

# Development and Performance Evaluation of Hydroxyl Radical Generator using Electron Emission Type High Voltage and Low Current Discharger

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## 전자방사식 고압 저전력 방전을 이용한 OH radical 발생기의 개발과 성능 평가

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**Abstract** In this study, we developed an electron-emission OH radical generator for waste water treatment. The stability of the circuitry was ensured by implementing stable pulse waves with a MOSFET and reducing the momentary current rise. The OH radical generator uses a high-voltage and low-current discharger. The performance of the device was evaluated experimentally, which showed that it is possible to produce a stable and uniform pulse waveform for the drain current of the power MOSFET, which is connected to the input side of an AC multiplying converter through negative feedback circuitry with CR-snubber architecture. It was also possible to reduce the excitation current of the converter and improve the stability of the oscillation circuit. In addition, the generator can generate hydroxyl radicals stably. The bactericidal activities were also evaluated, and the germicidal power for *E. coli*, *S. aureus*, and *S. flexneri* was improved by 99.9% or more after 60 minutes.

**요약** 본 연구는 MOSFET을 이용하여 안정적인 구형파를 구현함과 동시에 순간적인 전류의 상승을 최대한 줄여 전체 회로의 안정성이 확보된 폐수처리용 전자방사식 OH radical generator를 개발하고자 하였다. 이와 같은 문제를 해결하고 안정성이 확보된 전자방사식 고압 저전력 방전을 이용한 폐수처리용 전자방사식 OH radical generator를 개발하고 그 성능을 평가하고자 하였다. 실험은 2016년 11월부터 2017년 3월까지 실시하였다. 그 결과 AC 체배 변환기의 입력단에 연결되는 Power MOSFET의 drain 전류는 CR-스너버 구조의 부궤환(negative feedback) 회로를 통해 안정적이고 일정한 펄스 파형을 제공할 수 있었으며 AC 체배 변환기의 여기 전류(excitation current) 감소 및 발진 회로의 안정성을 상승시킬 수 있었다. 또한 이와 같은 회로를 갖는 전자방사식 OH radical generator에서 OH radical을 안정정적으로 발생시킬 수 있었다. 또한 살균능을 평가한 결과, *E. coli*, *S. aureus*와 *S. flexneri*는 60분 후 최대 99.9%이상의 살균력을 보였다. 본 연구 결과를 기반으로, MOSFET을 이용하여 안정적인 구형파를 구현함과 동시에 순간적인 전류의 상승을 최대한 줄여 전체 회로의 안정성이 확보된 살균능이 우수한 폐수처리용 전자방사식 OH radical generator를 개발하였다.

**Keywords** : evaluating bactericidal activities, high voltage and low current discharger, MOSFET, OH radical, pulse wave

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## 1. Introduction

Since the Industrial Revolution, the advancement in scientific technology along with the rapid industrial growth, the population growth, and the elevated standard of living has accelerated water shortage owing to the growth of water demand and the indiscriminate development of ground water, as well as has caused serious pollution of water resources[1], which has become serious threats to the environment and the human health[2-7]. Such phenomena has aggravated to the level that natural cycles could not purify the pollution[8,9]. Accordingly, there has been requirement for eco-friendly water treatment technologies to secure sufficient quantity and quality of water based on immaculate treatment of polluted water resources.

In reality, the processing of industrial wastewater had been made difficult and aggravating, as it has been interlinking with national policies of development and growth. In addition, it has been difficult to find appropriate economical and efficient processing methods for livestock sludge or landfill leachate until today and the legacy biological treatment method have begun to reveal their limitations. To solve such problems, there have been some researches on physicochemical processing methods, of which utilization has been increasing progressively.

In comparison with general chemical treatments, the advanced oxidation processes(AOPs) are used to improve the oxidative power[10, 11]. AOPs are advanced techniques used to oxidize aquatic pollutants such as organic matters and non-degradable organic materials by injecting hydroxyl radicals, which have much powerful oxidative power than generic oxidizing agents used in most oxidation processes. Some advanced oxidation processes currently deployed in Korea include Fenton Oxidation, Ozone Oxidation, Photocatalytic Oxidation, Electronic Beam Oxidation, Ultrasonic Irradiation Oxidation and so on[10, 12]. Since these advanced oxidation processes use hydroxyl radicals with powerful oxidative power as oxidizing

agents, they are much more efficient in oxidizing organic matters than legacy chemical treatments. Although each of these treatments shows excellent performance, there are some troublesome shortcomings reported. Since incomplete removal of organic matter and excessive cost of hardware and operating expenses, these advanced oxidation processes have been used in pretreatment processes in biological treatments or in some limited extent, and have their limitation in processing waste water containing high concentration of organic matters. Thus, it is an urgent task for the protection of rapidly deteriorating water resources caused by rapid industrialization to develop technologies, which can overcome these shortcomings and process organic matters and non-biodegradable materials flawlessly.

