Residual Effects and Energy Cost of Gliding Arc Discharge Treatment on the Inactivation of *E. coli* in Water

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Abstract

The present study investigated the residual effect of gliding arc discharge (GAD) treatment on the inactivation of bacteria in a large volume of water (i.e., 20 L). Pure water and air were introduced to GAD separately, and then both the gas and water treated by plasma were sent to bacteria-containing water. Water contaminated by *Escherichia coli* (*E. coli*) was first treated by GAD for 10, 13 and 16, and 25 min and then stored for the next 4 h, during which time the *E. coli* concentration and pH were measured. In general, GAD produced the strong anti-microbial properties, a phenomenon which increased with plasma treatment time. More specifically, *E. coli* was partially inactivated (i.e., approximately 2.7-log reduction) with 16-min plasma treatment. However, *E. coli* was almost completely inactivated (over 99.9 % with 5-log reduction) during the subsequent 4-h storage period, a phenomenon that was attributed to the residual effect of the plasma treatment. The optimum plasma energy cost of the GAD treatment to inactivate *E. coli* in 20-L water in the present study was found to be approximately 0.57 kJ/L per 1-log reduction.

Key Words: Gliding arc discharge, E. coli, Bacterial inactivation, Residual effect, Energy cost of plasma treatment, Water treatment

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Nomenclature

E_d	deposited plasma energy	$k(T)$	rate constant
$V(t)$	voltage profile	E_a	activation energy
$I(t)$	current profile	R	gas constant
f	frequency	A, n	coefficients in rate constants
T	pulse period	T	plasma temperature

1. Introduction

Plasma treatment is a promising, non-chemical method for inactivation of microorganisms in water [1-4]. The inactivation effect has been attributed to the ability of plasma discharges to generate active plasma species, i.e., OH, O, O₃, H₂O₂, NO_x, UV and electric fields [5-9]. Each of these plasma species may play a role in the inactivation of microorganisms. However, most of the active species have very short half-lives on the order of μ s or less [10-13]. The exceptions to these are H₂O₂ and ozone, which have relatively long half-lives, and as a result, may be especially useful in the treatment of larger volumes of contaminated water [14, 15]. Ozone is often produced in air using a compressor equipped with dryer and cooler and then mixed with water in a two-step process [16-18]. On the other hand, H₂O₂ can be produced directly in water

through the dissociation of water molecules by plasma discharges [9, 19, 20]. Furthermore, hydrogen peroxide concentration can persist in water for relatively long durations, i.e., over a 10-min timescale [21]. The energy yield of 80 g/kWh for H_2O_2 formation was reported with a two-dimensional gliding arc discharge (GAD) with oxygen and argon gases in divergent-channel electrodes [19], whereas three-dimensional GAD used in the present study gave an energy yield of 7.3 g/kWh for H_2O_2 formation with air [21].

The GAD is able to generate a large amount of active plasma species because of its unique plasma properties and the gas flow inside the GAD generator. The GAD can be defined as an auto-oscillating periodic discharge between at least two diverging or non-diverging electrodes propelled by a gaseous flow [9], resulting in a sufficiently high degree of non-equilibrium to sustain a selective chemical process [22]. The arc discharge first ignites as thermal plasma at the locus of the smallest gap between the two electrodes. Then, the arc is forced to move downstream by a stream of gas and is convectively cooled by a stream of room-temperature gas, becoming a non-equilibrium discharge during the space-time evolution. The stream of gas in the GAD used in the present study flows through a cylindrical three-dimensional geometry, creating a reverse vortex flow (i.e., tornado) [9, 22-28].

The vortex flow regime provides excellent thermal insulation of active species from the cylindrical wall, reducing energy loss to the surroundings, thereby increasing its energy efficiency. Another benefit of the GAD in vortex flow is that the residence time of gas to be treated by plasma is relatively long, a desirable phenomenon for various chemical reactions. Thus, the rotating GAD can provide increased specific power input, reduced energy cost, and uniform treatment of gas [23, 27].

