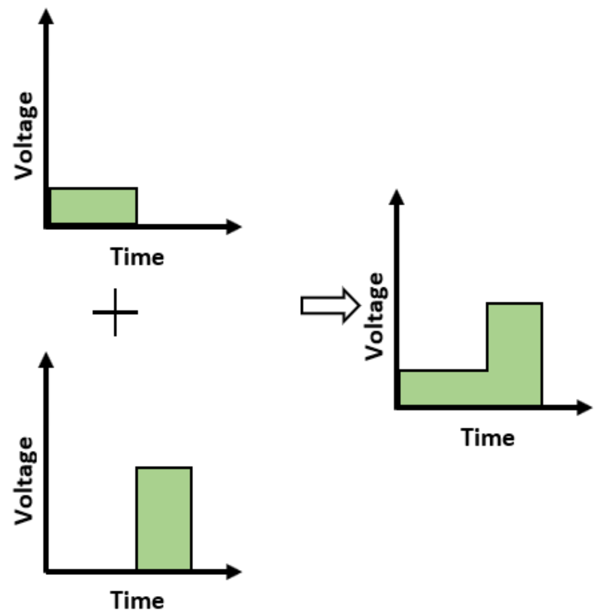


Fig. 18 Concept of pulse shaping function of LTD system.

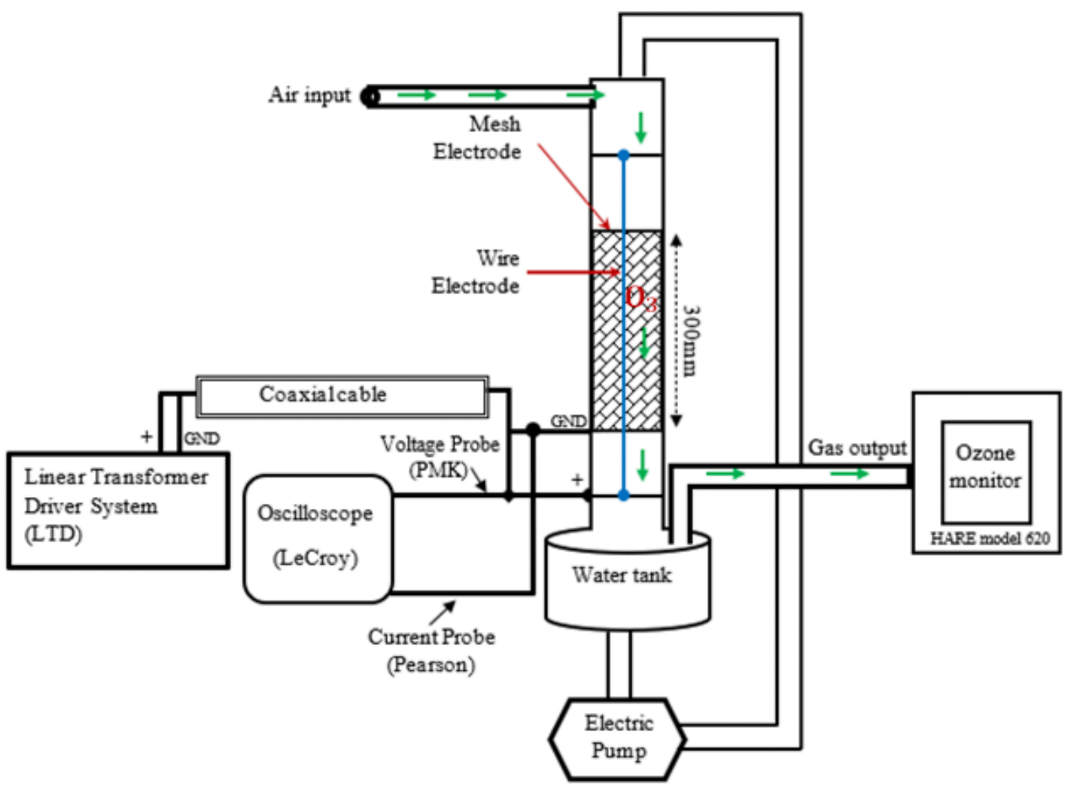


Fig. 19 Sketch of the experimental setup for evaluating ozone production in water treatment system.

2.3 EVALUATION OF OZONE GENERATION BY LOWERED VOLTAGE AT THE SECOND PART OF PULSE

In this part, the result of experiment that evaluating the effect of pulse discharge control on ozone production. There are a lot of studies for this effect on ozone production as in [74][75][76][77][78], but in these studies the average total input energy of pulse may be different due to different pulse width or the consideration for the electric field at the last part of pulsed discharge was not performed. Therefore, in this study, the average total input energy will be maintained as unchanged for all experiment and the effect of electric field at the last part was focused for discussion of ozone generation. Based on technical function of LTD system the pulse with lower voltage at the second part can be generated and these pulse shapes have been applied for evaluating production of ozone. The concept of these pulse shape will be shown is the following Fig. 20 to Fig. 23, in which the voltage of the second half is lowered in range of 1~0 when compared with the voltage of the first half.

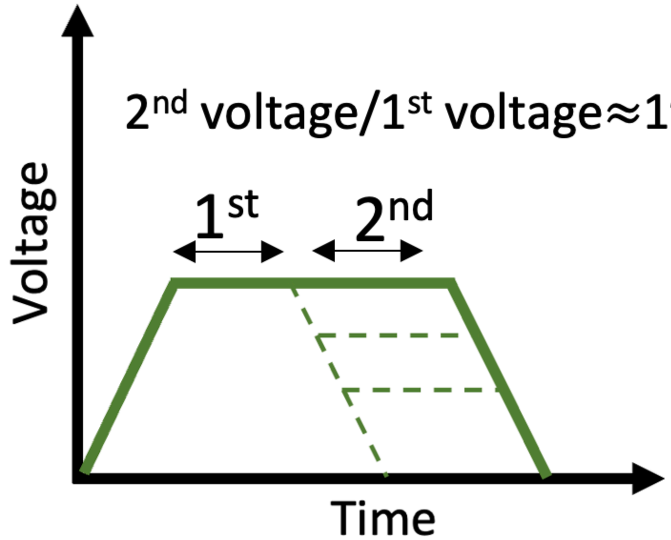


Fig. 20 Concept of pulse shape with lowered voltage at the second half.

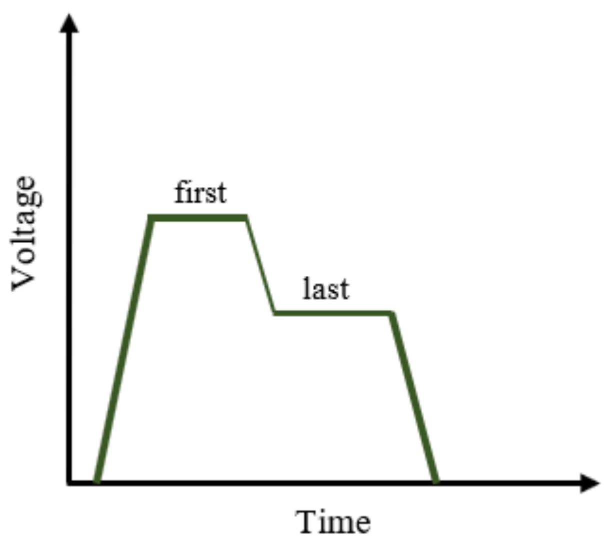


Fig. 21 Concept of pulse shape with lowered voltage at the second half (ratio of last to first is ~2/3)

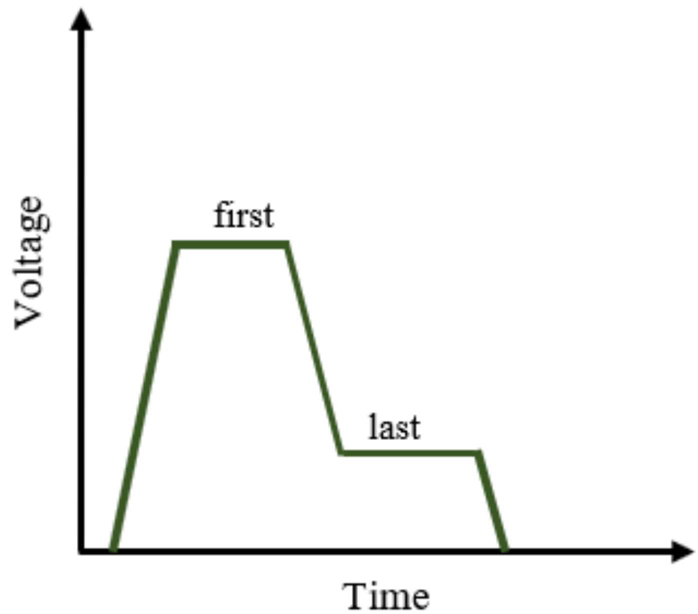


Fig. 22 Concept of pulse shape with lowered voltage at the second half (ratio of last to first is ~1/3)

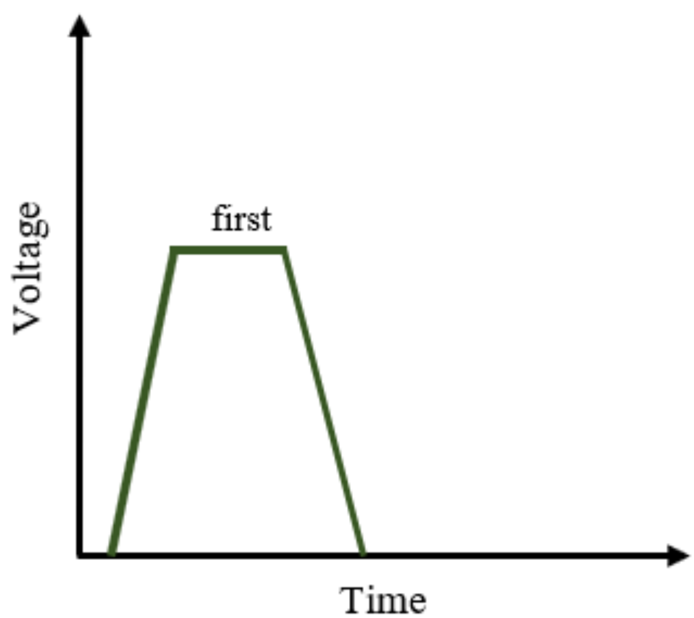


Fig. 23 Concept of pulse shape with lowered voltage at the second half (ratio of last to first is 0)

2.3.1 MEASUREMENT OF THE OUTPUT VOLTAGE AND CURRENT

The following Fig. 24 to Fig. 27 is the result of the measurement voltage and current applied to the electrodes by high-voltage probe (PMK, PHV4002-3) and current probe (Pearson, Model 110), respectively. Here, the measured value of current is considered as total of discharge current and displacement current. The displacement current can be calculated from the measurement of voltage and capacitance of electrodes. When the pulsed discharge has still not happened between electrodes yet, value of capacitance can be obtained approximately by fitting method. With value of displacement current, the discharge current of pulse can be calculated.