Taking into account the current situation of needing the development of advanced oxidation processes for waste water treatment, we tried to develop an electron emission type hydroxyl radical generator for waste water treatment based on electronic beam oxidation process with an electron generation device and an electromagnetic wave generator, which are applied in advanced water treatments and different from legacy water treatment techniques. However, most of these generators are designed to generate pulse wave by using operational errors of Bipolar Junction Transistor(BJT) with astable multi-vibrator type harmonic pulse generating circuitry. Since such architecture uses current rise/drop based on errors of resistance elements of the circuit, it is possible to cause serious damage to transistors owing to fluctuation of output voltage or momentary current rise (Gibbs phenomenon) of collector side. In addition, in terms of durability, it may cause serious damage to the booster circuit.

In this study, we tried to solve such problems by developing an electron emission type hydroxyl radical generator for wastewater treatment using reliable electron emission type high voltage and low current discharger, as well as evaluate its performance.

## 2. Method

### 2.1 Design of Electron Emission Type High Voltage and Low Current Discharger

Fig. 1 shows a diagram of astable multiple oscillator circuit, which is a circuit used to generate high frequency electromagnetic pulse for existing astable multi-vibrator. It is generating pulse waves based on operational errors of BJT at both sides. Since such architecture uses current rise/drop based on errors of resistance elements of the circuit, it is possible to cause serious damage to transistors owing to fluctuation of output voltage or momentary current rise of collector side. In addition, in terms of durability, it may cause serious damage to the booster circuit. To solve the above problem, we designed the device to improve the stability of entire circuitry by realizing stable pulse waves and at the same time by reducing the momentary current rise as much as possible, which has been available as we solve the Gibbs phenomenon of current by applying feedback circuits with CR-snubber architecture using volt age mode MOSFET which can operate stably, instead of current mode BJT. By taking into account operating environment of transistors and legacy circuits, we used SE-555 parts with relatively wide temperature rage for pulse generation. At this time, the frequency of the pulse waves was designed to optimize against the resonance characteristics of the AC voltage multiplying converter. The signal output from SE-555 is switched through IRF power MOSFET to control the input to the multiplying converter, and a

degeneration resistor was used at the source side of MOSFET to control entire power as well as to supply operating current without a hitch.

As mentioned above, the test was configured to improve the stability of entire circuit by minimizing the momentary current rise. SE-555 and IRF power MOSFET were used to control the entire power and at the same time to supply the operating current smoothly. Accordingly, the results as shown in Fig. 2 were obtained.

The current waveform shown in Fig. 2 was the result of a simulation stabilizing the current flow fluctuating instantaneously owing to the operation of the oscillating circuit by using CR-snubber. The drain current of power MOSFET connected to the input side of the AC multiplying converter was able to provide stable and uniform pulse waveforms through a negative feedback circuit with CR-snubber architecture. Accordingly, it was possible to reduce excitation current of the AC multiplying converter and to improve the stability of the oscillating circuit. At this time, the optimal frequency of the pulse generating circuit had to induce by taking into account the characteristics of operating frequency of the AC voltage multiplying converter.

For the characteristics of band pass filtering of the multi-step multiplier, it was able to get cost savings in terms of power efficiency and implementation if it is possible to optimize the specification of each element and the number of steps of the multiplier circuit.