The long residence time of treatment and the high concentration of H_2O_2 in water generated by the GAD are essential to bacterial inactivation [29]. Previously, it was reported that the concentration of H_2O_2 in water was found to be over 30 ppm as determined using a peroxide test strip in the case of 120 mL/min of water injection to GAD generator [21].

In our previous study [30], a small amount of water was injected to the plasma jet coming out of the exit of the GAD generator so that water molecules could be dissociated, resulting in the recombination of hydroxyl radicals, forming H_2O_2 [31], and at the same time, reducing the pH of water [4, 30, 32]. The combined effects of H_2O_2 and low-pH water on the inactivation of bacteria in a large volume of water (i.e., 20 L) were reported to be beneficial, where a 2-log reduction in counts of colony forming units (cfu) for *E. coli* with GAD treatment of 25 min was demonstrated with a total energy cost of 8.1 kJ/L per 1-log reduction [30, 33]. Recently, the total energy cost was further reduced to 6.5 kJ/L per 1-log reduction, i.e., an improvement by approximately 20 %, where microbubble generators were used to improve the mixing between plasma-treated water and gas [33].

When the plasma-treated water is kept in a closed reservoir, the low-pH in water can be maintained for an extended period of time. Hence, it is hypothesized that the anti-microbial power of plasma-treated water [8, 24] might also be maintained over the extended period of time after the completion of the plasma treatment.

In consideration of this hypothesis, the objective of the study was to investigate the residual effects of GAD treatment on the inactivation of bacteria for a large volume of water. In addition, the present study also investigated the plasma energy cost of the GAD treatments of water contaminated with *E. coli*, including the residual benefit of anti-microbial properties of plasma-treated water.

2. Experimental Methods

The experimental setup used in the present study is illustrated in Fig. 1. The test setup consisted of three major parts: the first part comprised of two identical GAD generators, each driven by its own power supply; the second part was made of air and water-transport systems to provide controlled flows of air and water to the GAD generators; and the third part was made of a reservoir tank with two microbubble generators to store bacteria-contaminated water and to receive plasma-treated air and distilled water. The basic approach in the study was to have both air and distilled water pass through the GAD generator first and, then, to introduce plasma-treated air and water to a large volume of contaminated water at the reservoir (see Fig. 1). In order to provide a fixed airflow of 3.8 scfm to each of the two GAD generators, an air compressor together with a valve and a pressure regulator was used. The compressed air was also sent to the top of distilled water reservoir, as shown in Fig. 1, so that water could be pushed out through an exit located at the bottom of the reservoir at the uniform flow rate of 120 mL/min, which was monitored by a flow meter (FL-3839G, Omega)

Since more detailed descriptions of GAD generator have been given elsewhere [21, 30], it will be described only briefly here. Each set of two stainless steel electrodes separated by a gap of 2.5 mm was connected to a high-voltage power supply, which delivered 200 W. Since two GAD generators were used in the study, and each GAD generator was powered by its own power supply, the total power consumption was 400 W for both plasma discharges. Compressed air was tangentially introduced to the gap space between two circular electrodes through six small nozzles in the GAD generator, resulting in a reverse vortex flow. Water was also injected tangentially through another set of six small nozzles to the plasma arc jet exiting from the center hole in the circular electrodes.

Fig. 2 shows the schematic circuit diagram of the power supply used in this study, which is designed to produce alternating high voltage (HV) and current at a frequency of 33.3 kHz (B&N Inc., South Korea). The power supply mainly consisted of four components; voltage rectifier with diode bridges, capacitors, high voltage transformer, and a transistor.

Fig. 3 shows voltage and current profiles produced by the AC HV power supply, which were measured and recorded by a digital phosphor oscilloscope (TDS3014C, Tektronix). For the measurement of the current, a magnetic core current probe was utilized (CM-10-L, Ion Physics Corporation, Fremont, NH), whereas the voltage was measured using a high voltage probe (P6015A, High Voltage Probe 1000X 75MHz, Tektronix). Peak-to-peak voltage (U) and current (I) were determined to be 5.6 kV (based on settings of 1.0 V per division x 1000 for the voltage probe) and 0.75 A (based on settings of 50 mV per division x 10 A/V for the current probe). The frequency (f) of HV pulses was 33.3 kHz, a value that was determined from pulse period (T) of 30 μ s measured with the oscilloscope.

