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An innovative non-thermal plasma reactor to eliminate microorganisms in water

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ABSTRACT

The growing need for scalable systems that can inactivate microbiological contaminants and recycle water in industrial operations has led to the development of a variety of new advanced oxidation process (AOP) technologies. In this paper, we report on the capability and techno-economics of a new AOP method to generate aqueous plasma species for inhibition of microbiological contaminants. The test microorganisms in this work were *Acidithiobacillus ferrooxidans* (a motile, Gram-negative bacterium that oxidizes sulfides to sulfates and ferrous iron to ferric iron, used as a model biofouling organism) and *Legionella gratiana* (a Gram-negative bacteria used as a surrogate of the human pathogen *Legionella pneumophila*, which can be a dangerous contaminant in cooling water systems). The cultured bacteria were dispersed in water and treated within a non-thermal plasma treatment system for varied exposure times, and then the bactericidal effects were measured. The results demonstrated plasma inhibition of *A. ferrooxidans*, with an approximate 6 log decrease in viability (assayed as most probable number) with 40 s of aqueous plasma treatment in the plasma treatment system. Likewise, *L. gratiana* viability was decreased, with an approximate 6 log decrease in viability with 20 s of aqueous plasma treatment (assayed as colony-forming units). Modeling the techno-economic aspects of these disinfection reactions in the treatment system indicated the potential for the technology to be competitive with existing AOP and aqueous chemical-based disinfection methods.

Keywords: Non-thermal plasma; Oxidation; Disinfection; Water treatment

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1. Introduction

Clean water is a limited resource, and new technologies are needed to economically recycle water from industrial-scale operations and to safely discharge water back into the environment. Thermoelectric power plants and other industrial-scale operations require large quantities of fresh water for use in cooling water systems [1]. Such cooling water systems provide suitable environments for the growth of many types of bacteria, including those that can cause deterioration of ferrous pipe materials [2,3], and those that can be pathogenic to humans [4]. Thus, microbial growth in industrial cooling water systems leads to the interrelated problems of biofouling, scaling, and corrosion, along with possible negative impacts on human health.

Typically, disinfectants are added into the industrial cooling water system circulation to control microbial growth, but this practice is limited by the expense of the disinfectants (e.g. chlorine) and the need for waste-stream cleanup (dechlorination) before environmental discharge [5,6]. Other methods for microbial control in such cooling systems include ultraviolet inactivation and/or filtration (including ultrafiltration and reverse osmosis). However, these methods can be limited by expense, lack of utility in turbid waters, and/or lack of suitable scalability [5,7].

Advanced oxidation processes (AOP) offer another avenue to control microbial-related contamination, biofouling, scaling, and corrosion in industrial cooling water systems. AOP methods control microbial proliferation through their generation of antimicrobial oxidants [8,9]. AOP are typically initiated via the addition of chemical disinfectants with oxidizing properties, and thus may suffer from many of the same drawbacks described above for chemical disinfectants. However, recent studies demonstrate that non-thermal plasmas can be used to generate reactive oxidative species in water and thus provide a possible solution for effective and efficient microbial control [10,11]. In addition, the energetic discharges from non-thermal plasmas possess sufficient kinetic energy to break organic bonds, synergistically adding to the oxidative antimicrobial effect [12–14].

Aqueous plasma generation strategies [15] that have focused on bacterial inactivation include systems with electrodes discharging in a gas phase in contact with an aqueous solution [12,16], systems with electrode configurations discharging within an aqueous solution [17–23], systems with capillary electrode configurations [24], systems with gliding-arc discharges [25,26], and systems with electrodes separated by dielectric barriers [27,28]. In general, however, these systems have high voltage and power requirements, slow reaction kinetics, and/or issues with their scalability. Denes and co-workers [29,30] developed a system which utilized non-thermal plasma chemistry that has the advantage of sustaining a pseudo-volumetric plasma submersed in an aqueous solution, where the transport of the microbial contaminants is controlled by convective mass transfer. This improvement enhanced the microbial decontamination rates considerably when compared to other point-to-plane non-thermal plasma systems [29–31]. In this system, a combined mixture of 16 different types of bacteria was treated with aqueous plasma, with approximately 2 log reduction in viability observed after 20 s of plasma exposure time [30].

While the initial disinfection investigation was promising, additional experimentation demonstrated that improvements to the reactor design, while still maintaining the pseudo-volumetric nature of the non-thermal plasma, could further enhance the efficacy of the system [32]. These modifications, in short, were postulated to (1) increase the interaction of the reactive species generated by the plasma and thereby increase oxidation and microbial inactivation rates, (2) lower the power required to initiate and sustain the plasma, and (3) provide a more robust reactor design that could be utilized in both batch and continuous flow configuration. An in-depth analysis of the proposed improvements and the effect on reactor performance from both kinetic analysis and computational fluid dynamics viewpoint are contained in Johnson et al. [32–34].