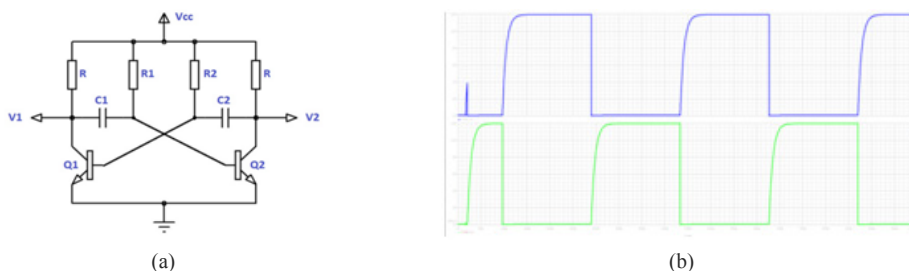


Fig. 1. High-frequency pulse generator circuit diagram of astable multi-vibrator structure (a) and the ideal pulse output waveform (b)

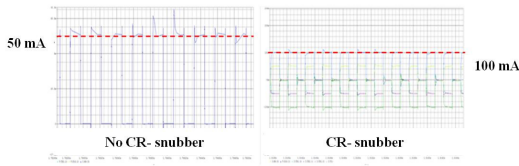


Fig. 2. Drain-side current waveform of power MOSFET based on the availability of CR-snubber

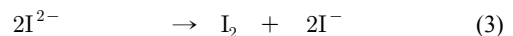
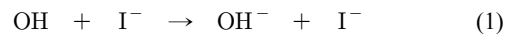
## 2.2 Analysis of the Generation of Hydroxyl Radicals

To measure the quantity of OH radicals generated at the electron emission type Hydroxyl radical generator for waste water treatment with highly stable circuitry, we applied existing studies of Chunde et al.[13]. and Son et al.[14], which proposed fluorescence analysis using potassium iodide(KI). To measure the generated quantity of OH radicals using KI, we measured the peak value appeared at 350 nm with a UV-Vis spectrophotometer[14]. In this study, to measure the quality of hydroxyl radicals generated at the hydroxyl radical generator, we measured the volume of generated hydroxyl radical dissolved in 6 L of distilled water at intervals of 10 minutes. After running the hydroxyl radical generator, we collected 50 ml of distilled water with dissolved hydroxyl radicals two times at intervals of 10 minutes, and then reacted them with KI 16.6 mg/L and 50.0 mg/L of KI and measured the absorbance. In general, when KI is dissolved in distilled water, it becomes  $K^+$  and  $I^-$ . At this time,  $I^-$  ions are oxidized by hydroxyl radicals as shown in Equation (1), and generated  $I_2^-$  ions as shown in the reaction of Equation (2).  $I^-$  ions and diatomic iodine ( $I_2$ ) ions in the solution are reacted as shown in Equation (4) to generate triiodide ( $I_3^-$ ) ions. The concentration of triiodide ions generated in the above reaction process can be displayed in term of an indirect indicator of hydroxyl radical generation. The reaction equation is as follows[13,14]

## 2.3 Evaluating Bactericidal Activities

To evaluate the bactericidal activity of the manufactured hydroxyl radical generator, the Korea

Testing & Research Institute was requested to perform the assessment. The method of evaluation was briefly described in the followings: First, put 6 L of sterilized distilled water in several sterilized containers and pollute then by inoculating the test strains. The strains used in the test include *Escherichia coli* ATCC 25922 (*E. coli*), *Staphylococcus aureus* ATCC 8538 (*S. aureus*), and *Shigella flexneri* ATCC 12022 (*S. flexne*). After culturing colon bacillus and *Staphylococcus aureus* in liquid media, dilute the cultured bacterial strain to make the initial bacterial count  $(1-9) \times 10^5$  CFU/ml. after inoculating the container with six liters of sterilized distilled water, measure the initial bacterial count, and then run the hydroxyl radical generator in the polluted solution. After 1, 10, and 60 minutes, respectively, collect the solution and measure the number of viable microbe cells. In the first dilution stage of every test, use the D/E neutralizing broth (DIFCO, USA) to neutralize the solution for the test. When culturing the bacteria in the culture media, the bacterial count is obtained by multiplying the bacterial count of the culture medium by a dilution factor. When the bacteria are not cultured in the culture medium, then multiply a dilution factor for the neutralization stage and display the value as less than 10 ( $<10$ ). The measurement of bacterial count in every stage was performed by using the Tryptic soy agar (DIFCO, USA), and the number of viable microbe cells was estimated by using Equation (5) and the bacterial reduction rate by using Equation (6).



where,  $N$  is the number of viable microbe cells,  $C$  is the number of colonies,  $D$  is the dilution factor,  $R$  is the reduction rate,  $A$  is the initial bacterial count, and  $B$  is the bacterial count after a certain period of time.