By integrating the voltage and current profiles over 1 pulse cycle, the deposited energy (E_d) into the GAD per 1 pulse cycle and per 1 s can be calculated, respectively, as:

$$E_d/\text{pulse} = \int_{t=0}^{t=2\pi} U(t) I(t) dt \quad (1)$$

$$E_d/\text{second} = \int_{t=0}^{t=2\pi} U(t) I(t) dt \times f \quad (2)$$

The values of E_d/pulse and E_d/second were approximately 8.92×10^{-4} J/pulse and 29.71 J/s.

These were used to calculate the plasma energy cost and D-value, which are given in the Result section.

The contaminated water reservoir was cleaned after each test with both sulfuric acid and tap water repeatedly and then dried in a fume hood. Then, the water reservoir was filled with 20 L

of distilled water to prepare for the next test, and the flexible tube from the GAD generator was re-connected to the reservoir.

The flow rate of water to each GAD generator was 120 mL/min at a uniform air flow rate of 3.8 scfm, an optimal condition for the present GAD generator [21, 30]. The concentrations of H₂O₂ in the plasma-treated water obtained at various test conditions of different flow rates of air and water through GAD generator were measured in the form of color changes with peroxide test strips (EMD Chemicals, Germany). Due to the limitation in collecting a sample of H₂O₂ in gas phase [21, 34], the concentration of H₂O₂ in water samples only was measured (see Fig. 4). The pH level in plasma-treated water was measured with a pH meter (HI 9125N, HANNA Instruments, Smithfield, RI). Prior to each experiment, the pH meter was calibrated at three points (i.e., pH: 4, 7, and 10) using standard-pH solutions provided by the manufacturer, and each sample was measured 5 times, from which the mean value and standard deviation were obtained. The temperature of water in the reservoir was also measured using a thermometer (52 II, Fluke) whenever pH measurement was done.

Escherichia coli (*E. coli*) was used for the present bacterial inactivation experiments. Cultures of *E. coli* were grown for 18 h at 37°C in an incubator and diluted using the most probable number method [35], yielding a range of concentrations between 10⁵ – 10⁶ colony forming units per milliliter (cfu/mL) as initial test conditions. The cfu measurements in all water samples were conducted using an aerobic heterotrophic plate count (HPC) method by the serial dilutions of samples [36, 37]. For bacterial inactivation tests, cultures of *E. coli* were added to the reservoir filled with distilled water of 20 L. Immediately after adding *E. coli* to water, the water reservoir was continuously stirred using an electrical stirrer (Eurostar/IKA, Staufen,

Germany) at 650 rpm to disperse *E. coli* uniformly in the reservoir. After stirring water for 5 min, 50-mL samples were collected in sterile tubes for cfu counts.

Prior to plasma inactivation tests, a control test (see Fig. 5) was performed with an initial bacterial count of 10^5 cfu/mL in the reservoir holding 20 L of contaminated distilled water, where an additional 2.8 L of distilled water was introduced through the GAD generator at a flow rate of 2×120 mL/min for 16 min as per procedures but with the plasma power turned off. The water was then kept at the reservoir for the next 4 h, during which time the cfu numbers in water was determined.

In cases with plasma treatment, the initial concentration of *E. coli* in the reservoir varied from 10^5 to 10^6 cfu/mL. Plasma treatments were conducted for 10, 13, 16 and 25 min, during which both plasma-treated water and air were continuously delivered to the reservoir. The initial volume of the contaminated water in the reservoir was 20 L in all tests. For the cases of 10, 13, 16 and 25-min treatments, approximately 2.4, 3.1, 3.8 and 6 L of plasma-treated water, respectively, were added to the reservoir during the treatment.

