I. INTRODUCTION

Effective use of water resources are required as a result of the global increase in population and industrial development [1]. Sewage disposal is performed on the basis of condensation, precipitation, filtration, and disinfection. There are methods for wastewater control such as activated carbon adsorption [2], chemical treatment [3], activated sludge [4], corona discharge [5], and enzymatic polymerization [6]. With the current water disposal technology, extermination of cryptosporidium and disassembly of volatile organic compounds, such as trichloroethylene, are difficult. Therefore, a new water treatment technology is required. Discharge in the water is mentioned as one of them [7-17]. Decomposition of harmful persistent organic compounds and sterilization of microorganisms are expected during discharge in water through the occurrence of ultraviolet rays, shock waves, ozone, and OH radicals. The application range of water purification technology is broad and the requirement of large equipments could be an obstacle in the use at small-scale facilities.

In this paper, water treatment is experimentally investigated by using low voltage discharge below 10 kV in the water. Short gap discharge in still water and large gap discharge in bubbling water are used for water treatment.

Abstract—Water treatment is experimentally investigated by using low voltage discharge below 10 kV in the water. Low voltage discharge in water is suitable for practical application of water treatment, since necessary apparatus could be small, inexpensive, and characterized by low power consumption. In this research on the examination of water treatment by low voltage discharge in the water, discoloration processing of indigo carmine solution and sterilization of Escherichia coli were carried out. Short gap discharge in still water and large gap discharge in bubbling water are experimentally compared as water treatment methods. In both discoloration and sterilization, the treatment performance is higher with large discharge gap with gas bubbling than that of the short gap discharge in still water. Air, nitrogen, and oxygen are used to bubble the water for treatment. The treatment performance improves when oxygen is blown in the solution. These results could be the effect of generation of active radical species such as ozone, and also due to the combination of churning effect and the increase of treatment volume.

Keywords—Low voltage discharge, sterilization, discoloration, ozone

II. EXPERIMENTAL SETUP AND METHOD

A. Pulsed Power Supply

When voltage is applied to electrodes, generation of oxygen and hydrogen by electrolysis and temperature rise occur through the Joule effect, since the electric conductivity of liquid, such as water, is high.

For this reason, it is necessary to apply a pulsed voltage to cause a discharge in a liquid. A pulsed power supply is used for water treatment, suggesting its advantage for discharge in water [18, 19].

In this research, pulsed voltage is generated using a spark gap switch as shown in Fig. 1. The spherical diameter of the spark gap switch was 24 mm. Pulse voltage was generated by the following methods:

1) AC100V is increased to 8 kV using a neon transformer.
2) Half-wave rectification of the high voltage is carried out using a diode.
3) A capacitor is charged with the voltage which carried out half-wave rectification.
4) The energy charged by the capacitor is momentarily sent to the electrodes with a spark gap.

B. Experimental Setup

The electrode system and the experimental apparatus are shown in Fig. 2. Different gases are introduced into the water treatment reactor, which contains two metallic electrodes.

When water between the electrodes is bubbled by...
gas, an injection needle was used as a needle electrode. A tungsten wire (Ø 0.5 mm) was used as a discharge electrode to increase the electric field for short gap discharge and when no gas is bubbled in the water. A punched metal disc was used as a plate electrode connected to ground with a diameter Ø 40 mm. The needle electrode was placed beneath the plate electrode, thus when the gas was pumped into the water, the formed bubbles will rise towards the plate electrode. The discharge gap was set at 10 mm for the electrode configuration with needle electrode and at 0.3 mm for the wire electrode.

The electrical characteristics were measured by a high voltage probe (Tektronix, P6015), a current probe (Tektronix, P6021), and a digital oscilloscope (Tektronix, TDS2014B). The concentration of indigo carmine solution before and after discoloration was measured by HPLC (Agilent, 1100Series) and UV Spectrophotometer (SHIMADZU, UV-3100PC). Dissolved ozone concentration in the water was measured using an ozone monitor (Ebara Jitsugyo, PL-620A).

C. Control of Organic Compounds

The effect of water treatment was examined with indigo carmine solution as an organic compound. Initial concentration of indigo carmine solution is 20 mg/L, and the volume of sample liquid is 50 mL. Discharge time was 5, 10, 15, 20, and 25 minutes, respectively. Two electrode systems were used in this research. A point – plain electrode with gas bubbling has a 10 mm discharge gap. The other electrode also has point - plain electrode without bubbling. The gap was set at 0.3 mm, ensuring a low discharge voltage in the water.

D. Sterilization of Microorganisms

Microorganisms such as pathogenic bacteria are regarded as a critical issue in water environment management. The sterilization of various types of bacilli is covered in this study [18-20]. In this research, Escherichia coli HB 101 (Gram-negative bacteria) were used as the object of the sterilization. The volume of sample liquid was 50 mL. Discharge time was 5, 10, 15, 20, and 25 minutes, respectively. After the discharge treatment, the laboratory dishes were incubated in the incubator at 37 °C for 15 hours. Sterilization effect of discharge in the water was inspected by comparing the number of colonies with and without discharge. The electrode systems used in the series of experiments were the same as the treatment of indigo carmine.