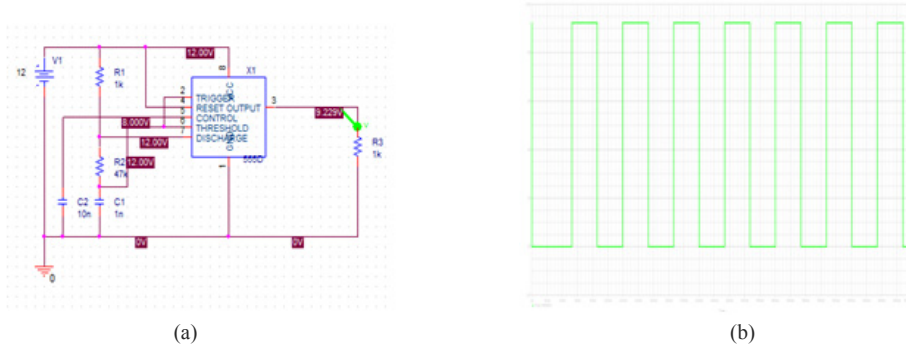
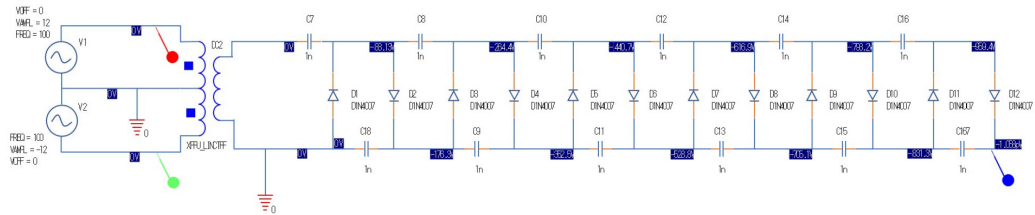


Fig. 3. OrCAD circuit diagram of E-555 (a) and pulse output waveform of SE-555 (b)

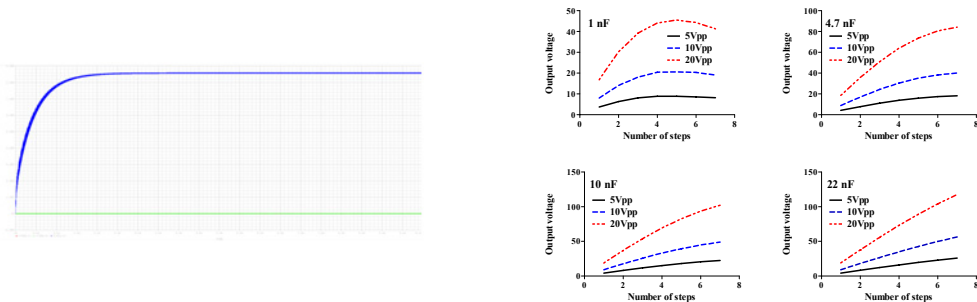
### 3. Results and Discussion

Fig. 3 shows OrCAD circuit diagram and pulse output waveform of SE-555 designed to generate pulse waves with the frequency of 27 kHz. Based on each resistance value and capacitor value, it is possible to control the frequency of pulse wave. As shown in the following output waveform, this device can produce accurate duty ratio, as it has rather short pulse rise time compared to the existing astable multiple oscillator circuit.

The multi-step multiplier for voltage charging was used to boost the output voltage of the AC voltage multiplying converter to 10 kV by using ceramic capacitors and diodes with withstand voltage of 2 kV or more. The configuration of each step in the multi-step multiplier is the Villard voltage doubler type, of which output is 10 kV DC with half-wave rectification conducted by using two diodes and two capacitors in each step. In addition, in a multi-step multiplier, the combination of capacitors and diodes may act as series C-R circuit, so it may be related to



(a) Design of transformer and simulation of interworking with Villard voltage doubler



(b) Input waveform and output DC waveform of dual input transformer

(c) Output voltage according to the number of steps and the capacity of the capacitor

Fig. 4. Circuit used to check coupling and operation of AC voltage multiplier and multi-step multiplier and the simulation result.

the characteristics of frequency. It may affect the discharging cycle when generating pulses owing to the change of the time constant according to the capacity of the capacitors. As a result, the result as shown in Figure 6 can be obtained.