Two water samples of 50 mL each (including at $t = 0$, i.e., before treatment) were collected for cfu and pH measurements during the plasma treatment period, and 8 additional samples of 50 mL each were also collected from the contaminated water reservoir every 30 min (at $t = 30, 60, 90, 120, 150, 180, 210$ and 240 min) during the storage period (see Fig. 6A – 6D). 1-mL water sample for cfu measurement at each time period was collected using a sterile pipette from 50-mL water sample, which was continuously stirred with a vortex mixer (Fisher Scientific: Genie 2). Each 1-mL sample water was spread on Brain Heart Infusion Agar plate (Fisher Scientific) inside a clean bench within 5 min after collecting the 50-mL water sample from the reservoir. All agar plates containing water samples were dried for 30 min at room temperature

inside the clean bench and incubated for 18 h at 37°C [38, 39]. The number of colony forming units was determined using a colony counter (Digital Colony Counter/HYC-560, Korea), which has a measurement error (i.e., sensitivity) of 7.9 % in the measurement range for 10^2 cfu or greater. Prior to performing cfu counts of *E. coli* on agar plates after incubating, the HPC images were recorded with a digital camera. The above measurements of the cfu of *E. coli* were repeated three times for each water sample, from which both the mean value of cfu/mL and the standard deviation were obtained and shown in Figs. 5 and 6.

3. Results

Fig. 4 shows the results of H_2O_2 concentrations in water collected from the connecting line between GAD generator and water reservoir tank at 3 different air flow rates and 5 different water flow rates. As the water flow rate varied from 140 to 20 mL/min, the concentration of H_2O_2 in the plasma-treated water increased from 20 to 100 ppm. The maximum concentration of H_2O_2 of 100 ppm was obtained at the lowest water flow rate of 20 mL/min. In the present study, a uniform water flow rate of 120 mL/min was used for all bacterial inactivation tests because the large water flow rate provided a minimal temperature increase in plasma-treated water [21, 30].

Fig. 5 shows photographic HPC images for the control case, where the control sample (i.e., without plasma treatment) maintained its initial cfu number of 10^5 per mL during 16-min period of the sham treatment under identical test conditions (i.e., with the same water and air flow rates through GAD generator), and the cfu/mL number was maintained within ± 15 % during the next 4-h storage period. Significant levels of cultured *E. coli* were observed in the HPC images in water collected from the reservoir in the control test when the water was not treated with plasma.

Fig. 6A – 6C show photographic HPC images obtained in the cases of 10, 13, and 16-min plasma treatments with an initial *E. coli* concentration of 10^5 cfu/mL. Fig. 6D shows the corresponding images for the case of 25-min plasma treatment with an initial *E. coli* concentration of 10^6 cfu/mL.

The energy cost for the present plasma treatment was determined using the values of the deposited energy (E_d) obtained from the integration of the voltage and current profiles as:

$$\text{Energy cost/L (kJ/L)} = 2 \times (E_d/\text{s}) \times \text{treatment time (min)} \times 60 \text{ s} / 20 \text{ L} \quad (3)$$

where the constant 2 is for two GAD generators used in the present study.

For the cases of 10, 13, 16, and 25-min plasma treatment, the plasma energy costs were determined as 1.78, 2.31, 2.85, and 4.56 kJ/L, respectively.

The cfu data for the cases with plasma treatments were consistently reduced with increasing plasma treatment time (i.e., plasma intensity). In addition, the pH significantly reduced with the plasma treatment and remained at a reduced level during the subsequent storage period.

Fig. 6A also shows the cfu counts for the case of 10-min plasma treatment, followed by 4-h storage. The value of cfu/mL decreased to 5130 ± 110 (i.e., 1.5-log reduction) at the end of the plasma treatment, which was further reduced to 35 ± 14 cfu/mL (i.e., 2.1-log reduction) at the end of the 4-h storage period, resulting in a total 3.6-log reduction.