To test the postulated improvements, the present study is focused on fabricating and testing the oxidative plasma system described in [34], which advances the volumetric plasma generation capabilities described in Johnson et al. [32], for the bactericidal treatment of cooling water systems. The intent is to determine if the oxidative plasma system has the potential to overcome the physical and economic drawbacks of plasma-based and classical AOPs as described above. The plasma discharge characteristics as well as the disinfection ability of the reactor were tested and reported herein. The antimicrobial properties of the plasma were experimentally determined using a model biofouling organism *Acidithiobacillus ferrooxidans* (a motile, Gram-negative bacterium that oxidizes sulfides to sulfates and ferrous iron to ferric iron) [35] and *Legionella gratiana* (a Gram-negative surrogate of the human pathogen *Legionella pneumophila*, a common contaminant in cooling water systems that causes Legionnaires' disease) [36]. The antimicrobial studies were conducted in fresh water [37]; the intention was to make the aqueous samples representative of the fresh water typically used in power plant cooling tower systems [38]. Additionally, a cost analysis based on the bacterial inactivation kinetics

is presented in order to explore the feasibility of the proposed plasma system to effectively treat thermoelectric power plant cooling tower water.

2. Materials and methods

The goal of the present study was to develop and test a non-thermal plasma treatment system and evaluate the feasibility of its use for bacterial inactivation in aqueous samples. In this plasma reactor, electricity is used to generate an oxidizing plasma in aqueous solution, producing a mixture of antimicrobial agents including UV radiation, high-energy electrons, ozone, hydrogen peroxide, hydroxyl radicals, and others [32,34]. The main features of the plasma reactor are briefly described here, while a more in-depth, conceptual discussion of the design principles can be found elsewhere [32–34,39].

2.1. Plasma reactor system

A schematic representation of the plasma reactor is depicted in Fig. 1. This schematic illustrates the introduction of aqueous suspensions of bacteria that also contained components of the bacterial growth media (A) entering the tubular chamber where the plasma is generated (B**)** through an inlet (C**)** in the upper reactor flange (D**)** and exiting as a treated effluent (E**)** through outlets (F), respectively, in the lower reactor flange (G) after traveling axially (H) through the tubular plasma chamber. A hollow, rotatable, electrically conducting shaft (I) that has a surface parallel to the wall of the tubular plasma chamber (J), which serves as the positive electrode, and has an array of hollow pin electrodes (K) designed to release fine gas bubbles [34], which serve as the negative electrode, affixed and protruding perpendicularly from the conducting shaft surface. For the inactivation experiments described below, 40 pin electrodes were utilized in the plasma reactor.

The hollow pin electrodes protrude through an electrically insulating sheath constructed from alumina ceramic or teflon (L), which rotates with the electrically conducting shaft and is utilized to electrically insulate the pin electrodes up to the point at which they contact the contaminated fluid. The thickness of the insulating sheath can also be varied to manipulate the available fluid volume in the tubular plasma chamber. The electrode gap (M) between the tips of discharge pin electrodes (N) and the inner wall of the outer cylinder (O) can be adjusted to distances on the order of a 100 μm to a few centimeters and is set to approximately 1.5 mm for the bacterial inactivation experiments in the present study.

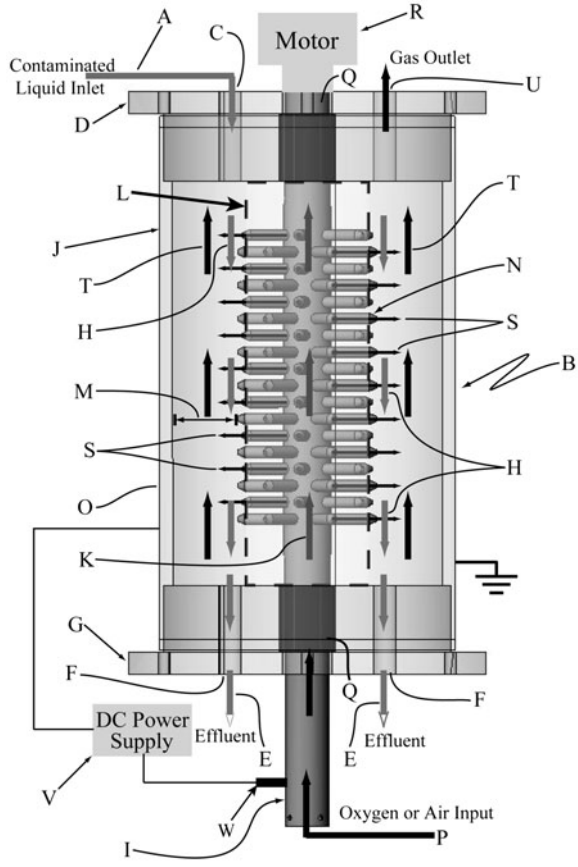

Fig. 1. A schematic illustration of the plasma reactor employed in the described experiments. Approximate flow patterns of the liquid and gas are highlighted in addition to alphabetical labeling of all important reactor components [34]. Briefly, the schematic is marked to show the aqueous suspensions of bacteria (A), the tubular plasma reactor chamber (B), solution inlet (C) through the upper reactor flange (D), treated effluent solution (E), outlets (F) in the lower reactor flange (G), direction of solution travel (H), a hollow, rotatable, electrically conducting shaft (I), chamber outer wall and positive electrode (J), the array of hollow pin electrodes (K), which serve as the negative electrode, an electrically insulating sheath (L), the electrode gap (M) between the tips of discharge pin electrodes (N) and the inner wall of the outer cylinder (O), compressed air (P), shaft bearings (Q), electric motor (R), direction of gas exiting pin electrodes (S), direction of gas flow though reactor (T), gas outlet (U), the DC power supply (V), and an electrically connecting carbon brush (W).