Fig. 4 shows a circuit used to check the coupling of the AC voltage multiplier and the multi-step multiplier as well as their operations, and the simulation result. It shows DC voltage at each stage according to the number of steps and the capacity of the capacitor of the multi-step multiplier. At this time,  $V_{pp}$  means the amplitude of the input AC voltage. The figure shows that there is the optimized number of steps for the capacity of the capacitor, and the larger the capacity becomes; the more the number of steps becomes available. However, as shown in the above, as the large capacity of the capacitor would increase value of the time constant, it will not make the discharges occurred uniformly when the pulses are generated. Therefore, it is necessary to use optimal number of steps and appropriate capacity for the capacitor:

Fig. 5 shows the results of simulations and measurements performed to check the operating characteristics of the AC voltage multiplying converter and the multi-step multiplier of the 10 kV multi-step booster circuit. For power MOSFET of the pulse generating circuit, IRF-610 was used by taking into account the appropriateness of switching speed for the output of SE-555 and the current characteristics, and the maximum drain current was 3.3A. The results showed that the measured values and the simulated values were similar, and it was possible to check the operation of

the AC voltage multiplying converter and the multi-step multiplier through the results.

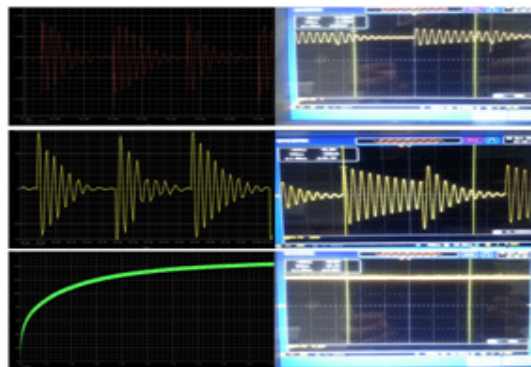


Fig. 5. Results of simulations and measurements performed to evaluate operating characteristics of the AC voltage multiplying converter and the multi-step multiplier of the 10 kV multi-step booster circuit. Input and output waveforms of the AC voltage multiplying converter and output waveform of the multi-step multiplier.

To measure the quantity of hydroxyl radicals generated in the hydroxyl radical generator, we used two settings for the capacity of KI: 16.6 g/L and 50 g/L. Fig. 6 shows the result. As shown in the figure, when the time of injecting hydroxyl radicals is longer, the generation of hydroxyl radicals is increased. When injecting hydroxyl radicals for an hour, it increased by about  $269.32 \pm 4.68$ (SD)% before the injection of hydroxyl radicals. In addition, when measuring the quantity of hydroxyl radicals after leaving it alone for an hour from the point of stopping the injection of hydroxyl radicals, the results shows the decrease of about 1.27%; however, as this value is within the limit

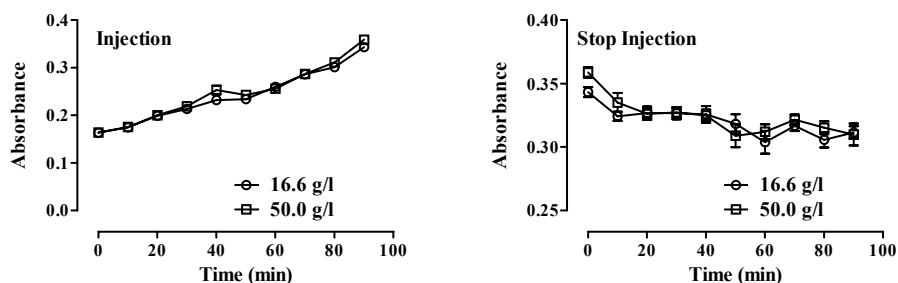


Fig. 6. The results of measuring absorbance with the passage of time



of error, so it shall be deemed as almost similar. It maintains the increase of about  $254.59 \pm 5.27(\text{SD})\%$  before injecting hydroxyl radicals. From the result, it is possible to generate hydroxyl radicals reliably with circuitry based on SE-555 and IRF power MOSFET. In addition, when using two volumes of 16.6 g/L KI and 50 g/L KI, they are almost identical within the limit of error of the test results.