Fig. 6B and 6C show the cfu results for the cases of 13 and 16-min plasma treatments, respectively. The results in Fig. 6B show 2-log reduction in cfu/mL at the end of the plasma treatment with an additional 2-log reduction during the subsequent storage period, resulting in a total 4-log reduction. The results in Fig. 6C show nearly 2.7-log reduction at the end of the 16-

min plasma treatment with an additional 2.3-log reduction during the subsequent storage period, resulting in a total 5-log reduction at the plasma energy cost of 2.85 kJ/L.

Fig. 6D shows the cfu counts for the case of 25-min plasma treatments with an initial condition of 10^6 cfu/mL. At the end of the plasma treatment, there was 3.5-log reduction followed by an additional 2.5-log reduction after 2-h storage, resulting in a total 6-log reduction, a better reduction performance than the case of the 16-min treatment, but at a significantly increased plasma energy cost of 4.56 kJ/L.

Using the data on the log-reduction, the plasma energy cost including the residual effects in plasma-treated water becomes 0.50, 0.58, 0.57 and 0.74 kJ/L/1-log reduction for the cases of 10, 13, 16, and 25-min plasma treatment, respectively. This residual effect of the anti-microbial properties significantly improves the plasma energy cost. The best plasma energy cost to inactivate *E. coli* in 20-L water in the present study was found to be approximately 0.57 kJ/L per 1-log reduction in cfu/mL of *E. coli* corresponding to the 16-plasma treatment. Accordingly, the case with the 16-plasma treatment was considered the best performance in terms of plasma energy cost and bacterial inactivation capability among the tests conducted in the present study.

Fig. 7 shows the variations in *E. coli* concentrations for all cases in the present study, including the control case (i.e., no treatment), whereas Fig. 8 provides the variations in pH during the plasma treatment and subsequent storage periods. The temperature increase in the plasma-treated water was found to be less than $6\text{ }^{\circ}\text{C} \pm 0.05\%$ after 25-min plasma treatment. However, since the plasma-treated water was introduced to a large volume of water in the reservoir, the temperature rise in the contaminated water in the reservoir was negligible in this study.

Comparing the bacterial inactivation results with the pH variations in all plasma-treated cases, the reductions in pH and *E. coli* concentration followed similar trends. Pooling together all of the test data across 5 arms of this study (10, 13, 16, 25 min of plasma treatment and no treatment control), reflecting a range of treatment durations and storage times, a moderately strong correlation was observed between pH and *E. coli* concentration using Student's t-test ($R=0.52$, $p=0.0004$). For the plasma treatment cases with 10, 13, 16 and 25 min, the mean pH values in the contaminated water in the reservoir decreased from the initial pH of 6.13 ± 0.03 to 3.95 ± 0.03 , 3.83 ± 0.01 , 3.68 ± 0.02 and 3.62 ± 0.00 , respectively, at the end of the plasma treatment, which were maintained almost constant during the storage period. Since the reductions in *E. coli* concentration almost coincided with those in pH in the contaminated water, one can attribute the residual effects of the plasma treatment to the low-pH of water.

4. Discussion

Our previous studies reported that the GAD generator produced H_2O_2 in plasma-treated water [21, 30]. Experimental results in the present study indicate that the pH in the treated water was reduced to as low as 3.62 in a large volume of water. The phenomenon of the formation of acidic water can be attributed to H_3O^+ ions and nitric acid (HNO_3) generated from the plasma discharge [40].

When water is directly exposed to GAD, the following reactions occur with the dissociation of water molecules [4, 9, 20, 41]:

Then, H₂O₂ is formed from the recombination of hydroxyl radicals [9, 19, 20].

In the overall set of reactions, the concentration of H₂O₂ in water increases with plasma treatment [21, 40]. A significant pH drop observed in the present study can be attributed to positive charges (M⁺) created in the plasma discharge that reach the water molecules and exchange charges with the water molecules, resulting in the creation of H₃O⁺ ions and OH radicals, both playing an important role in sterilization [17, 19, 24, 40] through Equations (4-4) to (4-5).

