Pulsed arc electrohydraulic discharge (PAED) offers concurrent treatment of chemical and microbial target compounds in water by several mechanisms. Here, Escherichia coli, Bacillus subtilis spore, and MS2 bacteriophage inactivation by PAED were investigated using two bench-scale reactors (0.7 and 3 L). A plasma channel was created between a pair of iron electrodes set 0.5 mm apart in these reactors. Pulsed applied voltage was supplied at approximately 0.3 kJ/pulse (~100 μsec). In the 0.7-L reactor, median E. coli, B. subtilis, and MS2 reductions of 2.4-, 4.6-, and 3.7-log, respectively, were observed after approximately 80 seconds of treatment in water with a conductivity of 14.7 mS; reductions of 2.8-, 4.0-, and 3.7-log, respectively, were observed in treated drinking water filter effluent (conductivity of 610 μS). In the 3-L reactor, at a conductivity of 500 to 600 μS, 1.9- and approximately 0.9-log median reductions of 3.7-log, respectively, were observed in treated drinking water filter effluent (conductivity of 610 μS). In the 3-L reactor, at a conductivity of 4.3 mS, median E. coli reductions of 0.5-log were achieved after 50 pulses (400 sec). Further work is necessary to elucidate the primary mechanism(s) of disinfection acting in the PAED system and how they relate to the reactor design, applied power requirements, and disinfection of specific target microorganisms.

**Key words:** pulsed arc electrohydraulic discharge, disinfection, E. coli, pathogens, drinking water, small systems

**Introduction**

Many drinking water systems are challenged with difficult-to-treat target compounds such as organics (e.g., NDMA [N-nitrosodimethylamine]) and pathogens (e.g., Cryptosporidium, viruses, etc.). To be effectively removed from drinking water supplies, many of these recalcitrant compounds require specialized, and often target-specific, treatment technologies. The necessity of multiple treatment technologies can be quite costly, especially for small systems. Pulsed arc electrohydraulic discharge (PAED) can effectively treat many recalcitrant organic compounds in water (Angeloni et al. 2006; Locke et al. 2006). Relative to currently available technologies, it has the potential to treat a wide range of target compounds effectively and concurrently, making it a particularly applicable treatment technology for small systems. PAED is a direct plasma technology that may treat water by several mechanisms including: pressure waves, ultraviolet (UV) irradiation, radical reactions, electron processes, ionic reactions, and thermal dissociation (Karpal Vel Leitner et al. 2005). The advantage of utilizing direct plasma technologies such as PAED is that they are capable of simultaneously taking advantage of all of these mechanisms.

It is important to contrast PAED systems with pulsed power electrohydraulic discharge systems (PPED). PAED employs the rapid discharge of stored electrical charge across a pair of submerged electrodes to generate electrohydraulic discharges forming a local plasma region. PAED systems operate at a frequencies of $10^2$ to $10^5$ Hz with peak currents above $10^5$ A and voltage rise occurring on the order of microseconds with operating voltages of 1 to 10 kV. An arc channel generates strong pressure waves with a cavitation zone containing plasma bubbles and transient supercritical water conditions. These systems can generate strong UV radiation and high radical densities, which have been observed to be short-lived in the cavitation zone (Chang et al. 2007). In contrast, PPED systems operate at frequencies of $10^3$ to $10^4$ Hz with peak currents in the range of $10^2$ to $10^3$ A. The voltage rise occurs on the order of nanoseconds with $10^2$ to $10^3$ kV (Chang et al. 2007). These systems generate strong pressure waves and electromagnetic pulses with weak UV radiation. These fundamental differences are due to the power supplies used and result in differing availability of the various inactivation mechanisms (Chang et al. 2007). Reviews of plasma technologies, including pulsed power systems, and the potential application of PAED for the treatment of microbial and chemical target compounds in drinking water are provided elsewhere (Emelko et al. 2003; Chang et al. 2007).