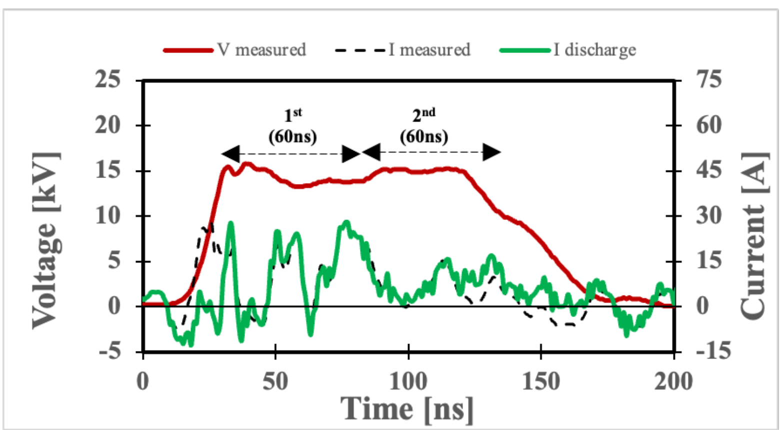


Fig. 24 Result of measurement the output with pulse shape of lowered voltage at the second half (ratio of last to first is 1) marked as waveform (a).

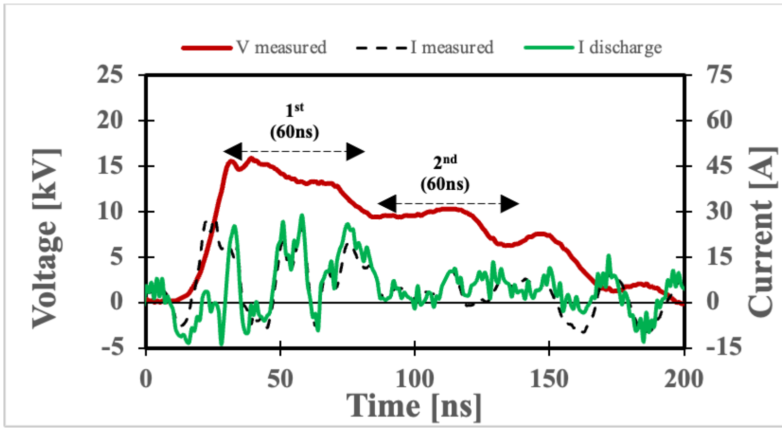


Fig. 25 Result of measurement the output with pulse shape of lowered voltage at the second half (ratio of last to first is ~2/3) marked as waveform (b).

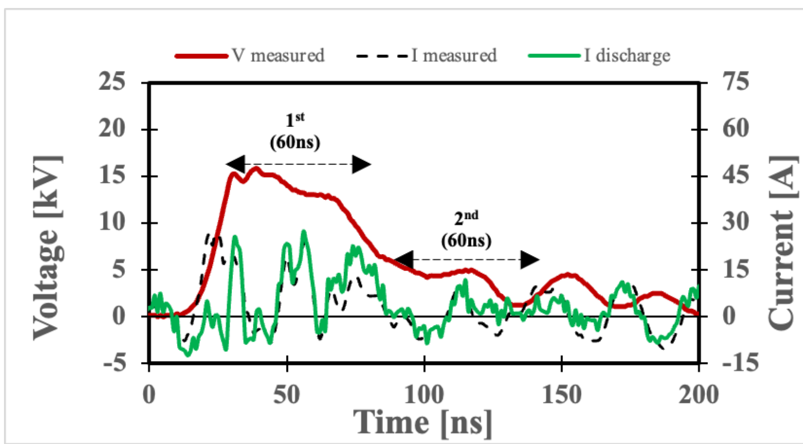


Fig. 26 Result of measurement the output with pulse shape of lowered voltage at the second half (ratio of last to first is ~1/3) marked as waveform (c).

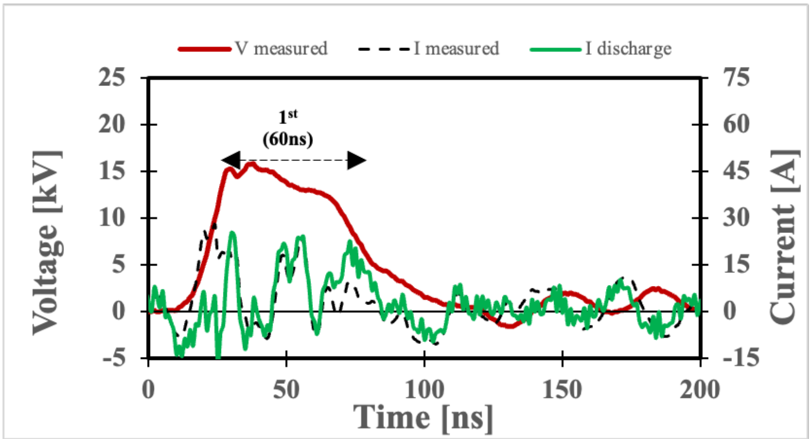


Fig. 27 Result of measurement the output with pulse shape of lowered voltage at the second half (ratio of last to first is 0/1) marked as waveform (d).

In order to maintain the total average input energy of pulse as unchanged, the repetition rate of each case was adjusted. The following Table. 6 and Table. 7 shows the method of generating high-voltage pulse with these pulse shape in which number of ON-LTD modules is different for each case, and the average input power of each case, respectively [70].

Table. 6 Number of ON-LTD modules for each part of pulse of different waveform.

Waveform	Number of LTD modules are switched ON	
	First part	Second part
(a)	23	23
(b)	23	15
(c)	23	7
(d)	23	0

Table. 7 Parameter and calculated result of each waveform.

Waveform	Repetition rate [Hz]	Discharge energy of each pulse [mJ]	Average input power [J/s]
(a)	50	15.6	0.78
(b)	78	9.95	0.78
(c)	118	6.57	0.78
(d)	138	5.6	0.77

2.3.2 EVALUATION OF OZONE PRODUCTION

Here, the result of evaluating ozone production by applying each waveform is shown. The following Fig. 28 to Fig. 29 shows the concentration of produced ozone and energy efficiency of ozone producing. Here, mass of ozone produced by a unit of energy was analyzed for evaluating energy efficiency. About this calculation, by multiplying measured concentration of ozone with air flow rate (~16.5 L/min), volume or mass of ozone produced per a unit of time can be obtained. Then total energy of pulse output per a unit of time can be calculated by multiplying energy of pulse with frequency of pulse. Finally, mass of ozone produced per a unit of time divided by total energy of pulse output per a unit of time is the mass of ozone produced by a unit of energy from pulsed discharge (yield of ozone production).

As the result, when the voltage at the second part of pulse is lowered from waveform (a) to waveform (d), the concentration of ozone produced in water treatment equipment and caught by ozone monitor is increased clearly. The energy efficiency of ozone producing is also increased from waveform (a) to waveform (d), while the total average input energy for each waveform from (a) to (d) is maintained as unchanged.

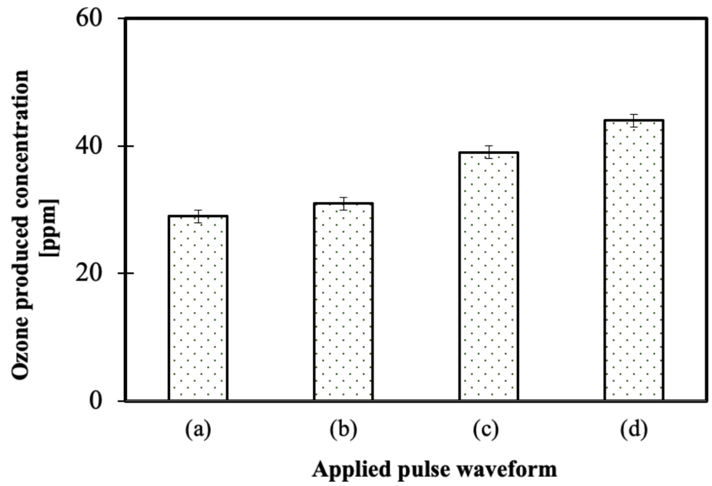


Fig. 28 Result of measurement the ozone concentration with applying waveform from (a) to (d).

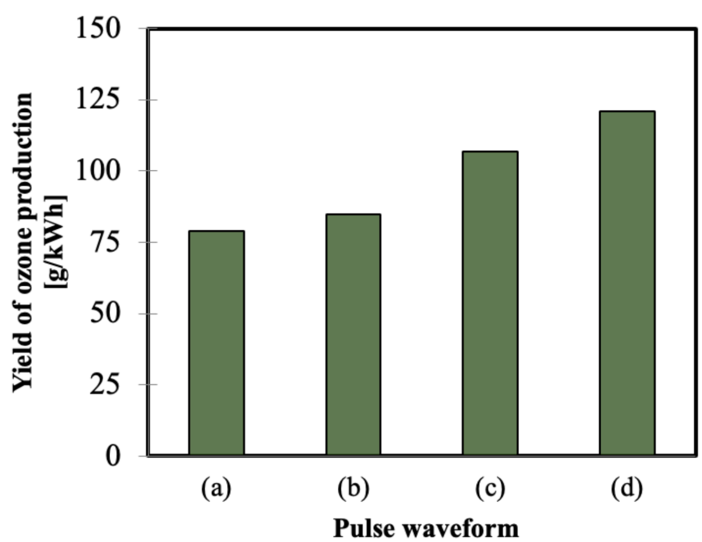


Fig. 29 Calculated result of ozone producing efficiency with applying waveform from (a) to (d).

2.3.3 DISCUSSION OF THE EXPERIMENT RESULT

2.3.3.1 Discussion the effect of electric field

In the following, the discussion of the above experiment result will be introduced and analyzed.

As in the following Fig. 30, when the high voltage is applied to the positive electrode, here is the wire electrode for this experiment. The primary streamer discharge will be generated, and the streamer head will move to the grounded electrode, which is mesh electrode in this experiment equipment. At the time primary streamer head has already gone to ground, the phenomenon of neutralization of ion in this area will appear which led to the decrease of electric field [79] [80]. Therefore, at this time the production of ozone may become more difficult, and it can be considered as a physical explanation for the reason that lead to decreases in energy efficiency in total.

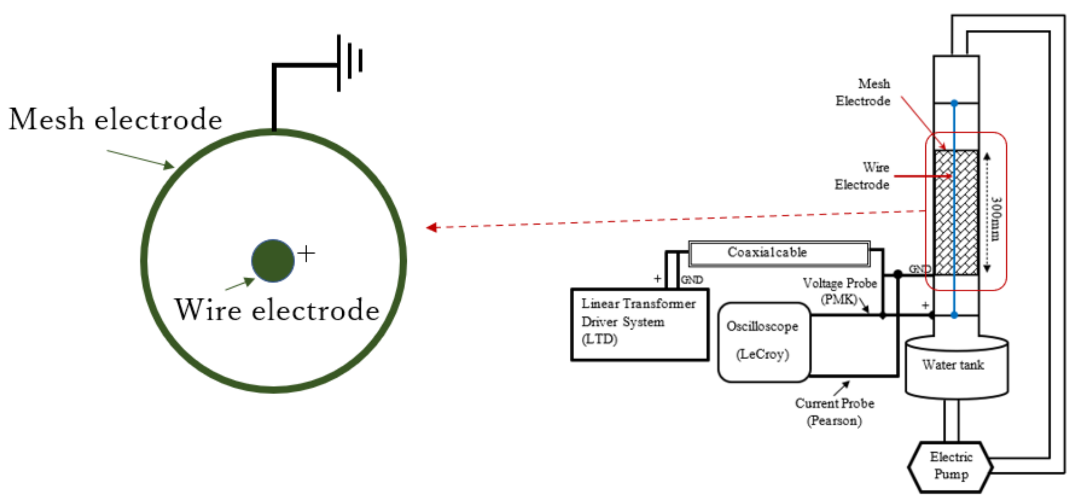


Fig. 30 Enlarged structure of the electrode in the water treatment equipment.