When air is the carrier gas, nitrogen oxides can be formed from gas phase reactions of dissociated nitrogen and oxygen [9, 40-45]

The nitrogen dioxide (NO₂) affects the pH of the water through the formation of acids and ions. The reaction between NO₂, see Equation (5-4), and hydroxyl radicals OH, see Equations (4-1) to (4-4), can generate HNO₃, which results in acidic water [40, 41, 43, 46].

All chemical reactions and the rate constants in the form of an Arrhenius equation, which depend on plasma temperature (T), are listed in Table 1 in the following format:

$$k(T) = A (T / 298 \text{ K})^n \exp(-E_a / RT) \quad (6)$$

where E_a is the activation Energy, R is the gas constant, and A and n are coefficients.

Table 1 presents these chemical reactions grouped by the temperature range. The four chemical reactions are shown in the high-temperature plasma zone at $T = 2,000$ K or above. The OH radicals shown in the reactions of (4-1) and (4-2) are normally associated with the dissociation of the water vapor, as high energy electrons (radiation) with 5.1 eV/mol necessary for H_2O dissociation pass through the water, yielding hydrogen and hydroxyl radicals [9, 20]. The other reactions of (5-1) and (5-2) in the high-temperature plasma zone explain the vibrational excitation of N_2 and O_2 molecules by the high electron energies between 1 and 3 eV/mol [9, 47]. Distilled water was introduced to the above plasma arc jet in the present study (see Fig. 2), so that the vibrational excitation occurred first in the high-temperature zone, immediately followed by the dissociation of water molecules.

When the plasma jet and treated water exiting from the high-temperature plasma zone of GAD generator, both of them become cooled as they move in the connecting pipe to the reservoir. Subsequently, the excited species (ie., OH, N and O radicals) are rapidly quenched and then recombined to H_2O_2 and HNO_3 through the reactions from (4-3) to (4-5) and (5-3) to (5-5) in the low-temperature plasma zone [9, 48].

A number of studies on the antimicrobial effects of plasma have indicated OH and NO as the effective species [5-9]. The generation of these two reactive species depends on the humidity of air injected into the GAD [47, 49]. For example, the concentration of OH generated by the GAD can increase much higher than that of NO as the humidity of the carrier gas increases. The OH formed from the dissociation of water can lead to the formation of H_2O_2 due to the recombination of OH as explained above. Hence, when pure water is injected into the

plasma jet exiting from the GAD, the concentration of H_2O_2 becomes much higher than that of nitric acid (HNO_3) formed from the reaction of OH and NO_x . [40, 47]

The plasma water treatment system in the present study increased H_2O_2 concentration in water and decreased pH to a low level of 3.6. These two effects are proposed as the main mechanism for *E. coli* inactivation by the plasma treatment. Note that H_2O_2 itself is not a strong oxidizer. Also, note that acidic water alone is not a strong oxidizer. However H_2O_2 in the presence of acidic water becomes a very strong oxidizer and an effective tool for the inactivation of microorganisms, making a direct impact on the outer membrane of microorganisms due to the peroxidation of a cell membrane [1, 8]. In the present study, as the plasma-treated water was pumped to a large volume of bacteria-filled water, the optimal bacterial inactivation was achieved with 16-min treatment, resulting from the combined effect of the high concentration of H_2O_2 and low-pH level in water. In other words, the plasma treatment in the case of 25-min treatment gave a slightly better performance in the bacterial inactivation than the case of 16-min treatment, but at a significantly increased energy cost. Further inactivation was obtained through the residual effect of the plasma treatment as manifested by reduced cfu values by letting the plasma-treated water sit for several hours.

There have been a number of studies on *E. coli* inactivation by other types of plasma such as corona, capillary and DBD discharges directly in water [9, 50-56]. D-value, which can be defined as the energy cost per L per 1-log reduction (i.e., 90 %), varied from 3.3 to 33.3 kJ/L per 1-log reduction [52-54]. The D-value obtained with a 3-dimensional GAD in the present study in 20-L water was 0.57 kJ/L per 1-log reduction for 16-min treatment case, indicating that the present GAD method of bacterial inactivation showed at least 5.7 times more energy-efficient than those systems reported in the literature [9, 50-56].