In the past, the obstacle that has precluded the application of direct plasma techniques for drinking water treatment has been cost-prohibitive power sourcing. A recent development in pulsed power supply configurations has allowed PAED to be powered from...
a 110-V source. This could ultimately lead to decreased capital and operating costs relative to other commercially available water treatment technologies such as UV systems, which are also plasma technologies because UV emissions are generated from plasma inside UV lamps. For example, low pressure UV lamps are referred to as glow discharge plasma sources and medium pressure UV lamps are barrier discharge plasma sources, regardless of any coatings the lamps may have. Preliminary investigations conducted with a limited number of target compounds have indicated that effective water treatment with PAED utilizes less than 50% of the kilowatt-hours required by other plasma technologies (e.g., UV systems) for equivalent levels of treatment (Uchida et al. 2002). While formal efficiency analyses comparing PAED with UV and ozonation systems cannot be conducted until disinfection mechanisms and optimal reactor design have been established, a general comparison is possible. Conventional lamps for low pressure and medium pressure UV systems typically cannot exceed electrical-UV conversion efficiencies of 35 and 60% respectively (Duley 1996). The majority of commercially available ozonation systems operate in the range of 80 to 200 g of O3 per kWh (Duley 1996). Moreover, the efficiency of the power supply for both types of systems is typically between 40 and 70% (Chang 2001). In contrast, a PAED system delivering combinations of UV radiation, radical formation, thermal emission, and pressure wave formation will not have such limitations because the majority of the input power will be converted in the water matrix directly to the inactivation mechanisms (e.g., pressure wave formation, UV radiation, radical formation, thermal emission, etc.) when properly optimized power supplies are used. This occurs because the PAED system is a closed system; therefore, the energy conservation law of thermodynamics applies.

To provide a preliminary assessment of PAED’s ability to disinfect drinking water and wastewater, Escherichia coli, Bacillus subtilis, and MS2 bacteriophage inactivation by PAED were evaluated in the present investigation. E. coli was selected to bridge previously reported direct plasma work with PPED (Ching et al. 2001) and PAED (Emelko et al. 2003; Lee et al. 2004) treatment systems. The relative ease of disinfection of E. coli, as compared with protozoan pathogens such as Cryptosporidium parvum, is commonly recognized. E. coli disinfection has been achieved with several treatment technologies, including UV radiation (Harris et al. 1987). Many strains of E. coli have demonstrated an ability to repair after irradiation with low- and medium-pressure UV radiation (Harris et al. 1987; Zimmer and Slawson 2002). E. coli inactivation with electrohydraulic discharges was first reported by Ching et al. (2001).

The inactivation of aerobic spores of B. subtilis and MS2 bacteriophage was also investigated. Spores of B. subtilis have been used historically as surrogates for the inactivation of Cryptosporidium by ozonation. Providing a comparison with UV treatment, B. subtilis require germicidal fluences of UV at levels 2 to 3 times higher than those required for Cryptosporidium inactivation (Hijnen et al. 2002). Given that UV treatment has limited efficacy against viruses, a preliminary investigation of MS2 bacteriophage inactivation was also conducted herein because MS2 is a well-studied surrogate parameter for virus inactivation during drinking water treatment.

Experimental Setup

E. coli and B. subtilis Inoculum Preparation

Escherichia coli (ATCC 11229, Manassas, Va.) were grown in nutrient broth (Sigma-Aldrich Canada Ltd., Oakville, Ont.) at optimal conditions (37°C while shaken at 225 rpm). To utilize the environmentally relevant growth phase of E. coli, a culture was grown and collected during the stationary phase of growth. This was confirmed using optical density measurements at 600 nm. After 20 to 23 hours of growth, the E. coli suspension was centrifuged at 400 x g for 8 minutes (Sorvall FA-Micro, Dupont Canada, Mississauga, Ont.). The supernatant was drawn off using aseptic technique, and the pellet was resuspended in 0.01 M phosphate buffered saline ([PBS], 24.72 g of Na2HPO4, 3.60 g of NaH2PO4·H2O, and 170.0 g of NaCl, pH 7.4) to yield a final concentration of approximately 10⁷ colony forming units (cfu)/mL. A portion of this stock suspension was removed and serially diluted in 0.01 M PBS to determine the initial E. coli concentration. After PAED treatment, the E. coli concentration in the treated water was determined by collecting the entire volume of water from the reactor. Each of the samples collected during the experiments was processed in duplicate or triplicate. Samples drawn at regular intervals were held for less than 24 hours at 4°C prior to plating, incubation, and counting.