Before the introduction of the bacterial suspensions, compressed air (P) is introduced into the system through the shaft (I), which is rotated on bearings (Q) by a motor (R) at rates from 250 to 1,000 rpm and is fixed at 1,000 rpm for the inactivation experiments. The air is expelled into the tubular reactor chamber through the center of the hollow pin

electrodes. The gas introduction system is designed to ensure an equal volumetric flow rate of the gas as finely dispersed bubbles through each pin electrode (N), independent of their position along the shaft (I). Once introduced into the reactor chamber, the gas exits (S) from the center of each hollow pin electrode and rises (T), thereby flowing counter-currently to the aqueous bacterial suspensions (H) that are being treated within the reactor chamber, and exits the tubular plasma chamber (J) through gas outlet (U) in upper flange (D).

It should also be noted that the bores through each pin electrode have been fitted with porous stainless steel gas diffusors. The diffusors have an average pore size of 40 μm and serve the function of distributing the input gas more evenly along the tip of the pin electrode. It is expected that a well-dispersed introduction of gas into fluid will stabilize the plasma, increase electrode life, and promote pollutant oxidation, as previously demonstrated by Johnson et al. [31]. Fine bubbles exiting the center of the pin electrodes are also expected to further reduce the localized density of the fluid adjacent to the discharge pin electrode surface, thereby reducing the voltage required, with a consequent reduction in power consumption for initiating and sustaining the plasma. A reduction in total current can also be realized, thereby reducing the current density at each pin electrode resulting in an anticipated increase in electrode life. A systematic investigation of the effect of the gas diffusors on reactor performance is beyond the scope of the experiments described here and is currently underway. Qualitatively, however, the utilization of the gas diffusion has a positive impact in terms of initiating and sustaining the plasma.

To initiate and sustain a plasma discharge at the tip of the pin electrodes (N) that propagates to the inner wall of the stationary outer cylinder (O), a DC power supply (V) is placed in electrical connection with the conducting shaft (I) using a carbon brush (W) isolated in a spark plug housing. In order to complete the circuit, the return connection is in contact with the stationary outer cylinder (O), which is electrically insulated from the inner shaft by insulating bearings (Q). The DC power supply utilized to initiate and sustain the plasma discharge was an electronically controlled Pinnacle® Plus + Pulsed-DC Power Supply from Advanced Energy Industries, Inc. in Fort Collins, Colorado, with features that support precise control of plasma intensity and duration. A photograph and short video illustrating the plasma chamber before and during plasma ignition from a characteristic *L. gratiana* inactivation experiment is contained online in the supplementary information.

2.2. Bacterial strains and growth conditions

Growth of *A. ferrooxidans* (ATCC 13598) was initiated from a lyophilized stock, followed by static incubation in ATCC medium 2039 at 26˚C for two weeks. *L. gratiana* (ATCC 49413) from lyophilized stocks was grown on charcoal yeast extract (CYE) agar at 37˚C with 5% CO_2 for three days and then subcultured in CYE broth and statically incubated at 37˚C with 5% CO_2 for one week to generate the inoculum.

In order to investigate the effect of the pin electrode diameter on plasma power consumption, variations of the CYE broth were used as a model influent media. These variations are discussed below and referred to as solutions A, B, and C. As in the plasma-mediated bacterial inactivation experiments, the CYE media was prepared in a 1:50 dilution, but did not contain viable bacteria. Solution A is simply the CYE media at the 50:1 dilution. Solution B underwent filtration through 6 μm Whatman® qualitative filter paper (Grade 3). Solution C was centrifuged at 13,000 rpm for 20 min after filtration. The goal of these additional steps was to remove the carbon-based particulates from the solution media that could also be affecting the plasma power consumption. For all of the above described solutions, the total dissolved solids (TDS) concentration was manipulated through the addition of NaCl (Fisher Scientific, 99.6%).

2.3. Plasma-mediated bacterial inactivation

Bacterial cultures were diluted in Millipore MilliQ water to a concentration of approximately 10^7 CFU/ml (a 1:50 dilution), treated with aqueous plasma, and collected from the plasma reactor at time zero prior to initiating plasma treatment, and at 20 s intervals throughout the course of plasma treatment. This interval was chosen for practical considerations associated with withdrawing samples from the reactor. Plasma generation conditions within the reactor for the *A. ferrooxidans* experiments were 300 mL fill volume, air flow rate 200 standard cubic feet per hour (SCFH), spin rate 1,000 rpm, current 3 A, and 300–700 V. Plasma generation conditions within the reactor for the *L. gratiana* runs were 300 mL fill volume, air flow rate 250 SCFH, spin rate 1,000 rpm, current 3.5 A, and 500 V.