III. RESULTS AND DISCUSSION

A. Electrical Characteristics

Image of discharge in bubbled water is shown in Fig. 3. When the pulse voltage was applied with bubbled water, the gap distance was set at 10 mm. The exposure of the needle electrode was 0.5 mm. Bubbles with bright discharges were observed. Diameter of bubbles were distributed between 1.0 ~ 3.0 mm.

Fig. 3 shows the discharge voltage waveform and corresponding discharge current with ion-exchanged water. Rising time of pulse was about 350 ns.
The applied voltage was 8 kV and the power consumption of the whole equipment was about 36 W. Peak of discharge current was observed at about 2 A.

In the series of experiments, ion-exchanged water, tap water, and indigo carmine solution in ion-exchanged water were used. The electric conductivity of various liquids is shown in Table I. The peak value of the discharge voltage becomes lower as the value of electric conductivity of the liquid becomes higher. A high frequency discharge current was measured at the rising slope of the discharge voltage.

Fig. 5 shows the image of discharge in water without bubbling. The discharge gap between the electrodes was set at 0.3 mm. The exposed length of needle electrode (tungsten wire \(0.5\) mm) is set at 0.5 mm. Tense arc discharges were observed between the electrodes.

Waveforms of discharge voltage and current are shown in Fig. 6. Without bubbling in ion-exchanged water, oscillation of discharge voltage and discharge current was observed. Discharge current was about 2 times higher than that measured at the large gap setting (Fig. 4).

**TABLE I**

<table>
<thead>
<tr>
<th>Type of water</th>
<th>Electric conductivity [(\mu)S/cm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ion-exchanged water</td>
<td>2</td>
</tr>
<tr>
<td>Tap water</td>
<td>125</td>
</tr>
<tr>
<td>Indigo carmine solution in ion-exchanged water</td>
<td>5400</td>
</tr>
</tbody>
</table>

**TABLE II**

<table>
<thead>
<tr>
<th>Without bubbling</th>
<th>Bubbling (Air, (O_2), (N_2))</th>
<th>Discoloration of indigo carmine</th>
<th>Sterilization of (E.\ coli)</th>
<th>Discoloration of indigo carmine</th>
<th>Sterilization of (E.\ coli)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Discharge voltage [kV]</td>
<td>4.5</td>
<td>7</td>
<td>3</td>
<td>5</td>
<td></td>
</tr>
<tr>
<td>Discharge gap [mm]</td>
<td>0.3</td>
<td>0.3</td>
<td>10</td>
<td>10</td>
<td></td>
</tr>
<tr>
<td>(R_2) [k(\Omega)]</td>
<td>0.3</td>
<td>0.3</td>
<td>12</td>
<td>12</td>
<td></td>
</tr>
</tbody>
</table>

Experimental conditions with both electrode systems are shown in Table II.

**B. Treatment of Indigo Carmine Solution**

Indigo has \(H\) type chromophoric group, therefore absorbs the visible light of a specific wavelength [23]. The chemical structural formula of indigo carmine is shown in Fig. 7. The double band could be broken by the oxidation reaction of ozone with isatin sulfonic acid [24].
Absorbance spectrum of indigo carmine solution with an UV spectrophotometer at various discharge time is shown in Fig. 8. The results were obtained without bubbling any gas.

In the spectrum before discharge treatment (0 min), the peaks at 200 nm, 251 nm, 287 nm, and 611 nm represent the absorption by the organic compound contained in indigo carmine.

The peak of 611 nm and 287 nm decreases with the increase in discharge time, along with the discoloration of indigo carmine solution. The solution after treatment of 30 minutes was almost transparent.

On the other hand, absorbance spectrum (200 nm and 251 nm) increases with electric discharge time. Fig. 9 shows the absorbance spectrum of ion-exchanged water at various discharge time.

After treatment in water, the peak of 200 nm increases. Hydrogen peroxide, which is a long-lived active species, could be generated by recombination reaction of OH radicals through discharge in water [25].

\[ \begin{align*} 
H_2O + e & \rightarrow OH^* + H \\
OH^* + OH^* & \rightarrow H_2O_2 
\end{align*} \]

In the presence of air and O₂ bubbles the following reaction will occur:

\[ e + O_2 \rightarrow e^+ O(P) + O(1D) \]  

The peak near 200 nm represents the absorption of the light by hydrogen peroxide and the byproduct generated by decomposition of indigo carmine [25]. H₂O₂ concentration after 9 minutes of treatment time was 84 µg/L.

The photograph of the indigo carmine solution after treatment is shown in Fig. 10. It is clear that the color of solution has been changed as a result of the treatment. The color of the indigo carmine solution was especially brighter when O₂ or air was supplied during discharge, (a) and (b)).

Results of the decomposition of indigo carmine solution by using a HPLC is shown in Fig. 11.

The treatment performance is higher with gas bubbling than without bubbling. The treatment performance improves due to the presence of the gas bubbles as per Fig. 11. The generation of radicals inside the bubbles and churning effect contributed to decomposition of indigo carmine.