Endospores of B. subtilis (ATCC 6633) were obtained from Raven Biological Laboratories, Inc. (Omaha, Nebr.) as a 10 mL 40% ethanol in deionized water suspension containing approximately 10⁹ endospores per mL. B. subtilis endospores were stored in the dark at 4°C until use. From a pure stock suspension, a streak plate was made and allowed to grow for 24 hours at 37°C. After the incubation period, the plates were then stored in a refrigerator at 4°C to place the colonies in stasis. Once a week, the plates were removed from the refrigerator and an inoculation loop was used to transfer a colony onto a fresh nutrient agar plate repeating the process. This maintenance process was used to keep the stock colony growing by providing a new source of nutrients for the bacteria.

B. subtilis were in vegetative cell form on the nutrient agar plates and in spore form during the experiments. To achieve sporulation, an inoculation loop was used to isolate a colony on the original nutrient agar plates and to transfer it to a nutrient agar plate that contained 0.3 g/L of MnSO4 (BDH Inc., Toronto, Ont.). Manganese sulphate was added to the agar to encourage sporulation.
The suspension was heated to 60°C, described by Rice et al. (1996) with one minor deviation; undergone during disinfection. By plating and incubation, regardless of the damage this procedure accounted only for cells that reproduce. The stock suspension was then centrifuged at 10,000 × g for 10 minutes and washed 2 to 3 times with sterilized deionized water. This process allowed the spores to collect near the bottom of the bottle and the vegetative debris to be washed off the surface. Sterilized water was used to wash the resulting pellet by gently swirling it over the pellet. After the rinsing cycle, the suspension was again centrifuged. Spore staining using the Shaeffer-Fulton method was used to verify that the stock suspension was primarily in the spore form (Prescott et al. 1990). The spores appeared green under the microscope while the vegetative cells appeared pink.

**E. coli and B. subtilis Enumeration**

All samples were collected in sterile glass bottles using aseptic technique. Initial and treated *E. coli* concentrations were determined using serial dilutions and the membrane filtration technique specified in Standard Method 9222 (APHA et al. 1999). In this protocol, 0.45-μm nominal pore size filters (GN-6, Pall Life Sciences, Ann Arbor, Mich.) with a diameter of 47 mm were used to filter each sample. mTEC agar (Becton, Dickinson, and Company, Sparks, Md.) was used to grow the colonies after membrane filtration, as specified in Standard Method 9213D (APHA et al. 1999). The mTEC agar is a selective medium that isolates for *E. coli* bacteria. The mTEC agar was sterilized in the autoclave prior to use. The use of mTEC agar was necessary to quantify *E. coli* separately from *B. subtilis* because both organisms were treated concurrently during the experiments. The number of colony forming units was counted after incubation in the dark at 35°C for 2 hours and 44.5°C for 22 hours. This procedure accounts only for cells that reproduce by plating and incubation, regardless of the damage undergone during disinfection.

*B. subtilis* was enumerated using the method described by Rice et al. (1996) with one minor deviation; the suspension was heated to 60°C rather than 80°C as described by Francis et al. (2001). This method generally consisted of filtration of samples onto 47-mm, 0.45-μm gridded cellulose acetate membranes (Pall Gelman Corporation #66278, Ann Arbor, Mich.) and growth at 37°C for 24 hours on plates of nutrient agar with trypan blue (0.015 g/L). Spores were identified by their blue color.