Viable numbers of *A. ferrooxidans* were directly assessed using the five-tube most probable number (MPN) technique employing the growth conditions described above [40]. In the five-tube MPN technique, samples are serially diluted (five replicate tubes per dilution factor) to extinction allowing for the presence of viable bacteria in the tubes with lowest dilution factor and the absence in the tubes with higher

dilution factor. Based on the dilution factor, and the number of the tubes (of a single dilution factor) which contains viable organisms, an estimate of the original bacterial concentration can be extrapolated. Such a method is used to estimate the quantity of viable microorganisms, including *A. ferrooxidans*, which cannot be enumerated by direct plate count methods. Direct plate counts are obtained by plating serial dilutions of the sample onto agar, counting of colony-forming units (CFU) following bacterial growth, and extrapolating bacterial concentrations from the number of counted CFU and dilution factor. The numbers of viable *L. gratiana* were determined by direct plate counts following the previously established growth parameters for this microorganism. Additionally, the plasma-induced inactivation kinetics of *L. gratiana* were indirectly assessed by measuring ATP (an indicator of cell viability) using the Ultrasnap ATP test (Hygiena, Camarillo, CA) and Pi-102 luminometer (Hygiena) throughout the course of treatment. The manufacturer's instructions were followed with the following exceptions: (a) the swab component of the Ultrasnap assay tube was removed and (b) 0.5 ml of the bacterial suspension was assayed.

3. Results and discussion

3.1. Bactericidal effect of the plasma reactor

The plasma reactor was infused with bacterial suspensions, which also contained components from the growth media. Plasma treatment caused an approximately 3 log reduction in viable *A. ferrooxidans* within 20 s of plasma treatment, and an almost complete (6 log) inactivation of this bacterium was observed following plasma treatments of 40–60 s, as demonstrated in Fig. 2. Similarly, exposure to the plasma conditions within the reactor caused approximately 6 log reduction in *L. gratiana* viable counts within 20 s of plasma treatment, and further inactivation (approximately 7 logs) following 40 and 60 s of plasma treatment as indicated in Fig. 3(A). The indirect assessment of *L. gratiana* viability by an ATP assay indicated a significant decrease (an indication that bacterial inactivation occurred) in cellular ATP levels within 20 s of plasma treatment, and further reductions of ATP levels after 40 and 60 s of plasma treatment, for which the data are shown in Fig. 3(B). The fact that ATP was present after 40 s of treatment indicates that bacteria were still present in the sample; however, considering that no bacterial colonies were present when cultured on solid media, this demonstrates that the remaining bacteria were injured by the treatment process, such that they could not replicate (indicating they were not viable). These results

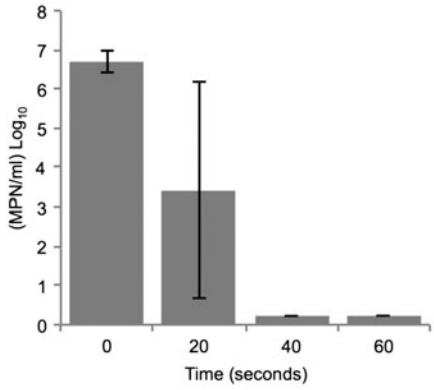

Fig. 2. Reduction in numbers of viable *A. ferrooxidans* following treatment in the plasma reactor. Aliquots of the bacteria-containing water suspensions were withdrawn from the plasma reactor at defined intervals (0, 20, 40, and 60 s) and enumerated by the MPN method. The average MPN/ml (and standard deviations) from three replicates is reported.

support the conclusion that the plasma treatment exhibited significant antimicrobial effects [41,42].

In comparison with other characterized disinfection procedures for *A. ferrooxidans* and *L. gratiana*, the inactivation kinetics observed with the non-thermal plasma system are favorable [5,43–45]. Ultraviolet light (UV) treatment of *A. ferrooxidans* with 56 ergs/mm^2/s treatment resulted in approximately 4.5 logs of reduction within 40 s of treatment (whereas a 6 log inactivation was achieved with the non-thermal plasma within 40 s) [45]. Similarly, a 6 log reduction was achieved for *L. gratiana* within 20 s using the non-thermal plasma treatment. For *Legionella* spp., UV light and other oxidizing sanitizing agents including halogens, ozone, hydrogen peroxide, and potassium permanganate require contact times ranging from minutes to days to achieve bacterial reduction similar to that of the plasma system (5).