2.3.3.2 Discussion the effect of temperature

The production of ozone is known for being affected by temperature; with higher temperature the production of ozone will be decreased [81]. Therefore, the increase of temperature by pulsed discharge can be considered as reason for the decrease of ozone production. Despite of being classified as non-thermal plasma generation, streamer discharge may increase a bit in the temperature, so the following analyzation is performed to evaluate change in temperature by pulsed discharge under condition of atmospheric pressure and room temperature.

At first, the kinetic energy can be performed as $\frac{1}{2}m\overline{v^2}$, and the following relation equation is available [82],

$$\frac{1}{2}m\overline{v^2} = \frac{3}{2}kT \qquad (1)$$

Here, T is the room temperature (~300 K) and $k = 1.38 \times 10^{-23}$J/K is the Boltzmann constant. With the amount of total molecules in the discharge area-between electrodes, the total kinetic energy of air molecules can be calculated. In the discharge place of the system applied in this study, the total air molecules are approximately 9.11×10^{21} so the total kinetic energy of air here is ≈ 56.6J.

For the discharge energy of 15.6mJ at repetitive rate of 50Hz we have the following analyzation. With the repetitive rate of 50Hz and air flow rate of 16.5 L/min, the speed of air flow through the discharge area will be approximately 0.22m/s [70]. It means that gas need 1.4s to flow from the top to bottom of the discharge area with 300mm-length, and with 50Hz-repetition rate the pulsed discharge will collide air approximately 68 times during this duration.

If the discharge energy is assumed as the kinetic energy, from (1) the following relation can be considered.

$$\frac{15.6 \times 10^{-3}}{9.11 \times 10^{21}} \times 68 + \frac{56.6}{9.11 \times 10^{21}} = \frac{3}{2} kT$$

$$\Rightarrow T \approx 306\text{K}$$

Herer, it may be considered that the temperature is raised a little bit than the room temperature of 300K and may affect the production of ozone as mentioned above. However, the different of this analyzed temperature between each pulse waveform applied in these experiments is very small due to the approximately unchanged input energy of each waveform.

2.3.3.3 Analyzation the reduced electric field

The reduced electric field in the discharge area is analyzed in this part. Fig. 31 shows the cross section of the mesh electrode and wire electrode and their position. The r_0 is radius of the cross section of mesh electrode, which is 19mm. And a is 0.14mm, which is radius of the cross section of wire electrode. Then relation between voltage, charge and electric field in cylinder is performed as following (2)(3) referred [83],

$$V = \frac{Q}{2\pi\varepsilon_0} ln\frac{r_0}{a} \tag{2}$$

$$E = \frac{Q}{2\pi\varepsilon_0 r} = \frac{V}{r \times ln\frac{r_0}{a}} \tag{3}$$

From equation (3), the electric field can be calculated and performed in Fig. 32, and by being divided by density of air in the discharge area the reduced electric field can be obtained. Fig. 33 shows the calculated reduced electric field relating with position of r between wire electrode and mesh electrode. When the value of r is expanded to the mesh electrode, the electric field and reduced electric field decreases clearly.

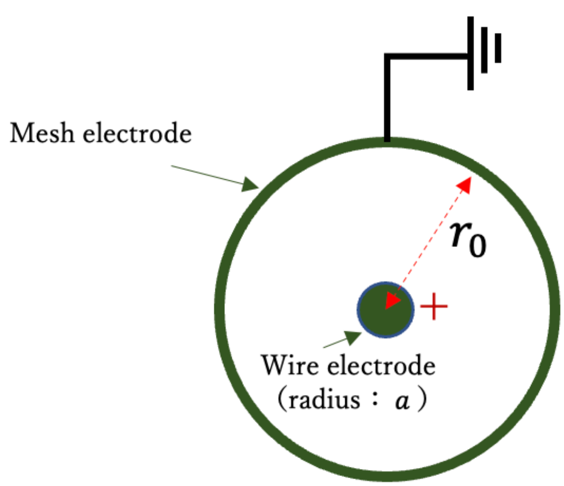


Fig. 31 Cross section view of electrodes in the discharge area.

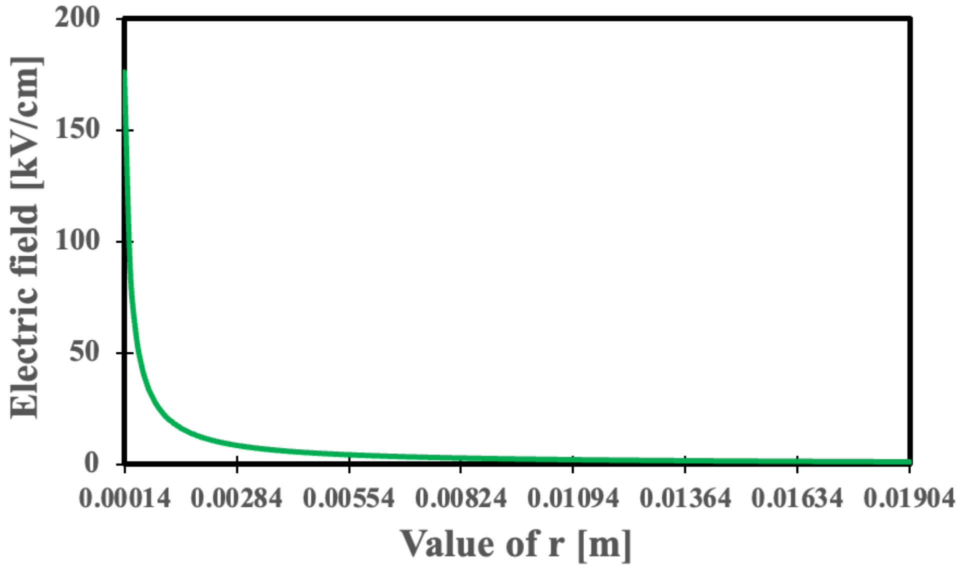


Fig. 32 Value of electric field related with each position between electrodes.

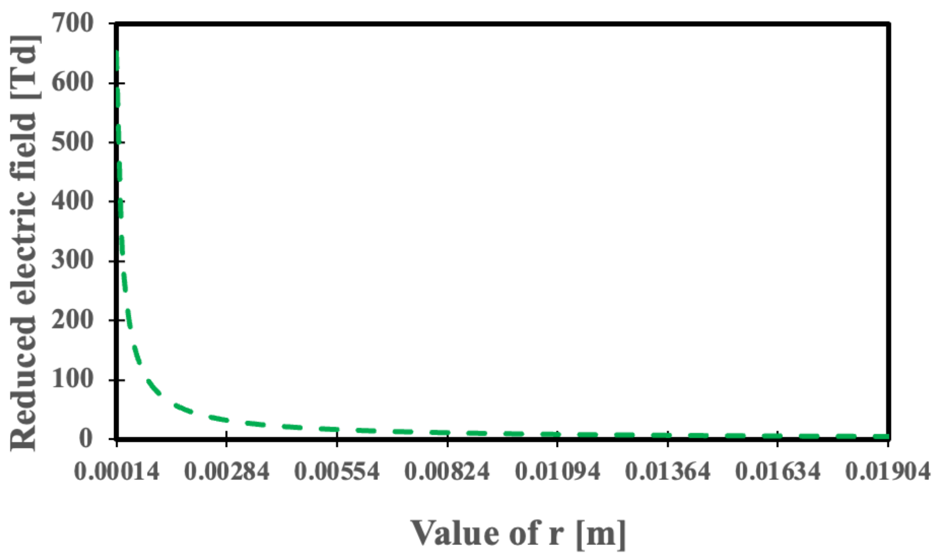


Fig. 33 Value of reduced electric field of each position from wire electrode to mesh electrode.

About the effect of reduced electric field on production of ozone, the following analyzation can be considered. S. Pekarek [84] showed that the energy of electron is decreased when the reduced electric field is decreased. The energy of electron is the main factor affect to the production of ozone, due to the following reaction (4)(5), in which electron plays main role in separating oxygen for the next reaction of ozone production.

$$O_2 + e \rightarrow 2O\cdot + e \quad (4)$$

$$O_2 + O\cdot + M \rightarrow O_3 + M \quad (5)$$

Then A. T. Bell et al. [85] showed the relation between reaction rate constant of (4) and rate of electric field and pressure. With decrease of the rate of electric field and pressure, the reaction rate constant is decreased, which can be considered for the decrease of ozone production in final. Here, the rate of electric field and pressure have close relation with reduced electric field, and they can be converted together, so from this point it can be conclude that the reduced electric field has effect on the production of ozone in total. Furthermore, study in [86] has shown relation between the reduced electric field and coefficient of ionization, in which with the decrease of reduced electric field the coefficient of ionization is decreased, and this coefficient has relation with the flow of discharge current. Finally, from their studies, it can be concluded that the reduced electric field has effect on production of ozone.

2.4 RESULTS OF EVALUATING OZONE GENERATION BY NANOSECOND DOUBLE-PULSE WITH VARIED PULSE INTERVAL

In this section the evaluation of the effect of pulse interval on production of ozone in water treatment equipment will be shown and analyzed as [70].

2.4.1 MEASUREMENT OF THE OUTPUT VOLTAGE AND CURRENT

The following Fig. 34 shows the concept of applied pulse waveform for the experiment in this part. Here, the interval between pulse will be 1, 2, 5, 10, 50, 100, 500, 1000 μs, and 10 ms. And Fig. 35 to Fig. 37 shows the measured output of the voltage and current with varied pulse interval from 1 μs to 5 μs, respectively. And the Table. 8 shows the calculated energy of each pulse. As in Table. 8 with shorter pulse interval, the difference in discharge energy of the first pulse and second pulse is so far clearly. In order to explain for this phenomenon of high voltage pulse control, as shown in [83][87], M. R. Kazemi et al. has mentioned that the electric field may be decreased by phenomenon of ionization when the primary voltage pulse is applied to the electrode.

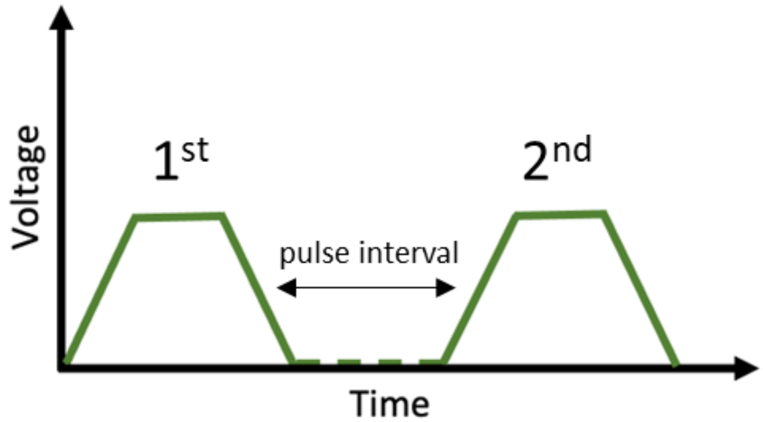


Fig. 34 Concept of double-pulse waveform applied for evaluation of ozone production.

