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Residual effects and energy cost of gliding arc discharge treatment on the inactivation of *Escherichia 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.

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, O_3, H_2O_2, 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 μs or less [10–13]. The exceptions to these are H_2O_2 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, H_2O_2 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

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Nomenclature

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

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 *Escherichia 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 75 MHz, 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 × 1000 for the voltage probe) and 0.75 A (based on settings of 50 mV per division × 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 μs 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:

$$E_d/\mathrm{pulse} = \int_{l=0}^{l=2\pi} U(t)l(t)dt \tag{1}$$

$$E_d/\mathrm{second} = \int_{l=0}^{l=2\pi} U(t)l(t)dt \times f \tag{2}$$

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 H_2O_2 in the