3.2. Plasma treatment system scale-up

3.2.1. Bacterial inactivation kinetic analysis

Due to the rapid inactivation kinetics as well as the favorable comparison discussed above, an additional analysis is included to determine the viability of the proposed system on a scale suitable for thermoelectric power plant cooling tower water treatment. In order to perform the analysis, the rate at which the laboratory-scale plasma treatment system can inactivate the microorganisms evaluated above must first be determined. The dimensional form of the species conservation equation

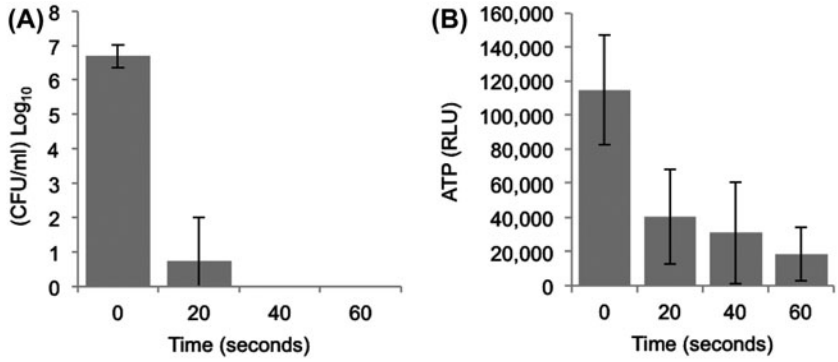

Fig. 3. Reduction in viable *L. gratiana* following treatment with the plasma reactor. Aliquots of the bacterial suspension were extracted from the plasma reactor at defined intervals (0, 20, 40, and 60 s) and enumerated by direct plate counts (A). Alternatively, bacterial viability was indirectly assessed by monitoring changes in ATP levels following reactor treatment, where a decrease in ATP indicates cell death (B). The average CFU/ml and RLU from three replicates (and standard deviations) are reported.

$$\frac{\partial C_i}{\partial t} + \underline{v} \cdot \nabla C_i = D_{i_{mix}} \nabla^2 C_i + r_i \tag{1}$$

can be used to model the kinetics associated with the inactivation of bacteria in the plasma treatment system. This equation can be simplified to the standard design equation for a batch reactor

$$\frac{\partial C_i}{\partial t} = r_i \tag{2}$$

where i is the bacterial species of interest, t is the treatment time, r describes the inactivation of bacterial species i, and C is the time-dependent concentration of bacterial species i in the plasma treatment system. Given that the treatment of the contaminated water is achieved within a short time frame, as demonstrated in Figs. 2 and 3, a key assumption that requires validation is that the inactivation kinetics are mass-transfer limited and will therefore be pseudo-first-order in nature. This assumption allows Eq. (2) to be further expressed as

$$\frac{\mathrm{d}C_i}{\mathrm{d}t} = kC_i \tag{3}$$

where k is the rate of bacterial inactivation. Solving Eq. (3) yields

$$\mathrm{Ln}(C_i) = kt \tag{4}$$

To empirically demonstrate that this assumption is valid, the log(bacterial species concentration) for *A. ferrooxidans* and *L. gratiana*, in units of MPN/mL and CFU/mL, respectively, is converted to the Ln (bacterial species concentration) and plotted vs. treatment time. If the assumption is valid, the relationship between the Ln(species concentration) and time is linear with the slope of the linear fit corresponding to the pseudo-first-order inactivation rate constant. This is in fact demonstrated for *A. ferrooxidans* in Fig. 4.

The same analysis could be conducted for *L. gratiana*; however, there are only two non-zero data points. This is due to the fact that the disinfection is sufficiently facile that significant inactivation is achieved, within the ability to detect viable bacteria, before the third

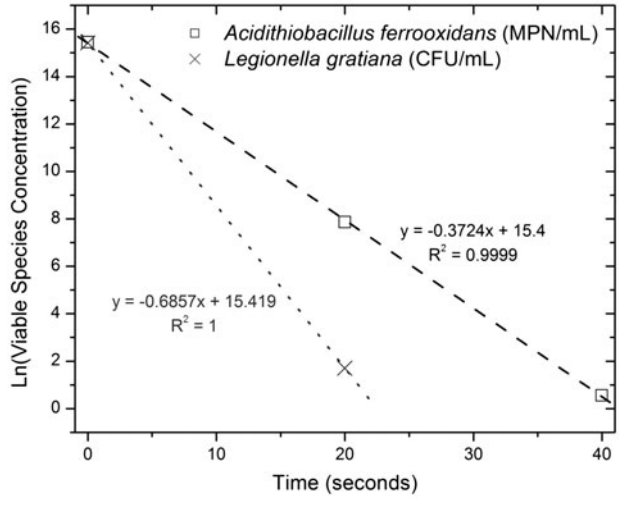

Fig. 4. Natural log of *A. ferrooxidans* and *L. gratiana* concentration, in units of MPN and CFU/mL, respectively, as a function of time. A linear fit of the data is also included for which the slope of the lines corresponds to the pseudo-first-order rate of bacterial inactivation.

sample is collected at 40 s. A linear regression based just on the two non-zero data points is not appropriate. However, as discussed above, the inactivation of *A. ferrooxidans* has been demonstrated to be pseudo-first-order. Because the plasma reactor is operated in a similar manner when inactivating *A. ferrooxidans* and the inactivation kinetics are more facile in comparison, the disinfection mechanisms of the reactor for the two species should be consistent. These considerations justify the assumption that the microbial inactivation kinetics for *L. gratiana* also are pseudo-first-order. The data for *L. gratiana* inactivation are plotted in Fig. 4 in conjunction with the inactivation data for *A. ferrooxidans* for comparison.