5. Conclusions

The present study reports the efficacy of plasma water treatment with a 3-dimensional GAD on the inactivation of contaminated water. Plasma treatment decreased the pH of water to 3.62 as GAD produced H_3O^+ ions and HNO_3 . The efficacy of active plasma species such as H_2O_2 and low-pH on the inactivation of a large volume of water (i.e., 20 L) has been demonstrated with plasma treatment of 10, 13, 16 and 25 min. Although the plasma treatment provided only partial inactivation at the end of the plasma treatment, remaining *E. coli* in water was further inactivated during the subsequent storage period by the residual effect of H_2O_2 and low-pH. The synergetic effect of H_2O_2 and low-pH in water is enhanced by a powerful residual effect during the post-treatment storage period, where no additional energy is consumed. Additional studies are needed to better understand the scale-up issue in the treatment of a significantly large volume of water.

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List of Figures

Fig. 1. Experimental setup for the present plasma-water-treatment system using gliding arc discharge; HV = high-voltage power supply, MBG = microbubble generator, PG = pressure regulator.

Fig. 2. Circuit diagram of a power supply connecting with gliding arc discharge generator; 1 = Voltage rectifier with diode bridge, 2 = Capacitors, 3 = Transistor to control primary coil, 4 = Primary coil, 5 = Secondary H.V. coil, 6 = Electromagnetic interference filter, Ch-1 = Voltage measurement with HV probe (x 1000), Ch-2 = Current measurement with magnetic core probe (x 10 A/V)

Fig. 3. Profiles of voltage and current used to generate the gliding arc discharge in the present study. (U = voltage, I = current, T = 30 μ s, f = 33.3 kHz)

Fig. 4. Variations of H₂O₂ concentrations in water at various air flow rates and water injection flow rates through gliding arc discharge generator.

Fig. 5. Control test: Results of E. coli inactivation experiment with sham plasma treatment with water injection flow rate of 120 mL/min and air flow rate of 3.8 scfm.

Fig. 6A. Plasma treatment for 10 min: Results of E. coli inactivation experiments and pH variations with an initial water volume of 20 L at reservoir with 1.78 kJ/L of plasma energy consumption

Fig. 6B. Plasma treatment for 13 min: Results of E. coli inactivation experiments and pH variations with an initial water volume of 20 L at reservoir with 2.31 kJ/L of plasma energy consumption.

Fig. 6C. Plasma treatment for 16 min: Results of *E. coli* inactivation experiments and pH variations with an initial water volume of 20 L at reservoir with 2.85 kJ/L of plasma energy consumption.

Fig. 6D. Plasma treatment for 25 min: Results of *E. coli* inactivation experiments and pH variations with an initial water volume of 20 L at reservoir with 4.56 kJ/L of plasma energy consumption.

Fig. 7. Results of *E. coli* inactivation experiments with various treatment times (plasma energy) in an initial water volume of 20 L at reservoir.

Fig. 8. Results of pH changes experiments with various treatment times (plasma energy) in an initial water volume of 20 L at reservoir.

Table 1. Chemical reactions and rate functions [9, 20, 41, 44, 45, 46]