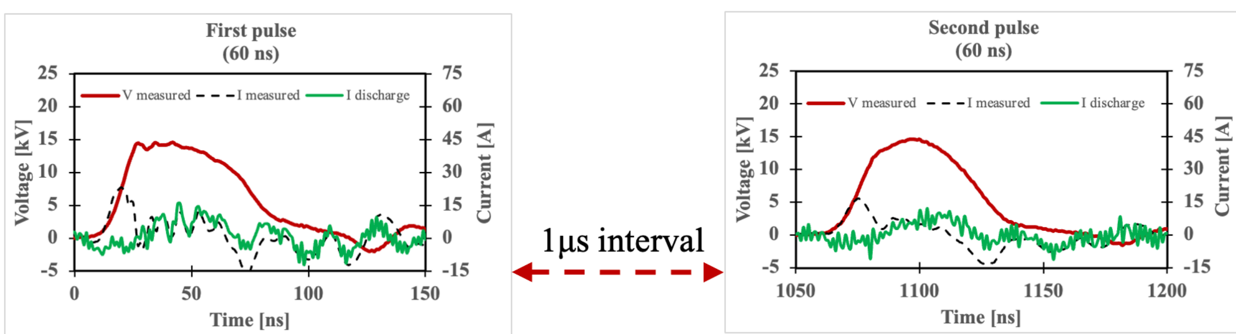


Fig. 35 Output voltage and current of pulses with 1 μs pulse interval.

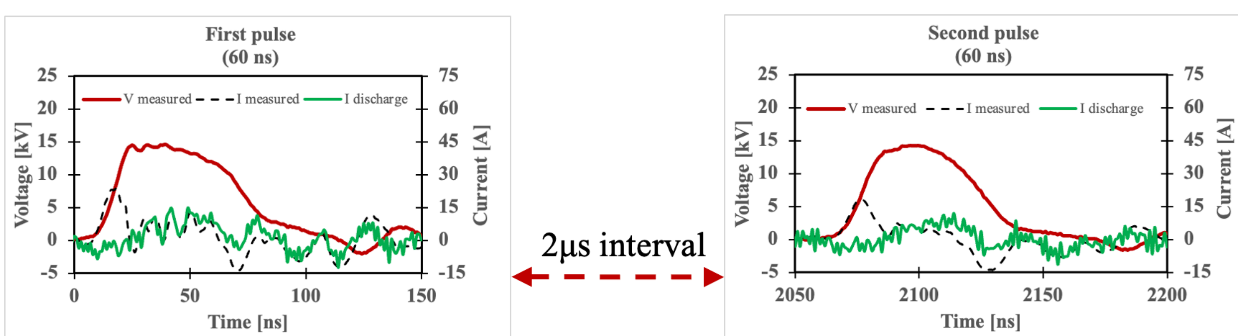


Fig. 36 Output voltage and current of pulses with 2 μs pulse interval.

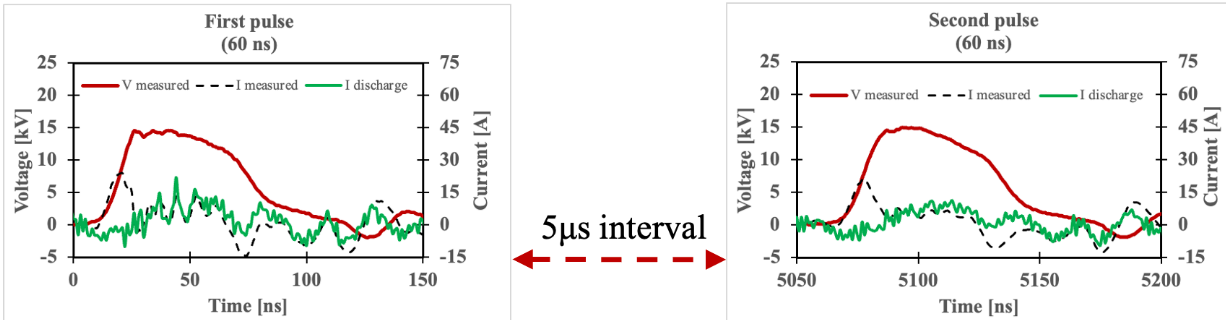


Fig. 37 Output voltage and current of pulses with 5 μs pulse interval.

Table. 8 Calculated result of discharge energy of each pulse in double-pulse waveform.

Pulse interval	Discharge energy of the first pulse with 60 ns pulse width [mJ]	Discharge energy of the second pulse with 60 ns pulse width [mJ]
1μs	3.44	2.29
2μs	3.76	2.17
5μs	3.73	3.34
10μs	4.01	3.43
50μs	4.01	3.68
100μs	3.95	3.54
500μs	4.02	3.94
1000μs	4.00	3.99
10ms	4.01	4.01

2.4.2 EVALUATION OF OZONE PRODUCTION

The following Fig. 38 and Fig. 39 is the experiment result of measuring ozone production by applying double-pulse waveform to evaluate the effect of pulse interval between pulse on the production of ozone.

As the results from the Fig. 38 and Fig. 39, with extended pulse interval from 1 μs to 10 ms, the change in ozone production can be observed clearly. In detail, with pulse interval from 1 μs to 50 μs, the longer pulse interval will release higher concentration of ozone and energy efficiency, and when the pulse interval is extended from 50 μs to 10 ms, the change in ozone production is very small. The explanation for this phenomenon will be described more in detail in the discussion part of this section.

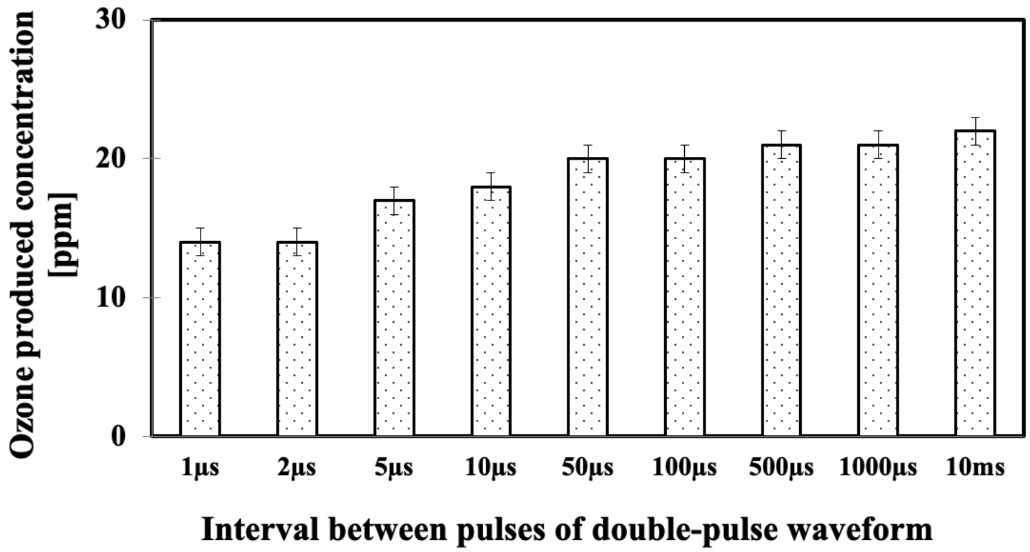


Fig. 38 The concentration of ozone produced with different pulse interval applied to the electrodes.

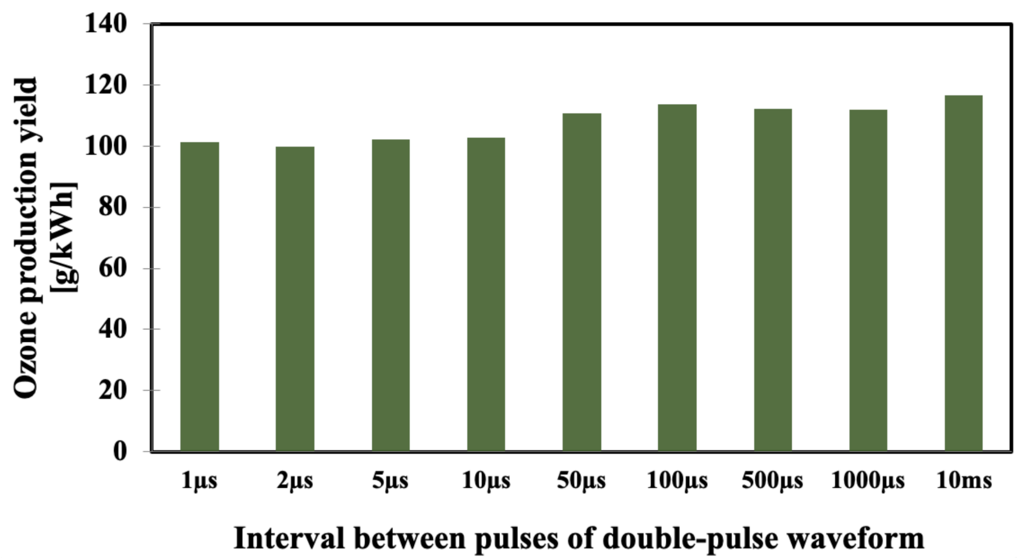


Fig. 39 The energy efficiency for production of ozone with different pulse interval applied to the electrodes.

2.4.3 DISCUSSION OF THE EXPERIMENT RESULT

In this part, the discussion and explanation for the above experiment result will be shown. As we have known, beside with ozone production reaction, there are a lot of ozone decomposing reaction as following [88],

$$O\cdot + O_3 \rightarrow O_2 + O_2 \qquad (6)$$

$$NO + O_3 \rightarrow NO_2 + O_2 \qquad (7)$$

$$NO_2 + O_3 \rightarrow NO_3 + O_2 \qquad (8)$$

And we have the reaction rate of ozone production reaction (9) can be performed as following (10),

$$O\cdot + O_2 + M \rightarrow O_3 + M \qquad (9)$$

$$\frac{d\,[O_3]}{dt} = k[O\cdot][O_2][N_2] \qquad (10)$$

here, $k = 8.7 \times 10^{-35} cm^3/s\ /particle$ is the reaction rate constant for the (9) with T=300 K temperature condition and M is nitrogen, this information is referred from [89].

In order to simplify this calculation, the change in concentration of ozone according to reaction time ($\frac{d\,[O_3]}{dt}$) will be considered as the reverse order of the change of O radical, and concentration of oxygen and nitrogen is as constant [70]. In the air, concentration of oxygen and nitrogen can be considered as 21% and 78%, respectively, and this information was referred from [90].

As the above setup for this calculation, needed time that O radical react with oxygen to produce ozone can be calculated by the following equation,

$$t = \frac{ln\,[O\cdot]_0 - ln[O\cdot]}{k'} \qquad (11)$$

Here, $k' = k[O_2][N_2]$

The following Fig. 40 is the result of above calculation of O radical reaction time. In other words, this result can be considered as the lifetime of O radical due to amount of produced O radical is set as only contributing to ozone production reaction.