**MS2 Bacteriophage Stock Preparation and Enumeration**

MS2 is a male-specific, unenveloped, and single-stranded RNA phage with 31% nucleic acid content. It has a diameter of approximately 25 nm, and a relatively low isoelectric point of 3.9 (Zerda 1982, as cited in Gerba 1984). The protein coating of MS2 is relatively hydrophobic (Shields 1986) and sensitive to interfacial forces that appear to cause its inactivation (Thompson and Yates 1999). The stock suspension of MS2 (ATCC 15597-B1, American Type Culture Collection, Manassas, Va.) was grown by adding 1 mL of MS2 in nutrient broth into a 200-mL culture of log phase *E. Coli* ATCC 15597 (American Type Culture. Collection, Manassas, Va.) that had been grown for 4 to 5 h at 37°C. After overnight (12 h) incubation, the *E. coli/MS2* suspension was centrifuged at 1,100 × g for 20 minutes. The supernatant was then filtered through a 0.2 μm filter (ZAPCAP-S Plus, Scheicher and Schuell, Dassel, Germany). The filtrate was stored at 4°C in sterile 250-mL glass flasks until use. MS2 was quantified with its bacterial host by the plaque-forming unit (pfu) assay using the double layer agar method of Adams (1959). Each sample was analyzed in duplicate. Countable numbers of 30 to 300 pfu per plate were targeted (You et al. 2003). The coefficient of variation in the plating was found to be consistently less than 10%.

**Experimental Apparatus Used During the 0.7-L Batch-Scale Experiments**

During the first phase of experimentation, a 0.3-kJ spark gap type PAED power supply (Yamatake et al. 2006) and a 0.7-L cylindrical batch reactor were utilized. The reactor was equipped with two adjustable arc electrodes located coaxially at the centre of the cylindrical reactor vessel, as discussed by Lee et al. (2004). Pulsed arc discharges were generated between the two iron rod electrodes by a spark gap type power supply with a fixed output of 0.3 kJ per pulse with a 2.2 kV charging voltage, a peak current of 3.5 kA, and peak power of 2.4 MW. During these preliminary experiments, 0.7 L of PBS at pH 7.4 with a conductivity of 14.7 mS (representing wastewater) or treated surface water (filter effluent with a conductivity of 14.7 mS) were placed within the reactor. *E. coli*, spores of *B. subtilis*, and MS2 bacteriophage were then suspended in the water inside the reactor. Water samples were collected from within the PAED reactor after 0, 6, 18, and 30 discharges, which corresponded to detention times of approximately 0, 15, 60, and 80 seconds respectively. Initial organism concentrations (at time = 0 s, corresponding to 0 discharges) were measured in duplicate before each experiment and were used to calculate microbe inactivation. Three replicate experiments were conducted to determine *E. coli*, *B. subtilis*, and MS2 inactivation as a function of PAED treatment time (corresponding to a specified number of electrohydraulic discharges for a given reactor configuration). Each sample (NSF International 2000). The plates were incubated for one week at 37°C or until sporulation of the culture was greater than 80%. A spore stain and/or a wet mount slide were used to check for the percentage of cells that were in the spore form. This method was modified as described below from Nicholson and Setlow (1990). After the one-week incubation period, the plates were rinsed with 10 to 15 mL of ultrapure sterilized water (MilliQ). To make the stock suspension, the cells were harvested from the surface of the agar by using sterilized water and rinsing into a common sterilized bottle. Once the agar plates were rinsed off, the stock suspension was heat shocked in a water bath at 80°C for 10 minutes to eradicate any remaining vegetative cells. The stock suspension was then centrifuged at 10,000 × g for 10 minutes and washed 2 to 3 times with sterilized deionized water. This process allowed the spores to collect near the bottom of the bottle and the vegetative debris to be washed off the surface. Sterilized water was used to wash the resulting pellet by gently swirling it over the pellet. After the rinsing cycle, the suspension was again centrifuged. Spore staining using the Shaeffer-Fulton method was used to verify that the stock suspension was primarily in the spore form (Prescott et al. 1990). The spores appeared green under the microscope while the vegetative cells appeared pink.
was processed in duplicate or triplicate. Reagent blanks (only PBS or only treated surface water) were analysed after each set of electrohydraulic discharge experiments to validate that the reactor cleaning procedure prevented carryover of microorganisms between experiments.

**Experimental Apparatus Used During the 3-L Batch-Scale Experiments**

The experimental apparatus consisted of a 0.3-kJ spark gap type PAED power supply, just as the one utilized with the 0.7-L batch reactor from the previous experiments, and a 3-L cylindrical batch reactor as shown in Fig. 1. Pulsed arc discharges were generated between two iron rod electrodes set 0.5 mm apart located eccentrically in the reactor. The power supply provided pulses with a charging voltage of approximately 3 kV per pulse (~100 μsec pulse duration, pulse frequency 0.4 to 0.8 Hz).