To calculate the pseudo-first-order rate constant associated with the disinfection kinetics, a linear regression is performed on the data plotted in Fig. 4. The slope, which is negative, corresponds to the rate at which the bacterial species is inactivated. Using the above analysis of the data, the rate constant for the inactivation of *A. ferrooxidans* and *L. gratiana* is calculated to be 0.3724 and 0.6857/s, respectively, for 40 pin electrodes and an electrode gap of 1.5 mm, the configuration of the laboratory-scale plasma treatment system described above.

In order to estimate the economic viability of an industrial-scale equivalent of the laboratory plasma reactor system utilizing the using kinetic data discussed above, the disinfection rates for each bacterial species must be scaled. The scaling procedure accounts for an increase in the electrode gap as well as an increase in the number of pin electrodes, for which both are envisioned in a commercial size treatment system. An introductory discussion for scaling a plasma reactor system is contained in Johnson et al. [32]. These inactivation rates are incorporated into a techno-economic model, which is subsequently discussed, to determine if the proposed plasma treatment system can economically and effectively disinfect thermoelectric power plant cooling tower water.

3.2.2. Techno-economic model

To calculate the operating costs associated with the tubular plasma reactor when scaled from a laboratory-scale batch reactor to an industrial-scale continuous flow (plug-flow) configuration, an initial microbial concentration consistent with what is observed in characteristic thermoelectric power plant cooling tower water with a required 2 log reduction in the viable microbial species are utilized as input parameters for the model. The DC power consumed has been experimentally determined to be 1.7 kW for *L. gratiana* and 1.6 kW for *A. ferrooxidans*. These values, as well as AC to DC power conversion factors ranging from 0.6 to 0.95, were subsequently used to calculate the cost associated with plasma generation. In addition to the plasma cost, the power consumption of the compressor, which is needed to supply air flow through the middle of the pin (discharge) electrodes, and the shaft motor, which is needed to spin the pin electrodes, are included in the analysis.

To determine the power requirement associated with pumping contaminated water through the tubular plasma reactor, the experimentally determined kinetic inactivation rates are utilized to determine the flow rate at which the reactor can provide a 2 log reduction in the contaminant species concentration. To calculate these volumetric flow rates, a set of dimensionless equations, for which a complete description of the derivation methodology is found elsewhere [32,34], are solved. Briefly, to determine the required residence time (τ) necessary to achieve the required 2 log reduction in viable bacteria, the dimensionless form of the species conservation equation (Eq. 5) is solved.

$$Pe_i\left(\frac{\mathrm{d}\varphi_i}{\mathrm{d}\zeta}\right) = \frac{\mathrm{d}^2\varphi_i}{\mathrm{d}\zeta^2} + \sum_{ij} Da_{ij} r^*_{ij} \tag{5}$$

In Eq. (5), the terms from left to right can be attributed to rate processes associated with convective mass transfer, diffusion, and/or sinks due to physical and chemical bacterial removal mechanisms. Note that the accumulation term is not included in Eq. (5) because of the steady-state operation of the idealized commercial plasma treatment system. Because of the turbulent conditions induced in the plasma reactor by the high spin rate of the pin electrodes, the diffusion term also can be neglected. The dimensionless species conservation then reduces to Eq. (6).

$$\frac{\mathrm{d}\varphi_i}{\mathrm{d}\zeta} = -\frac{L}{v_{\mathrm{avg}}}\left(k_{plasma}\varphi_i + k_{mt}C_{i_o}\varphi_i^2 + \frac{k_{\mathrm{photo}}}{C_{i_o}}\right) \tag{6}$$

In Eq. (6), the residence time is the reactor length, L, divided by the average fluid velocity, v_{avg}, and the inactivation of the bacterial species can be attributed to disinfection due to the plasma (and disinfection species produced by the plasma), a transfer of the bacterial species from the liquid phase to the gas phase that is expelled from the reactor, and disinfection due to exposure to the wide spectrum radiation produced by the plasma discharge. Thus, the above equation allows one to estimate the maximum fluid velocity through the reactor that results in the desired

reduction in viable bacteria numbers given the appropriate disinfection rate constants. Please see [32,39] for a complete description of the above derivation.

While deconvolution of the bacterial removal/inactivation rate constants associated with the plasma, entrainment in the gas phase, and radiation exposure is beyond the scope of this work—albeit the focus of upcoming research—the kinetic data obtained from the above analysis can be utilized, in combination with previously developed scaling factors for the number of pin electrodes and electrode gap, to estimate the efficacy of the plasma treatment system. The results from this estimation are contained in Fig. 5. As evidenced by the figure, while the inactivation of *A. ferrooxidans* is more costly when directly compared to *L. gratiana* due to the slower inactivation rate in the plasma treatment system, the total operating costs fall between just over \$0.02/1,000 L for a 65% power conversion efficiency and as low as \$0.013/1,000 L for a 95% power conversion efficiency when using \$0.12/kWh, which is based on the average cost of residential electricity as calculated by the US DOE. While the calculated operating cost is impressive, a route to further reduce the power consumption due to the plasma is to modify the pin electrodes.