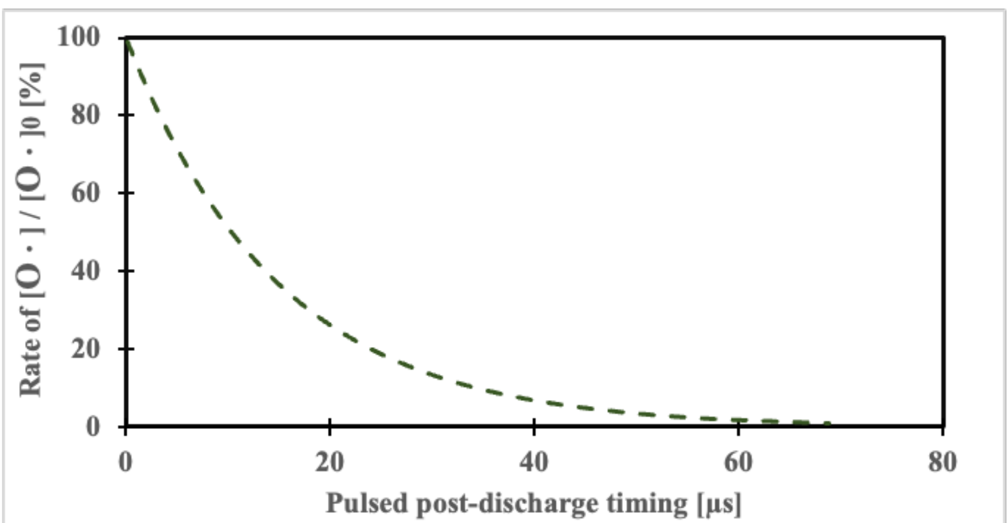


Fig. 40 The change in O radical concentration according with time after pulsed discharge applied.

From this result, it can be concluded that after about 50 μs the most of amount of O radical may disappear, and this result meet with the result of Fig. 38 and Fig. 39, in which with pulse interval longer than 50 μs the change in produced ozone concentration is small. In final, there are still some reactions related with O radical as following reaction [91], so the calculation in this part can be considered as one of the explainable reasons for change of ozone produced concentration.

$$O + NO + M \rightarrow NO_2 + M \quad (12)$$

$$O + NO_2 \rightarrow O_2 + NO \quad (13)$$

$$N + NO \rightarrow N_2 + O \quad (14)$$

Chapter III

EVALUATION OF OH RADICAL GENERATION

3.1 INTRODUCTION OF RESEARCH

In this section, the production of OH radical by applying a lot of different pulse shape will be shown. In detail, the effect of pulse with lower voltage at the behind part on OH radical production and the production of only the behind part of pulse is performed and analyzed [92].

Until now, there are a lot of studies about the production of OH radical by pulsed discharge [74] [93] [94], but as with the above evaluation of ozone production by pulsed discharge, the average input energy in these studies is not maintained due to different condition of pulsed discharge. Additionally, T. Sugai et al. in [73] performed experiment on evaluating this pulse shape on decomposing chemical connection-benzen ring of Indigo Carmine, but a direct evaluation on OH radical production had not been done yet. Therefore, in this part of this study the direct evaluation on OH radical production is performed with this pulse shape of lower voltage at the behind part of pulse with maintained as unchanged input energy for all cases and the OH production of only the last half phase of pulse is shown in addition.

3.2 THE SETUP OF EXPERIMENT AND EXPERIMENTAL METHOD FOR EVALUATING OH RADICAL PRODUCTION

The following Fig. 41 and Fig. 42 is the equivalent graph for the setup of water treatment system for evaluating OH radical production and the generated plasma caught in the discharge area. Next, OH radical evaluating method is shown in Fig. 43, based on unique characteristic of Terephthalic acid (TA) and 2-hydroxyterephthalic acid (HTA) [95] [96], fluorescence monitoring method [97] and Disodium Terephthalate (C8H4Na2O4) was applied to evaluate OH production in this water treatment system.

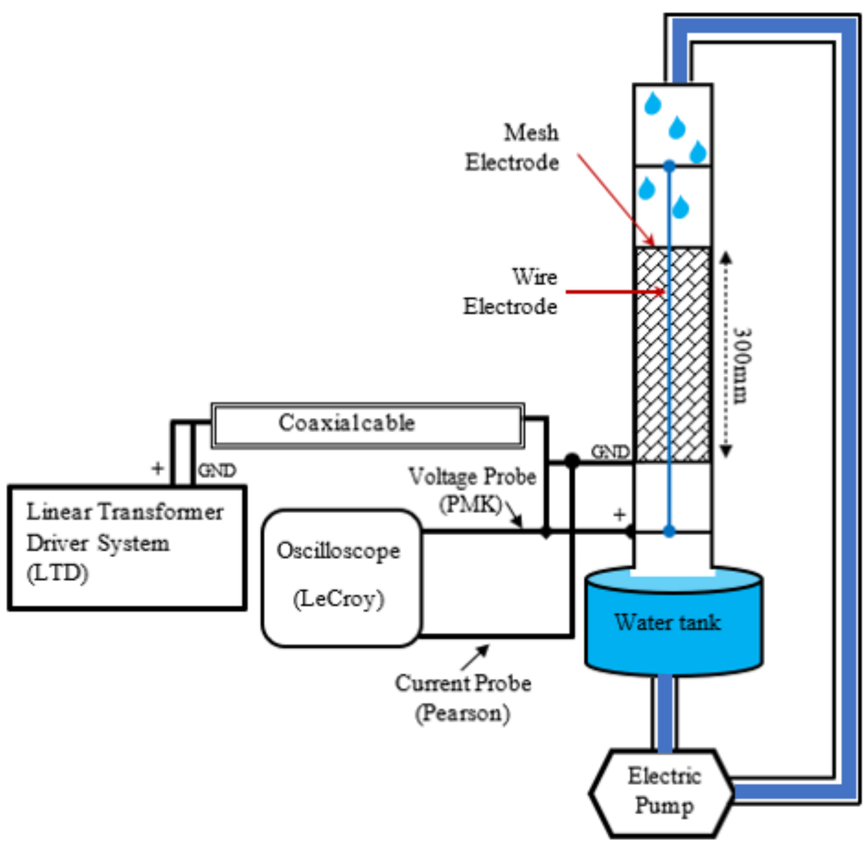


Fig. 41 Equivalent graph of water treatment equipment connected with pulse generator and measure equipment.

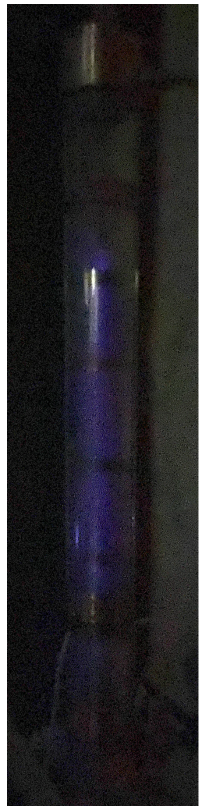


Fig. 42 Image of atmospheric pressure plasma caught in the discharge area.

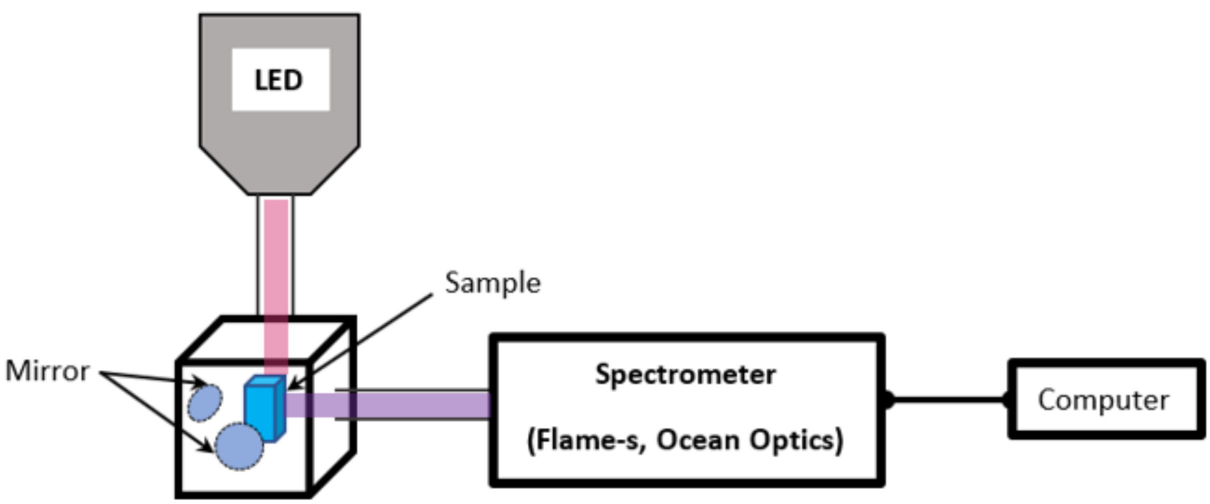


Fig. 43 Setup for OH radical evaluating experiment by fluorescence monitoring method.

3.3 EVALUATION OF OH RADICAL GENERATION BY WAVEFORM WITH LOWERING VOLTAGE AT THE SECOND PART OF PULSE

3.3.1 MEASUREMENT OF THE OUTPUT VOLTAGE AND CURRENT

In this part, the pulse control of suppressing voltage at the second part of pulse discharge was applied to evaluate production of OH radical in water treatment system. The Fig. 44 shows the concept of four different voltage at the second part of pulse. Then, the waveform in Fig. 45 to Fig. 48 is the measured results of the voltage waveform and current waveform, which was caught at the electrodes of the water treatment system. The streamer head of streamer discharge can spread from the positive electrode to ground with 0.8~1.2-mm/ns velocity, which was performed by D. Wang et al. in [98]. Therefore, the duration of 55 ns was selected for the first and second part of pulse to ensure that the primary streamer head had already reached grounded electrode with the gap of 19 mm between the positive electrode and grounded electrode.

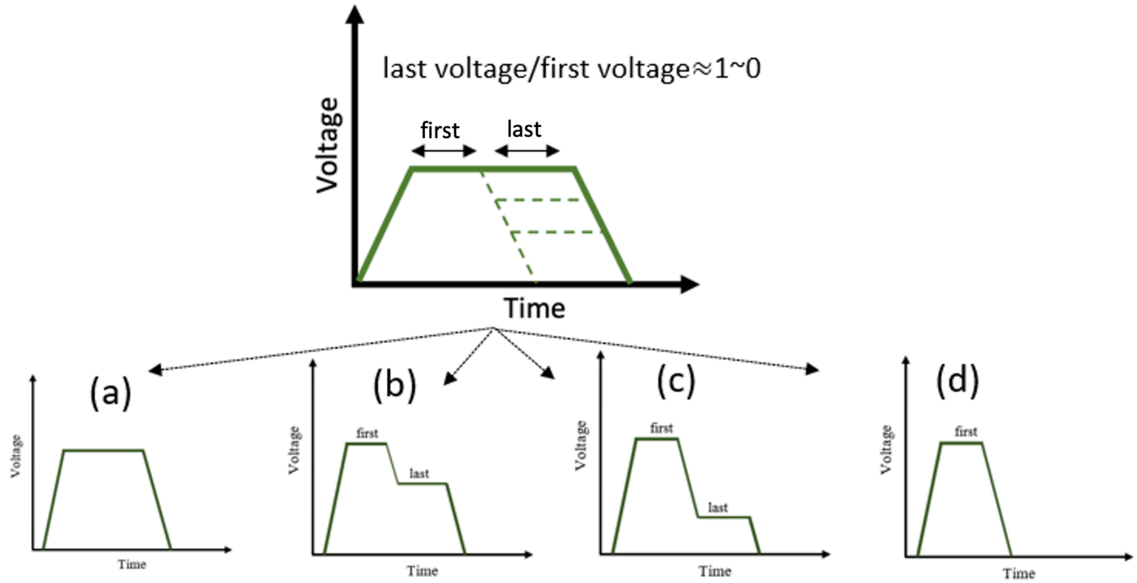


Fig. 44 Concept of applied pulse shape marked from (a) to (d) for evaluating OH radical production.