As described in detail elsewhere (Yamatake et al. 2006), applied voltages and discharge currents were measured by means of a high voltage probe (Tektronix, P6015A) and a current probe (Ion Physics Corporation, Model CM-01-L) with a digital oscilloscope (Tektronix, TDS 420A). The pulsed arc discharges also generate pressure waves, which represent one potential disinfection mechanism. Pressure measurements were collected simultaneously at four different locations (#1 to #4) along the walls of the reactor using Piezoelectric transducers (Columbia Research Laboratories Inc., Model 4103). Pressure waveforms were recorded with a digital oscilloscope (Tektronix, TDS 3014B). Detailed information regarding the discharge and pressure characteristics of the experimental apparatus is provided in Yamatake et al. (2006); it was noted therein that the pressure waves are strong enough to inactivate organisms via electroporation.

During batch-scale inactivation experiments, 3 L of PBS at pH 7.4 were placed within the reactor and then the solution was inoculated with *E. coli* or *B. subtilis*. To investigate the impact of conductivity on microbial disinfection by PAED, the molarity of the PBS was varied to yield conductivities of 8.5 and 4.3 mS (representing wastewaters) and 588 and 532 μS (representing typical North American drinking water sources). Ten replicate experiments were conducted to determine *E. coli* and *B. subtilis* inactivation as a function of PAED treatment time (corresponding to a specified number of electrohydraulic discharges for a given reactor configuration). The number of discharges corresponded to detention times ranging from 200 to 5,000 s. Initial organism concentrations (at time = 0 s, corresponding to 0 discharges) were measured in duplicate before each experiment and were used to calculate microbe inactivation. Each sample was processed in duplicate. Reagent blanks (only PBS solution) were analysed after each set of electrohydraulic discharge experiments utilizing microorganisms to validate that the reactor cleaning procedure prevented carryover of microorganisms between experiments.

**Results and Discussion**

*E. coli, B. subtilis, and MS2 Inactivation by PAED in the 0.7-L Reactor*

Three *E. coli, B. subtilis, and MS2* inactivation experiments were conducted at two different water conductivities: 14.7 mS and 610 μS. The charging voltage was 2.2 kV during all of these inactivation experiments. Samples were collected after 0, 6, 18, and 30 pulses or accumulative energy inputs of 0, 0.7, 2.1, and 3.6 kWh/m³ respectively, corresponding to detention times of 0, 15, 60, and 80 seconds respectively. *E. coli, B. subtilis, and MS2* inactivation were calculated as the difference between the log₁₀ of each the reactor concentration before treatment began and after the specified number of pulses were applied. In these experiments, inactivation of the three microbial target compounds was evaluated concurrently.

The inactivation data from the 0.7-L reactor treating 0.01 M PBS are presented in a box-and-whisker plot shown in Fig. 2. In these plots, the dash in the center represents the median (50th percentile) *E. coli, B. subtilis,
Disinfection with Pulsed Arc Electrohydraulic Discharge

or MS2 inactivation. The lower and upper portions of the box respectively indicate the 25th and 75th percentile levels of inactivation respectively. The lower and upper portions of the line (whisker) respectively indicate the minimum and maximum levels of \textit{E. coli}, \textit{B. subtilis}, or MS2 inactivation observed. These data demonstrate reproducible inactivation levels between the triplicate experiments conducted in the 0.01 M PBS. Median \textit{E. coli} inactivation in this water matrix ranged from approximately 0.03-log with 6 pulses (15 s) to 1.1-log with 18 pulses (60 s) and 2.4-log with 30 pulses (80 s). Median \textit{B. subtilis} inactivation in the 0.01 M PBS ranged from 0.4-log with 6 pulses (15 s) to 3.1-log with 18 pulses (60 s) and 4.6-log with 30 pulses (80 s). Median MS2 inactivation in the 0.01 M PBS ranged from 0.3-log with 6 pulses (15 s) to 2.9-log with 18 pulses (60 s) and 3.8-log with 30 pulses (80 s).