As dictated by the Paschen curve, a reduction in the radius of curvature can result in a reduction in the required power needed to generate and sustain an arc discharge in a point-to-plane electrode configuration [39]. To determine if a reduction in the radius of curvature would result in a decrease in the plasma power consumption for the plasma treatment system described above, the diameter of the electrode pins was reduced from approximately 0.95 to 0.64 cm. The average power required to initiate and sustain the plasma for the duration of the inactivation experiments is plotted in Fig. 6. As demonstrated by the figure, a reduction in the pin diameter resulted in a

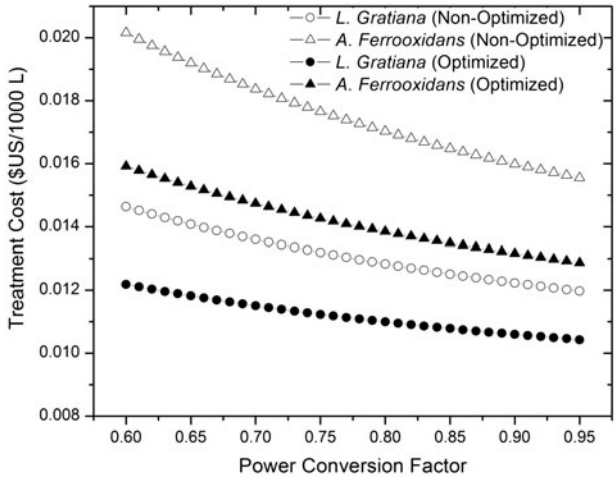

Fig. 5. A comparison of the cost required to achieve a 2 log reduction in viable bacteria when using the plasma treatment system under non-optimized and optimized plasma consumption conditions.

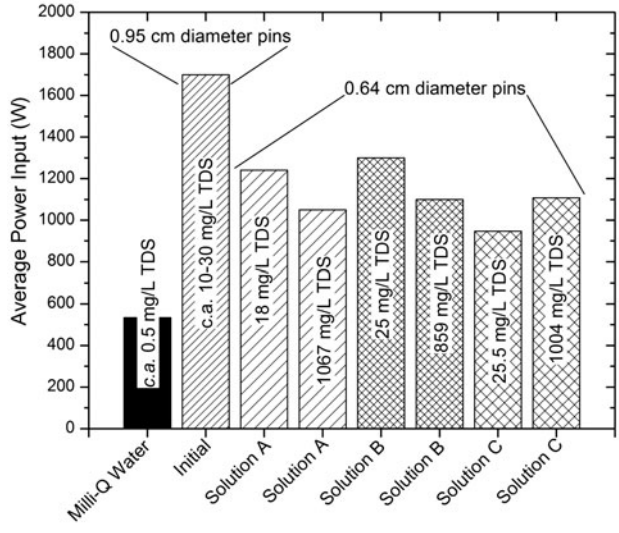

Fig. 6. A plot of the average power input required to initiate and sustain the plasma for two different electrode pin diameters. The power requirements for Millipore MilliQ water and contaminated solutions with artificially increased TDS also are included.

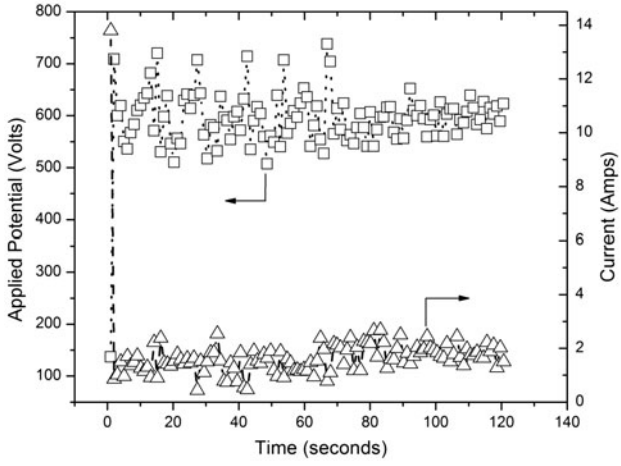

Fig. 7. A characteristic plot of the voltage and current response when a plasma is initiated and sustained in a media suitable for bacterial growth, solution C. Note the initial strike current is approximately 14 A, but stabilizes quickly at a value close to 2 A. Conversely, the initial strike voltage is considerably lower (approximately 150 V) when compared to the steady-state voltage of approximately 600 V.

reduction of 500 W in the power required for the plasma. To determine if a higher salt content, as is sometimes encountered in power plant cooling water, affected the power input, the contaminated solution TDS concentration was artificially increased with NaCl. As demonstrated by the data contained in Fig. 6, the added salinity did not have a negative effect on the plasma discharge, as the non-thermal discharge was initiated and sustained at TDS concentrations on the order of 1 g/L. A characteristic current–voltage plot for solution C is contained in Fig. 7 demonstrating the unique ability of the plasma treatment system to initiate and sustain a discharge under high salinity conditions. In addition to a decrease in power consumption with a decrease in the electrode diameter, it was also determined that in the plasma treatment system, a plasma can be initiated and sustained at approximately 500 W in a solution consisting of Millipore MilliQ water, indicating that constituents of the contaminated solution resulted in an increase in the plasma power. An investigation of the effect of these constituents is ongoing.