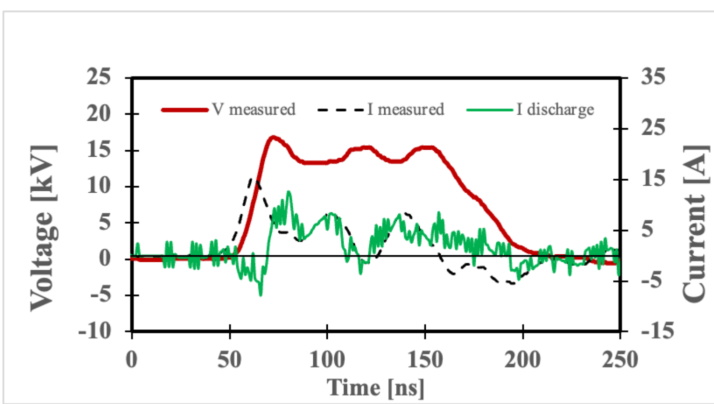


Fig. 45 Measured and calculated results of voltage and current waveform marked as waveform (a).

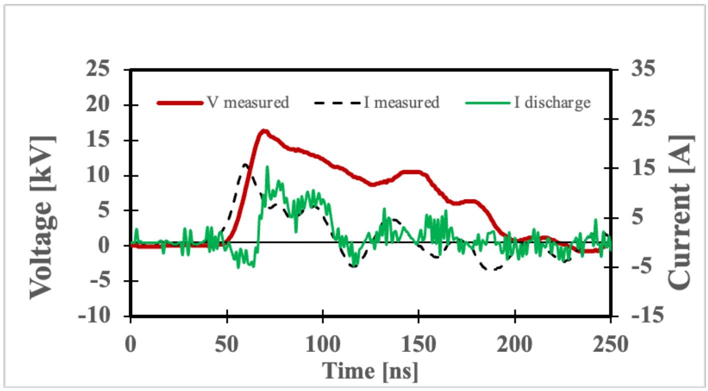


Fig. 46 Measured and calculated results of voltage and current waveform marked as waveform (b).

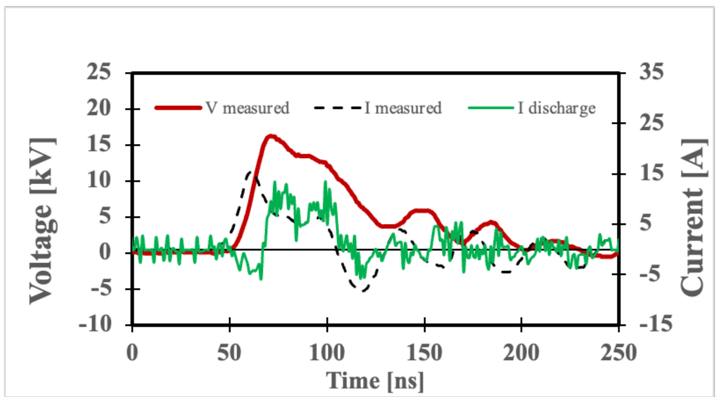


Fig. 47 Measured and calculated results of voltage and current waveform marked as waveform (c).

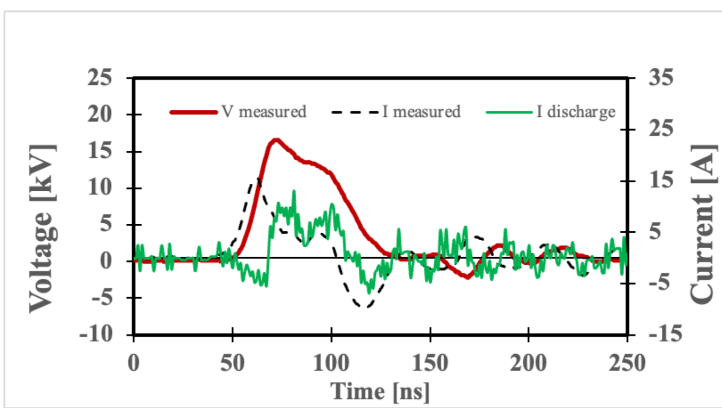


Fig. 48 Measured and calculated results of voltage and current waveform marked as waveform (d).

Following value in Table. 9 is the information of pulse and the calculated average input energy for one unit of time of each case. As we can see in this table, due to flexibly adjustable repetition rate by LTD system, the calculated result of average electrical power in the final column is maintained at about 0.5 J/s despite of different discharge energy of each waveform.

Table. 9 The information of each pulse with waveform (a) to (d) applied for evaluation.

Waveform	Repetition rate applied to each waveform [Hz]	Discharge energy of pulse [mJ]	Average input energy per unit of time [J/s]
(a)	78	6.38	0.5
(b)	106	4.71	0.5
(c)	131	3.79	0.5
(d)	166	3.0	0.5

3.3.2 RESULTS OF EVALUATING OH RADICAL PRODUCED IN WATER TREATMENT SYSTEM

The following Fig. 49 and Fig. 50 is the result of evaluating OH radical production and energy efficiency (when the sample solution had been treated for total 10 minutes), respectively. As in these figures, at the same average input energy, the lowest voltage of the second part-waveform (d) has significant effect on improving OH production and energy efficiency in total.

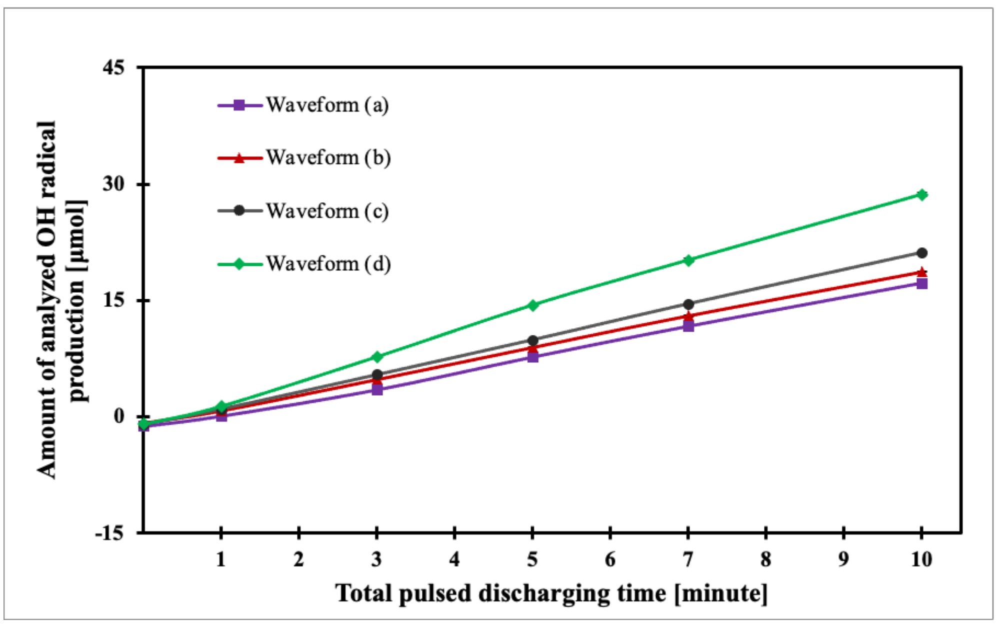


Fig. 49 Production of OH radical realized by each waveform from (a) to (d).

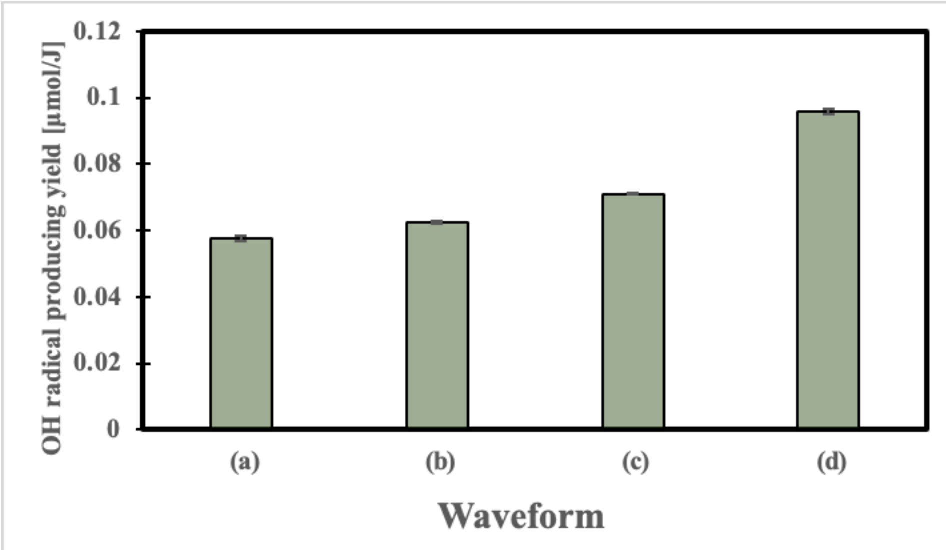


Fig. 50 Calculated energy efficiency for production of OH radical realized by each waveform from (a) to (d).

3.3.3 DISCUSSION OF THE EXPERIMENT RESULT

As mentioned in the previous section, the decrease of electric field due to number of neutralized ions in the discharge area when the primary streamer head had already touched the ground electrode [79] [80]. This phenomenon may lead to the difficult in generating OH radical by applying pulse longer duration at the same average input energy, and the effect of reduced electric field can be considered as a reason, which has been detailed in above Chapter II.

In the next section, the evaluation OH radical production of only the last part of pulse will be performed as an explanation for this expectation.

3.4 EVALUATION OH RADICAL GENERATION OF ONLY THE LAST PART OF PULSE

3.4.1 MEASUREMENT OF THE OUTPUT VOLTAGE AND CURRENT

In this section, the voltage at the last part of pulse was increased to boost the electric field in this phase, and the OH radical production of only this phase was evaluated and analyzed. The Fig. 51 to Fig. 54 shows the concept of pulse shape applied in these experiments for overview and in detail.

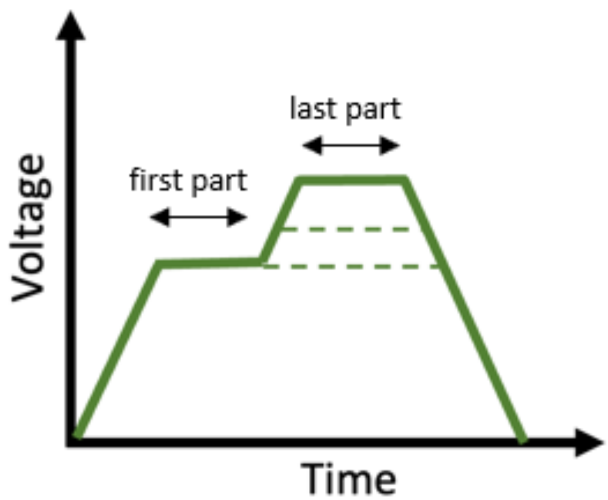


Fig. 51 Concept of waveform with adding up voltage at the behind.