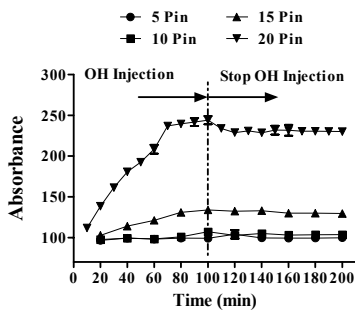


Fig. 7. The results of measuring generated hydroxyl radicals according to the number of discharger pins of the hydroxyl radical generator

Fig. 7 shows the results of measuring generated hydroxyl radicals according to the number of discharger pins of the hydroxyl radical generator. As shown in the figure, there is no significant difference when the number of discharger pins is 5 and 10, but when the number of discharger pins is 20, it increases up to 2.5 times. In addition, the longer the discharge time becomes, the larger the hydroxyl radical generation is, though the value does not increase after 70 minutes. That is, although the quantity of radicals decreases slightly after stopping the injection of hydroxyl radicals, it has maintained constantly during

the measurement of 100 minutes.

Both Table 1 and Fig. 8 show the evaluation result of bactericidal activities. They show the germicidal power of 17.2% for *E. coli* after a minute, 99.9% after 10 minutes and more than 99.9% after 60 minutes. In case of *S. aureus*, there is no germicidal power for a minute, then 99.9% after 10 minutes and more than 99.9% after 60 minutes. In addition, for *S. flexneri*, the germicidal power is 29.6% after a minute, 34.1% after 10 minutes, and more than 99.9% after 60 minutes.

Table 1. The evaluation result of bactericidal activities with the passage of time

Division	Initial	After 1 min	After 10 min	After 60 min	Etc
<i>E. coli</i>	$2.9 \times 10^5$	$2.4 \times 10^5$	60	< 10 (99.9% more than)	Gram negative (Total coliform)
<i>S. aureus</i>	$1.3 \times 10^5$	$1.4 \times 10^5$ (-)	40	< 10 (99.9% more than)	Gram positive (Bacteria)
<i>S. flexneri</i>	$4.4 \times 10^5$	$3.1 \times 10^5$	$2.9 \times 10^5$	< 10 (99.9% more than)	

#### 4. Conclusion

In this study, we were able to provide stable and uniform pulse waveforms for drain current of the power MOSFET connected to the input side of the AC multiplying converter through the negative feedback circuit with CR-snubber architecture, and it helped to reduce the excitation current of the AC multiplying converter and to improve the stability of the oscillating circuit. It was possible to adjust the frequency of pulse

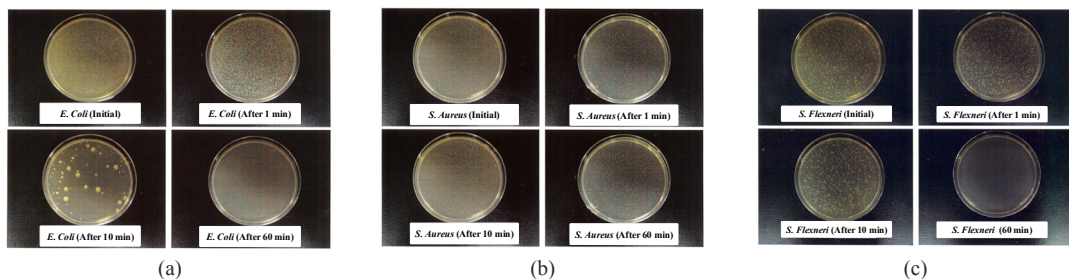


Fig. 8. Results photographs of bactericidal activities with the passage of time. (a) *E. coli*, (b) of *S. aureus*, (c) *S. flexneri*

waves with resistance value and capacitor value, respectively. Since the pulse, rise time is rather shorter than that of the existing astable multiple oscillator circuit, it was possible to generate accurate duty ratio. Hydroxyl radicals were generated stably from the hydroxyl radical generator by using reliable electron emission type high voltage and low current discharger, and the generation of hydroxyl radicals has been increased over time. After terminating the injection of hydroxyl radicals and leaving it unattended for an hour, it maintained the uniform state. Finally, the result of evaluating bactericidal activities showed the germicidal power of 17.2% for *E. coli* after a minute, 99.9% after 10 minutes and more than 99.9% after 60 minutes. In case of *S. aureus*, there is no germicidal power for a minute, then 99.9% after 10 minutes and more than 99.9% after 60 minutes. For *S. flexneri*, the germicidal power is 29.6% after a minute, 34.1 % after 10 minutes, and more than 99.9% after 60 minutes. Based on the results of this study, we developed an electron emission type OH radical generator with excellent sterilization ability for waste water treatment of which circuitry is ensured for stability by implementing stable pulse waves with MOSFET and at the same time by reducing the rise of momentary current.

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<Research Interests>

Biotechnology, Biomedical Engineering