Discharge processing was performed using tap water for 20 minutes. Dissolved ozone concentration was measured every 2 minutes, as shown in Fig. 12.

When O₂ was supplied to the solution, dissolved ozone concentration was about 20 % higher compared
with the case when air was supplied. At a discharge time of 20 minutes, the ozone concentration of 1.39 mg/L with O₂, and 1.15 mg/L with air was obtained. The bubbles in which ozone was produced are rapidly emerging at the surface of water. Thus the contact time between ozone and water is limited and does not favor a high dissolved ozone concentration, even when the bubbling gas was pure oxygen.

C.  Sterilization of E. coli

Fig. 13 shows the E. coli density versus discharge voltage at short and large discharge gaps as a semi-log plot. The D-value for E. coli sterilization after water treatment with no bubbles Dno bubbling = 17 min shows that when no gas was used the sterilization process was relatively slow. When N₂ and O₂ were used for producing bubbles at large discharge gaps the sterilization process was improved according to the D-values for nitrogen DN₂ = 14 min and for oxygen DO₂ = 11 min respectively. Sterilization using air bubbles was the most effective method, with a D-value of Dair = 10 min. A shorter period of time was necessary for the complete sterilization of E. coli when air or O₂ was used to bubble the water. This could be explained by the generation of radicals and ozone in the bubbles containing O₂ or air. These results were obtained at large discharge gaps and the volume of treated E. coli is bigger than that at short discharge gaps. In general, bacteria can be sterilized with an electric field of about 10⁴-10⁵ V/m [26]. In our cases with or without bubbling the intensity of the electric field is above this value, thus the electric field could affect the cell membrane.

Emission spectroscopy was used to measure the spectra of discharge in the water. The experimental setup consisted of a metal plate covered with dielectric and electrode connected to a ground with a diameter φ40mm. The needle electrode was placed above the plate electrode and N₂ gas was pumped in to the water at a gas flow rate of 2.5 L/min. The formed bubbles rise towards the surface of water. The discharge gap was set at 1 mm. The emission spectra were measured by an ICCD camera (Ryoushi-giken, SMCP–ICCD 1024 HAM-NDS/UV) and a spectrometer (Ryoushi-giken, VIS 351). A pulse generator (Tektronix, AFG 3102) was used to trigger a Marx Generator and the ICCD camera. The obtained data were transferred to a computer.

An experimental Marx generator with MOSFET switches was used as a pulsed high voltage power supply. Emission spectrum of the discharge in water showed N₂ second positive band system peaks in the ultraviolet region and OH peaks (Fig. 14). The spectrum was
obtained at a discharge voltage of -3 kV, rise time of 80 ns, pulse width of 500 ns, and discharge frequency of 1 kHz. The sterilization of bacteria could be considered as a result of the combined effects of high electric fields, UV radiation, ozone, and active radicals.

With the exception of heat resistant microorganisms, heat can also be used for sterilization. Fig. 15 shows the liquid medium temperature rise versus discharge time for short and large discharge gaps. The initial temperature of water was 20 °C. At large discharge gaps an increase of about 20 °C in the water temperature was measured. The water temperature increased only 10 °C after treatment at a short discharge gap. For both cases the water temperature does not assure the sterilization of E. coli.

The difference in the temperature rise was more than double for the treatment at large discharge gaps and using gas in comparison with the treatment at short discharge gaps.

At low voltages of discharge in bubbled water, electric discharge occurs when the space between the electrodes (mainly needle electrode tips) is filled with gas. In addition, due to heating through the Joule effect and electrolysis, the water temperature rises. The Joule effect is considered to be the cause of the heat increase when no discharge occurs.

The images representing the treated samples of E. coli at large discharge gaps and using O2 to produce bubbles in the water are shown in Fig. 16. The gas flow rate was set at 0.6 L/min. After 20 minutes of treatment,
it can be observed that there was no \textit{E. coli} in the treated sample (Fig. 16. d).

The treated samples at short discharge gap and without bubbles are presented in Fig. 16. In the treated sample for 20 minutes, it is observed that colonies of \textit{E. coli} still exist (Fig. 17. d).

IV. CONCLUSION

In this research, a low cost, low power consumption and small water disposal equipment is proposed. Fundamental examination of the low voltage discharge below 10 kV in the water was performed and the following knowledge is acquired.

1) In both discoloration and sterilization, the treatment performances are higher with gas flows and 10 mm discharge gap than that of the 0.3 mm discharge gap in the water without using any gas. This could be explained by the increase of treatment volume of solution and the churning effect.

2) The treatment performances improve when \textit{O}_2 was added into the discharge area. The indigo carmine concentration with \textit{O}_2 as added gas decreased from 20 mg/L to 0 mg/L when discharge time was 6 min and the discharge gap set at 10 mm. \textit{E. coli} density decreased 7 - digit after 20 min. of treatment when air and \textit{O}_2 were used and with a discharge gap of 10 mm. These results could be the effect of the generation of active radical species and ozone.

REFERENCES