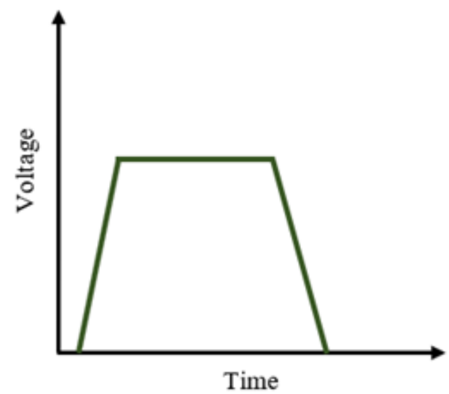


Fig. 52 Concept of pulse shape with same voltage for the first and last part.

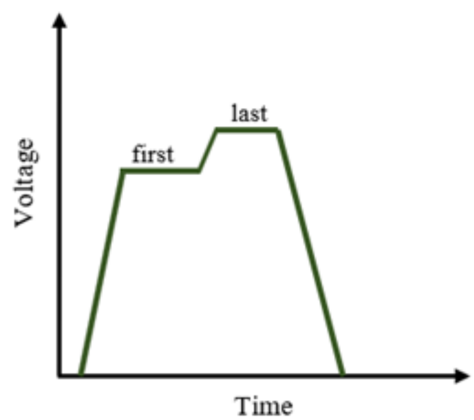


Fig. 53 Concept of pulse shape with higher voltage last part of pulse.

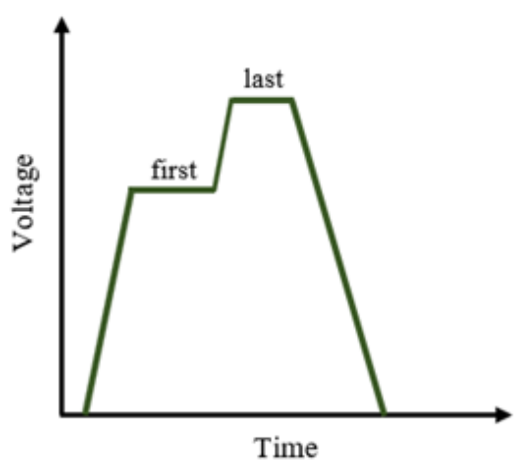


Fig. 54 Concept of pulse shape with more added up voltage last part of pulse.

In the next Fig. 55 to Fig. 57, measured output waveform of these concepts is shown. The Table. 10 shows the information of each waveform in Fig. 55 to Fig. 57.

In addition, the result of output with only 55-pulse-width waveform is shown in Fig. 58, this result was used for analyzation of the OH radical production of only the last phase by each waveform from Fig. 55 to Fig. 57.

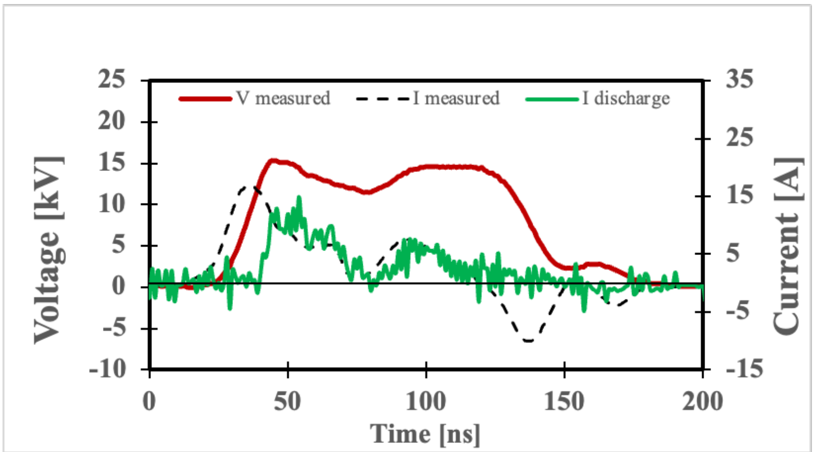


Fig. 55 Pulse shape with same voltage for all part of pulse marked as waveform number (1).

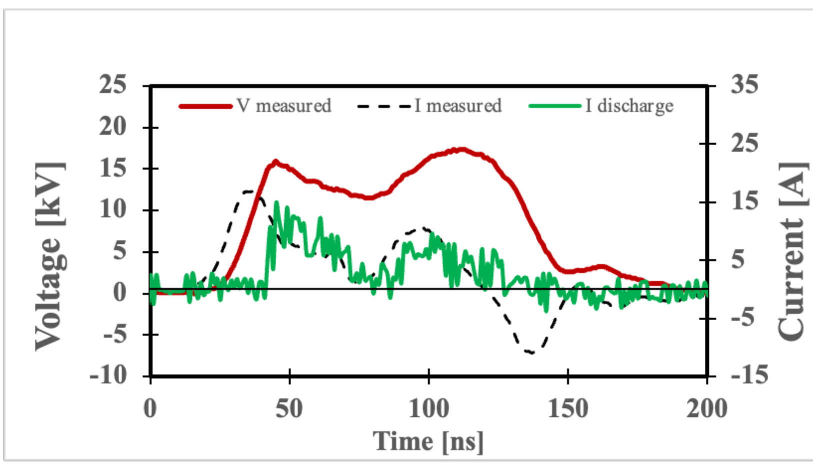


Fig. 56 Pulse shape with higher voltage for the last part of pulse marked as waveform number (2).

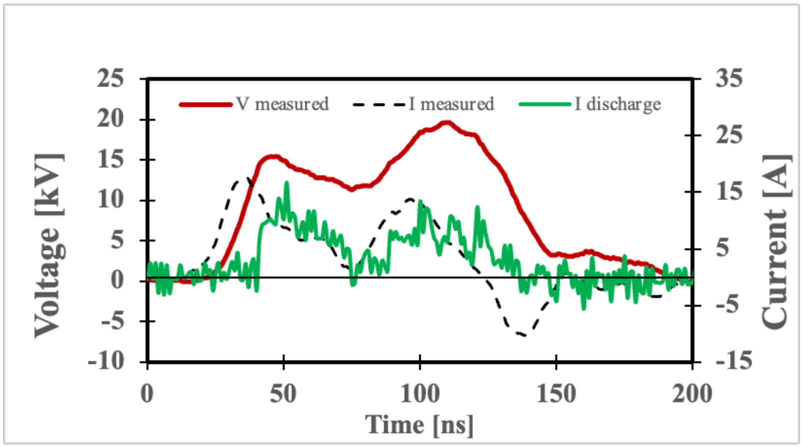


Fig. 57 Pulse shape with highest voltage for the last part of pulse marked as waveform number (3).

Table. 10 Information of pulsed discharge with waveform from number (1) to number (3).

Waveform	Repetition rate of each pulse waveform (Hz)	Repetition rate of waveform number 4 based on each waveform (Hz)
Number (1)	81	81
Number (2)	68	68
Number (3)	50	50

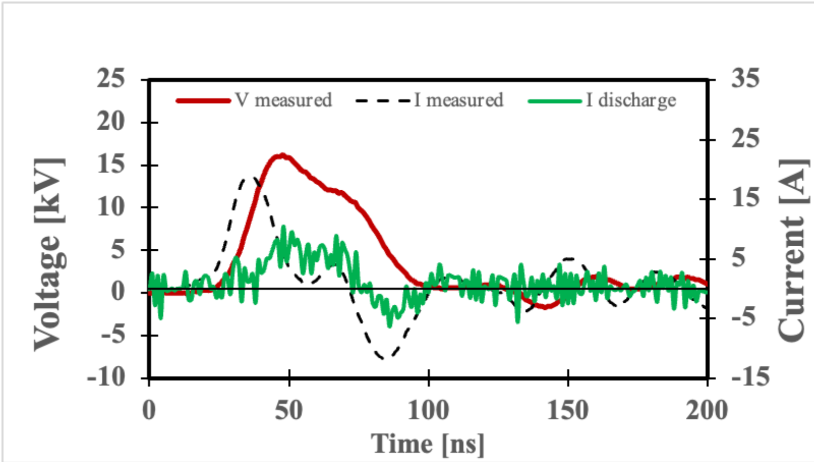


Fig. 58 Output of pulsed discharge with only single 55-ns-pulse-width waveform marked as waveform number (4).

In order to evaluate OH production of only the last phase of pulse with waveform marked number (1) to (3), the results of energy and amount of OH production of each waveform is withdrawn by the value of these information of waveform marked number (4). As a result, the following Fig. 59 to Fig. 61 is the imagine of this evaluating method, the dark side in the figure is the withdrawn part and the bright side is the evaluated part.

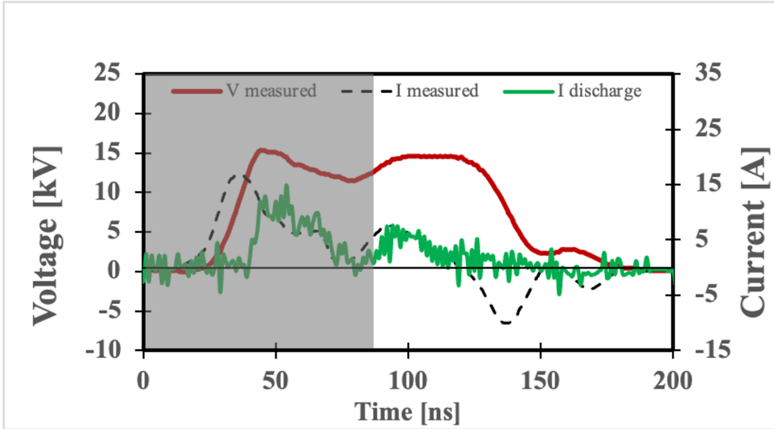


Fig. 59 Imagine of withdrawn value of waveform marked as waveform number (1).

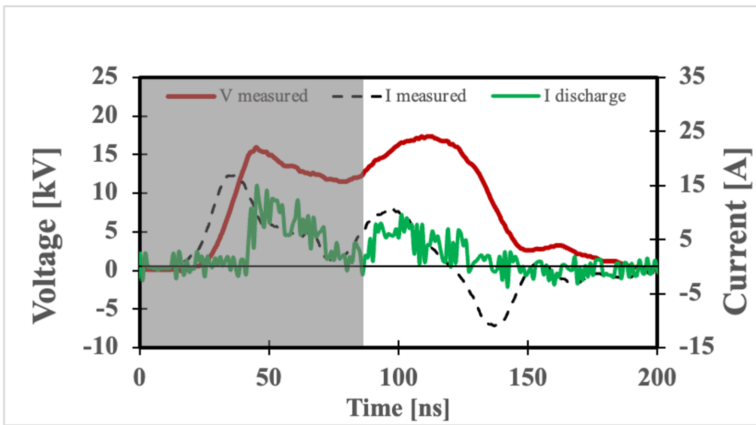


Fig. 60 Imagine of withdrawn value of waveform marked as waveform number (2).