When utilizing a more optimized electrode configuration, the results presented in Fig. 6 suggest that the plasma power can be reduced to 1.12 kW for *L. gratiana* and 1.06 kW for *A. ferrooxidans*. Utilizing the optimized plasma power consumption, values yield the second set of curves plotted in Fig. 5. As expected, the inactivation of *A. ferrooxidans* continues to be more costly when directly compared to *L. gratiana*, but the total operating cost falls considerably to between $0.016/1,000 L for a 65% power conversion efficiency and approximately $0.010/1,000 L for a 95% power conversion efficiency. Because of the effective inactivation efficiency associated with the tubular plasma reactor, as demonstrated both by the inactivation kinetics and the low operation cost, the associated volumetric flow rate calculated by the model is in excess of 0.1 MGD (million gallons per day) for one industrial-scale plasma reactor that is approximately 150 cm in length and 25 cm in diameter. This is beyond the physical ability to flow water through a single reactor due to frictional losses. This is a positive result as it indicates that the disinfection kinetics are not limiting, but that the fluid dynamics through the reactor is the most important design consideration as long as the plasma discharge can be maintained. Even though it has become obvious through this kinetic analysis and techno-economic model that the reactor module design developed is not fully optimized for microbial disinfection, the calculated operating cost is still impressive and provides a baseline for subsequent design improvements.

3.2.3. Key reactor design insights

A key metric for cooling water treatment is a log 2 reduction in the concentration of viable microbes when treated with the plasma reactor. As calculated by the techno-economic model, the volumetric flow rates that can be achieved while still accomplishing a log 2 reduction is impressive to the point that the water cannot physically be driven through the reactor. Thus, the plasma treatment system, when utilized to inactivate the bacterial species studied here, is not limited by the ability to inactivate the microorganisms. The system, therefore, needs to be redesigned with special attention being given to maximizing fluid flow through the reactor. This can be accomplished by increasing the cross-sectional area of the reactor in order to accommodate the highest flow rate without impacting the plasma discharge, while simultaneously decreasing the reactor length.

The techno-economic model revealed that the plasma generation and the fluid pumping are the main contributors to the operating cost associated with the plasma treatment system. This is highlighted in Table 1. Since 39–62% of the cost can be attributed to the plasma generation, the pin electrode array should be redesigned to minimize power consumption, while

Table 1
Breakdown of the estimated percent cost associated with the four main operating cost categories—plasma generation, fluid pumping, compressed air, and the shaft motor needed to spin the pin array

Percent of total consumed power	Initial power setting		Optimized power setting	
	L. gratiana (%)	*A. ferrooxidans* (%)	*L. gratiana* (%)	*A. ferrooxidans* (%)
Plasma generation	49.4	61.9	39.0	51.8
Pumping (Liquid)	48.8	35.3	58.5	44.7
Compressor (Air)	0.44	0.60	0.54	0.77
Pin array shaft motor	1.45	2.16	1.95	2.74

physically allowing the maximum amount of fluid to be passed through the reactor. As demonstrated above, a reduction in the power consumption could be achieved through modification of the pin electrodes to essentially reduce the effective voltage required to initiate plasma generation. The preliminary investigation described within utilizing pin electrodes with a reduced diameter resulted in a reduction of approximately 10% of the cost associated with the plasma generation. In the case of the optimized power configuration, the power consumption due to pumping the contaminated fluid is greater than that required to generate and sustain the plasma. Thus, reducing the energy required to pump the contaminated fluid through the reactor must also be addressed to further reduce the estimated treatment cost.

4. Conclusion

The bacterial inactivation studies focused on the use of the above-described plasma reactor system demonstrated facile inactivation kinetics, which were subsequently utilized in a techno-economic model, to estimate the cost associated with its use for cooling tower water disinfection. Both the disinfection and economic feasibility study results indicate the potential for this technology to compete with existing AOP and traditional aqueous-based disinfection technologies. In addition to demonstrating the technological and economic proficiency, the kinetic analysis and modeling efforts also elucidated possible design improvements in the industrial-scale plasma treatment system. Because the bacterial inactivation kinetics have been demonstrated to be extremely facile, the calculated flow rate based solely on the disinfection kinetics resulted in volumetric flow rates through the plasma treatment system that could not be supported. Based on these findings, a new scale-up plasma reactor design is underway that will accommodate the highest possible volumetric flow rate without impacting the plasma discharge or the facile inactivation kinetics.

In addition to informing a new reactor design, the facile kinetics have highlighted the need to better understand the microbial inactivation mechanism within the plasma system. It has been previously demonstrated that when plasma discharges of the type described above are initiated in an aqueous solution, many different reactive species—such as ·OH, ·H, ·O, O_2^-, $\cdot HO_2$, H_2O_2, O_3, and potentially others depending on the composition of the aqueous solution—as well as intense UV radiation are generated [20,46–50]. Microbial inactivation can, therefore, be initiated through chemical oxidative damage, cellular damage through exposure to UV radiation, and/or direct interaction with the plasma. Subsequent experiments are currently being designed to determine the most prevalent disinfection mechanism.

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