The inactivation results from the 0.7-L reactor treating treated surface water are presented in a box-and-whisker plot in Fig. 3. Like the results from Fig. 2, these data demonstrate reproducible inactivation levels between the triplicate experiments conducted in the water matrix. Median \textit{E. coli} inactivation in the treated surface water ranged from approximately 0.06-log with 6 pulses (15 s) to 1.2-log with 18 pulses (60 s) and 2.8-log with 30 pulses (80 s). Median \textit{B. subtilis} inactivation in the treated surface water ranged from 0.4-log with 6 pulses (15 s) to 2.8-log with 18 pulses (60 s) and 4.0-log with 30 pulses (80 s). Median MS2 inactivation in the treated surface water ranged from 0.4-log with 6 pulses (15 s or 0.7 kWh/m$^3$) to 2.2-log with 18 pulses (60 s or 2.1 kWh/m$^3$) and 3.7-log with 30 pulses (80 s or 3.6 kWh/m$^3$).

While the results of the inactivation experiments conducted in the 0.7-L reactor were relatively consistent over triplicate experiments conducted in the various water matrices, they were also somewhat surprising. The \textit{E. coli} inactivation levels observed in these experiments were low relative to those that could be obtained more quickly by conventional treatment technologies. Upon obtaining similar levels of \textit{E. coli} inactivation, Ching et al. (2001) noted that the mechanism of UV disinfection in the electrohydraulic discharge process is intrinsically different from that of low intensity lamp inactivation, thereby yielding differences in inactivation kinetics. While this point raised by Ching et al. (2001) does provide some perspective for the \textit{E. coli} inactivation results, it does not necessarily elucidate why relatively higher levels of \textit{B. subtilis} and MS2 were consistently observed in these experiments. As the relative levels of inactivation summarized in Fig. 2 and 3 are not consistent with the relative levels of inactivation that would be expected from conventional drinking water treatment technologies such as chlorination, ozonation, and UV irradiation, these outcomes underscore the importance of elucidating the disinfection mechanisms acting in the PAED system. Moreover, the differences in water conductivity between the 0.01 M PBS (Fig. 2) and the treated surface water (Fig. 3) did not appear to appreciably impact the inactivation performance of the PAED treatment configuration, which was somewhat surprising and further underscored the need to elucidate the mode of germicidal action that yielded these promising results.

\textbf{E. coli Inactivation by PAED in the 3-L Reactor}

Six \textit{E. coli} inactivation experiments were conducted at three different water conductivities: 8.5 and 4.3 mS, and 532 μS. The charging voltage during the \textit{E. coli} inactivation experiments was set to approximately 4.5 kV, with the exception of the experiment utilizing 8.5 mS water during which it was approximately 3.0 kV. The initial experiments with the higher conductivity waters involved sampling after 0, 25, 50, and 100 pulses or an accumulative energy input of 0, 0.7, 1.4, and 2.8 kWh/m$^3$, respectively. Subsequent experiments conducted with the lower conductivity water involved sampling after 0, 25, 50, and 100 pulses or an accumulative energy input of 0, 1.6, 3.2, and 6.3 kWh/m$^3$ respectively, and additional samples after 300, 400, and 500 pulses or 19, 25.3, and 31.6 kWh/m$^3$ respectively.
The \textit{E. coli} inactivation data are presented on a log\textsubscript{10} scale in Fig. 4. These data demonstrate highly reproducible inactivation levels between duplicate and triplicate experiments in the 4.3 mS and 532 μS waters respectively. Approximately 0.3-log inactivation of \textit{E. coli} was obtained with 50 pulses (200 s) and 100 pulses (400 s) applied to the 8.5 mS water. \textit{E. coli} inactivation in the 4.3 mS water matrix ranged from approximately 0.4-log with 25 pulses (200 s) to 0.5-log with 50 pulses (400 s) and 0.7-log with 100 pulses (600 s). \textit{E. coli} inactivation in the 532 μS water matrix ranged from approximately 0.4-log with 25 pulses (200 s) to 1.2-log with 300 pulses (1,900 s) and 1.9-log with 500 pulses (3,300 s). The general trends depicted in Fig. 4 demonstrate generally reproducible inactivation of the \textit{E. coli}, however, the operational conditions (e.g., required contact time) must be optimized further for PAED application to drinking water treatment systems.