Reaction Number	Reactions	Temp. range (K)	Reaction Order	Rate constant
1-1	$e + H_2O \rightarrow H + OH + e^-$	2,000 - 6,000	2	$5.8 \times 10^{-9} \exp(-440 \text{ kJ/RT})$
1-2	$e + H_2O \rightarrow H^- + OH$	2,000 - 6,000	2	$5.8 \times 10^{-9} \exp(-440 \text{ kJ/RT})$
1-3	$M^+ + H_2O \rightarrow H_2O^+ + M$	≈ 300	2	2.2×10^{-9}
1-4	$H_2O^+ + H_2O \rightarrow H_3O^+ + OH$	≈ 350	2	0.5×10^{-9}
1-5	$OH + OH + M \rightarrow H_2O_2 + M$	200 -1,500	2	$1.51 \times 10^{-11} (T/298)^{-0.37}$
2-1	$N_2 + e \rightarrow 2N + e^-$	$\approx 5,000$	2	2×10^{-11}
2-2	$O_2 + e \rightarrow 2O + e^-$	$\approx 20,000$	2	$10^{-10} - 10^{-9}$
2-3	$N + O + M \rightarrow NO + M$	196 - 327	3	$7.31 \times 10^{-33} \exp(1.12 \text{ kJ/RT})$
2-4	$NO + O + M \rightarrow NO^2 + M$	298	3	1.0×10^{-31}
2-5	$NO_2 + OH \rightarrow HNO_3$	200 - 400	2	4.0×10^{-11}
<div>- Rate constant units: First order: s^{-1} Second order: $cm^3 \text{ molecule}^{-1} s^{-1}$ Third order: $cm^6 \text{ molecule}^{-2} s^{-2}$</div> <div>- Rate constant: $k(T) = A (T / 298 \text{ K})^n \exp(-E_a / RT)$ - $R = 8.314472 \times 10^{-3} \text{ kJ mole}^{-1} \text{ K}$</div>

Fig. 1.

Fig. 2.

Fig. 3.

Fig. 4.

Control (No treatment)						
Time (min)	0	16	+30	+60	+120	+240
pH	6.15	6.03	6.02	6.04	6.05	6.09
cfu No./ml	10^5	10^5	10^5	10^5	10^4	10^5
HPC image	Water injection period			Storage period

Fig. 5.

10 min of plasma treatment and subsequent storage					
	Control	Treatment	Post plasma treatment		
Time (min)	0	10	+30	+60	+90
pH	6.15	3.93	3.93	3.93	3.93
cfu No/ml	10^5	5130 ± 110	2035 ± 75	678 ± 1	357 ± 3
HPC image
Post plasma treatment					
Time (min)	+120	+150	+180	+210	+240
pH	3.96	3.96	3.97	3.98	3.98
cfu No/ml	263 ± 4	108 ± 5	62 ± 6	42 ± 2	35 ± 14
HPC image

Fig. 6A.

13 min of plasma treatment and subsequent storage					
	Control	Treatment	Post plasma treatment		
Time (min)	0	13	+30	+60	+90
pH	6.15	3.84	3.83	3.83	3.83
cfu No/ml	10^5	880 ± 60	343 ± 47	195 ± 7	80 ± 3
HPC image
Post plasma treatment					
Time (min)	+120	+150	+180	+210	+240
pH	3.83	3.83	3.84	3.83	3.83
cfu No/ml	58 ± 10	52 ± 5	20 ± 5	15 ± 5	10 ± 3
HPC image

Fig. 6B.

16 min of plasma treatment and subsequent storage					
	Control	Treatment	Post plasma treatment		
Time (min)	0	16	+30	+60	+90
pH	6.10	3.67	3.67	3.68	3.68
cfu No/ml	10^5	339 ± 51	219 ± 24	54 ± 13	35 ± 10
HPC Image
Post plasma treatment					
Time (min)	+120	+150	+180	+210	+240
pH	3.69	3.69	3.69	3.69	3.69
cfu No/ml	10 ± 4	6.5 ± 2.5	4.5 ± 1	4 ± 1	2 ± 0
HPC image

Fig. 6C.

25 min of plasma treatment and subsequent storage				
	Control		Treatment	
Time (min)	0		25	
pH	6.10		3.62	
cfu No / ml	10 ⁶		542 ± 79	
HPC image
Post plasma treatment				
Time (min)	30	60	90	120
pH	3.62	3.62	3.62	3.62
cfu No / ml	203 ± 4	26 ± 6	6 ± 2	3 ± 2
HPC image

Fig. 6D.

Fig. 7.

Fig. 8.