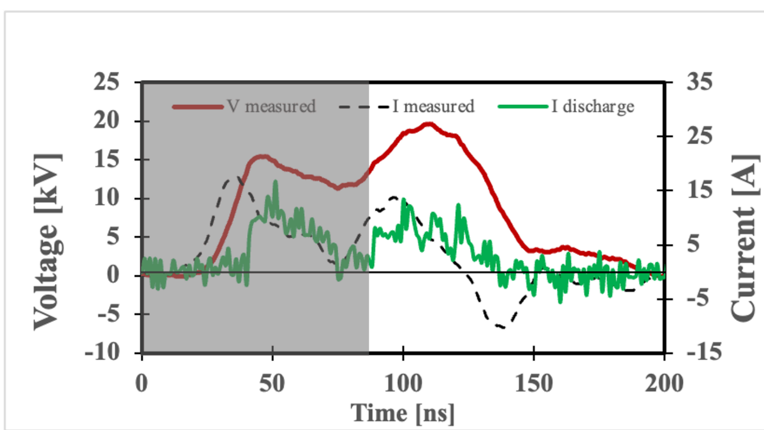


Fig. 61 Imagine of withdrawn value of waveform marked as waveform number (3).

3.4.2 RESULT OF THE EVALUATION AMOUNT OF OH RADICAL PRODUCED IN ONLY THE LAST PHASE OF PULSE AND DISCUSSION

In this part, amount of OH radical produced at only the last phase of pulse is introduced and analyzed. Fig. 59 and Fig. 63 is the amount of OH radical produced at only the last phase of pulse with waveform number (1) to (3) and the energy efficiency of the last phase for OH radical production, respectively. As in Fig. 62 and Fig. 63, when the voltage was added up more in the last part of pulse and only the last phase of pulsed discharge is analyzed, the amount of produced OH radical can be improved clearly, the energy efficiency is also improved by this adding up more voltage for the last part.

The possible explanation for this phenomenon is as following. Similar with being mentioned in the previous part, the neutralization of ion can be considered as physical reason for the decrease in electric field in the last phase. Therefore, analyzing for the last part of pulse only, by boosting electric field by adding up more voltage for the last part of pulse, the improvement in OH production and energy efficiency can be obtained.

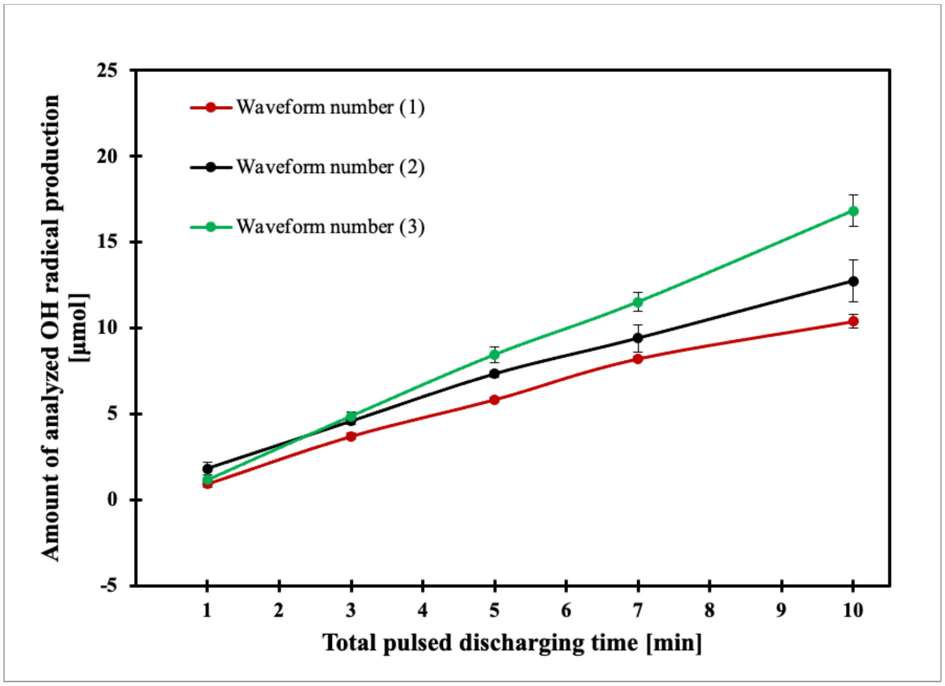


Fig. 62 The analyzed amount of produced OH radical.

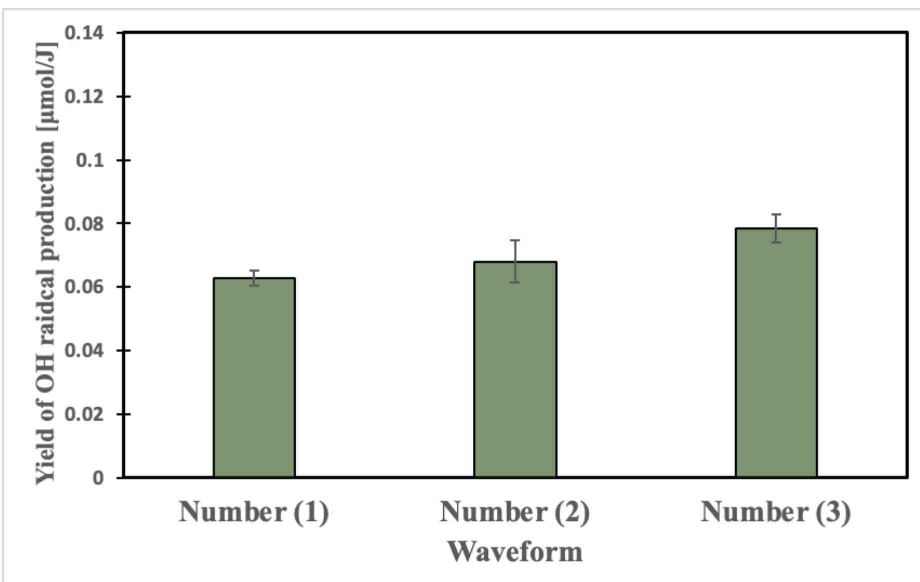


Fig. 63 The analyzed energy efficiency of OH radical production.

Chapter IV

CONCLUSION

In this study, pulsed power technology was mainly applied for investigating the effect of pulse shape (pulse duration) and period between each pulse on production of active species (ozone and OH radical) in the water treatment equipment. Linear Transformer Driver (LTD) system was used as the high-voltage pulse generator for all experiments in this study. Based on unique technical function of LTD system, a lot of complex-waveform high-voltage pulse has been generated efficiently. By the output pulse from LTD system, a lot of characteristics of pulsed discharge become possible for evaluating production and energy efficiency of active species. From the above Chapters, plenty of experiment condition have been considered for evaluating production of ozone and OH radical. The following are conclusion of these results,

- About the effect of characteristic of pulsed discharge on ozone production, it can be concluded that lower or unavailable of voltage at the second half of voltage pulse can release clearly high concentration of ozone produced in water treatment equipment. In these experiments, average total input energy with applying each waveform was maintained as unchanged, so these experiment results can be considered for investigating the effect of electric field - which may be decreased at the second phase due to neutralization of ions - when the first streamer head has already taught the ground on ozone production at this time.
- With applying pulse shape with varied pulse interval between two pulses, these experiment results can be summarized that when two pulsed discharges have shorter interval between application, it could be expected that more ozone decomposing

reaction will happen and lead to decrease the production of ozone in total and the energy efficiency is also wasted for this production.

- In the next part of this study, evaluation OH radical production was done to investigate effect of pulsed discharge on this production. From the experiment results, it can be confirmed that with the same average input energy lower voltage or disable voltage can lead to the increase in OH radical production in total. The reason for this phenomenon may be the same with above results of evaluating production of ozone, OH radical will be produced more difficultly at the second phase of pulsed discharge due to the decrease of electric field of the second phase.
- Finally, experiments of evaluating production of OH radical at only the second phase of pulsed discharge was done. From the results, it can be concluded that when the electric field of the second phase was strengthened again by adding more voltage to the second phase, the production of OH radical at this phase can be increased.

In conclusion, when the same average energy of pulse is applied, from the time that the primary streamer touched ground, production of active species will be decreased due to the decrease of electric field in the area of reactor. When the interval between pulses is longer than lifetime of the factor of active species decomposition, the production of active species can be improved.

From the mention above, in spite of some severe requirement of pulse generator, it can be considered that if the inefficiency pulse duration is decreased at the time electric field begins to decrease or effectively strengthening the electric field at the decreased phase, the production of active species can be improved and lead to improve the efficiency of water treatment in general. Furthermore, after the first voltage pulse is applied, a certain time is needed for the next application of the second pulse to improve the production of active species and water treatment efficiency for the last stage of application pulsed discharge.

Chapter V

ACKNOWLEDGMENT

In the first place, I would like to express my profound gratitude to Professor Weihua JIANG, who has been my academic supervisor for very long time. Professor JIANG is not only one of the most illustrious scientific professors in field of pulsed power technology, but he is also a very kind and zealous academic professor. Through working and communicating with Professor JIANG, I have learnt plenty of experiences, all of which is very valuable for me to go further professionally. Especially, Professor JIANG always encourages international students learn how to adapt to Japanese culture and thinking way; I am a foreign student and hope to work in Japan after graduation, so all direction from Professor JIANG is very meaningful for me in the future of my life in Japan. Every time I were stuck on my studies, Professor always encouraged and gave me valuable direction, which were as motivation for me to stand up and move forward on my studies.

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RESEARCH ACHIEVEMENT

Publication for international journal

1. **Phung Nhat Thanh, Taichi Sugai, Akira Tokuchi, Weihua Jiang, "The Effect of Pulse Discharge Control on Ozone Production," IEEE Transactions on Plasma Science, Vol. 50, No. 4, Apr. 2022**

2. **Phung Nhat Thanh, Taichi Sugai, Akira Tokuchi, Weihua Jiang, "The Influence of Pulse Shape on the Production of OH Radicals by Pulsed Discharge," IEEE Transactions on Plasma Science, Vol. 51, No. 1, Jan. 2023**

Presentation at Academic Conference

1. **Phung Nhat Thanh、須貝　太一、江　偉華、“パルス放電を用いた水処理装置の波形制御による活性種の測定、” 令和２年度核融合科学研究所共同研究研究会、online、2021 年１月７～８日**

2. **Phung Nhat Thanh, Taichi Sugai, Akira Tokuchi, Weihua Jiang, “The Influence of Shape and Timing of Applied Pulsed Voltage on Active Species Production in Electric Discharge Water Treatment Equipment,” P3-24, 8th Euro-Asian Pulsed Power Conference, Biarritz, France, online, 2021, Aug. 29th ~Sep. 2nd**

3. **Phung Nhat Thanh、須貝　太一、徳地　明、江　偉華、“大気圧パルス放電による OH ラジカル生成のための最適なパルス形状の調査、”EPP-22-048、放電・プラズマ・パルスパワー研究会、まちなかキャンパス長岡、2022 年 5 月 18～20 日**

4. **Phung Nhat Thanh, Taichi Sugai, Akira Tokuchi, Weihua Jiang, “Evaluating the Effect of Pulse Shape on OH Radical Production at Secondary Streamer Phase,” 2-1099, 9th Euro-Asian Pulsed Power Conference, Seoul, Korea, online, 2022, Sep. 18th ~22th**

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