During the triplicate experiments conducted with 532 μS water, it became particularly evident that the time between pulses varied during the experiments. Specifically, during the last of these experiments, it was observed that the average pulse time increased by approximately 25 μS between each sampling event (0, 50, 100, 200, 300, 400, and 500 pulses). This change in average pulse time was likely associated with the observed changes in water conductivity over the duration of each experiment, which had been observed to some extent during all of the experiments. Conductivity increased from 8.5 mS to 14.6 mS during the first experiment and from 4.3 mS to 4.4 mS and 4.5 mS during the second and third experiments respectively. The initial water conductivity was 532 μS at the start of the fourth, fifth, and sixth experiments; during the experiments, it increased to 559 μS, 577 μS, and 1,100 μS respectively. Since the sixth experiment was conducted for almost twice as long as the fourth and fifth experiments, the conductivity differences between these experiments are not necessarily surprising, though one would not necessarily expect them to be linear. The changes in water conductivity within the reactor was likely associated with sputtering of the iron electrodes and the associated addition of dissolved iron to the water matrix. A detailed analysis of particle formation from iron and titanium electrodes in this PAED system was provided by Ikeda et al. (2007). While water conductivity impacted overall treatment time, the replicate trends in inactivation in response to the number of discharges were quite consistent.

It should be noted that the total iron content (dissolved and particulate) in the water also increased considerably during these experiments. In all cases, the initial total iron concentration was <0.05 mg/L and increased to anywhere from 0.4 mg/L to >3 mg/L, depending on the experimental conditions (initial concentration, number of pulses applied, etc.). It is unfortunate that only total iron was measured during these experiments because the relative impact of dissolved iron on conductivity could have been further evaluated if dissolved iron concentrations had been specifically evaluated. Given that the U.S. maximum concentration limit in drinking water for iron is 0.3 mg/L, the issue of residual iron concentration must be addressed for PAED to be feasible as a drinking water treatment technology. Particulate iron was clearly visible after some of the inactivation experiments, indicating that the particulate iron concentration of the water within the reactors clearly increased. The particulate iron content of the water likely contributed to the turbidity changes that were also observed as a result of PAED treatment. In general, initial turbidities were approximately 0.3 NTU prior to PAED treatment (typical of North American tap waters) and increased to anywhere from approximately 2 to 7 NTU after PAED treatment. The conductivity, dissolved iron concentration, and turbidity concerns arising from PAED treatment must be minimized if PAED is to be applied to pilot- and full-scale drinking water treatment systems. Accordingly, future experiments will be conducted with electrodes made from other materials, such as titanium, and particulate and dissolved metal concentrations, water conductivity, and turbidity will be closely monitored. It is possible that the particulate iron concentrations were due to electrode sputtering despite thorough filing of the electrodes between experiments. Formation of fine particles of Fe(OH)\textsubscript{3}(s) in this type of system was reported by Ikeda et al. (2007) when Ti electrodes were used, however. This result indicates the possibility of photo-Fenton reactions in the PAED reactor, but further investigations are necessary for a more conclusive analysis.

\textbf{B. subtilis} Inactivation by PAED in the 3-L Reactor

Four \textit{B. subtilis} inactivation experiments were conducted in water having a conductivity of 588 μS. The charging voltage during three of the \textit{B. subtilis} inactivation experiments was approximately 4.5 kV. An additional low power experiment was conducted with a charging voltage of approximately 2.5 kV. These experiments involved sampling after 0, 100, 200, 300, 400, and (in some cases) 500 pulses.

\textbf{Fig. 4.} Inactivation of \textit{E. coli} by PAED in the 3-L reactor.
Disinfection with Pulsed Arc Electrohydraulic Discharge

The *B. subtilis* inactivation data are presented on a log₁₀ scale in Fig. 5. Similar to the *E. coli* data, the *B. subtilis* data demonstrate highly reproducible inactivation levels between duplicate experiments in the given water matrices. *B. subtilis* inactivation in the synthetic drinking water matrix ranged from approximately 0.2-log with 100 pulses (−600 to 1200 s or 2.8 kWh/m³) to 0.5- to 1.2-log with 500 pulses (−3,800 s or 13.9 kWh/m³). These data demonstrate reproducible inactivation of the *B. subtilis*; however, as with the *E. coli*, the operational conditions must be optimized further for PAED application to drinking water treatment systems.

The increases in average pulse time associated with the observed changes in water conductivity ranged from approximately 19 to approximately 80 μS between each sampling event. As observed with the *E. coli*, the total iron content (dissolved and particulate) in the water also increased considerably during these experiments. In all cases, the initial iron concentration was approximately 0.03 mg/L and increased to anywhere from 1.8 mg/L to >3 mg/L depending on the experimental conditions (initial concentration, number of pulses applied, etc.). Changes in water turbidity were also observed as a result of PAED treatment. Initial turbidities were approximatey 0.3 NTU prior to PAED treatment (typical of North American tap waters) and increased to anywhere from approximately 5 to 28 NTU after PAED treatment.

Sputtering of the electrodes was not quantitatively measured during this study, however, it was ensured that the gap distance was constant and the electrodes properly filed. Ikeda et al. (2007) investigated the use of both Fe and Ti electrodes and observed the formation of fine particles of Fe(OH)₃(s) in a similar PAED reactor, even when Ti electrodes were used. Their result indicates the possibility of photo-Fenton reactions in the PAED reactor; however, further investigations are necessary (e.g., larger reactor volume) for more conclusive analysis. Future experiments will be conducted with electrodes made from other materials (e.g., tungsten or titanium) in an attempt to minimize such impacts; however, as discussed above, it is also possible that photo-Fenton reactions were occurring in the PAED reactor.

The preliminary results from the one experiment utilizing the lower (~2.5 kV) charging voltage yielded levels of *B. subtilis* inactivation similar to those observed when utilizing a higher charging voltage (~4 kV). While this is only one experiment, it underscores the importance of investigating the mechanisms of disinfection so that they can be optimized to achieve maximum microbial inactivation.

The results presented herein clearly suggest that further work is necessary to elucidate the primary mechanism(s) of disinfection acting in the PAED system and how they relate to the reactor design, applied power requirements, and disinfection of specific target microorganisms. Unlike conventional technologies (e.g., UV, ozonation, and chlorination) that utilize one disinfection mechanism, PAED inactivation may be achieved by combining four mechanisms or more (shock waves, UV radiation, radical reactions, thermal emission, etc.). A mechanistic discussion requires the quantification of all of the relevant disinfection mechanisms and their relative and potentially synergistic contributions to disinfection. Accordingly, the role of the mechanisms and their relative and potentially synergistic contributions to disinfection may vary between reactor designs. Speculation regarding the relative contributions of the potential disinfection mechanisms (which are all present in varying degrees) is beyond the scope of the present investigation.

**Conclusions**

In preliminary investigations utilizing a 0.3-kJ spark gap type PAED power supply and a 0.7-L batch reactor, PAED was able to achieve 2.4-, 4.6-, and 3.8-log median inactivation of *E. coli*, *B. subtilis* spores, and MS2 bacteriophage in 0.01 M PBS after 30 pulses (80 s) of treatment. Similar levels of inactivation were observed in treated surface water, in which median observed inactivation was 2.8-log of *E. coli*, 4.0-log of *B. subtilis*, and 3.7-log of MS2. Subsequent investigations utilizing the same 0.3-kJ PAED power supply and a 3-L reactor resulted in approximately 1- to 2-log inactivation of *Escherichia coli* and *Bacillus subtilis* (after 500 pulses or 31.6 kWh/m³ accumulative energy input) in water matrices with conductivities typical of North American wastewaters (8.5 and 4.3 mS) and drinking water sources (532 μS). All of the data presented herein clearly demonstrate that PAED may offer an effective barrier for the protection of public health if the water quality impacts on conductivity, turbidity, and metal concentration are minimized (potentially by optimizing electrode material) and if treatment time can be further reduced.
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