

Development of an Electrochemical Process for Blackwater Disinfection in a Freestanding, Additive-Free Toilet

Katelyn L. Sellgren, Christopher W. Gregory, Michael I. Hunt, Ashkay S. Raut, Brian T. Hawkins, Charles B. Parker, Ethan J. D. Klem, Jeffrey R. Piascik, and Brian R. Stoner



RTI Press publication RR-0031-1704

This PDF document was made available from www.rti.org as a public service of RTI International. More information about RTI Press can be found at <http://www.rti.org/rtipress>.

RTI International is an independent, nonprofit research organization dedicated to improving the human condition by turning knowledge into practice. The RTI Press mission is to disseminate information about RTI research, analytic tools, and technical expertise to a national and international audience. RTI Press publications are peer-reviewed by at least two independent substantive experts and one or more Press editors.

Suggested Citation

Sellgren, K. L., Gregory, C. W., Hunt, M. I., Raut, A. S., Hawkins, B. T., Parker, C. B., Klem, E. J. D., Piascik, J. R., and Stoner, B. R. (2017). *Development of an Electrochemical Process for Blackwater Disinfection in a Freestanding, Additive-Free Toilet*. RTI Press Publication No. RR-0031-1704. Research Triangle Park, NC: RTI Press. <https://doi.org/10.3768/rtipress.2017.rr.0031.1704>

This publication is part of the
RTI Press Research Reports series.

RTI International
3040 East Cornwallis Road
PO Box 12194
Research Triangle Park, NC
27709-2194 USA

Tel: +1.919.541.6000
E-mail: rtipress@rti.org
Website: www.rti.org

©2017 RTI International. All rights reserved. Credit must be provided to the author and source of the document when the content is quoted. No part of this document may be reproduced without permission in writing from the publisher. RTI International is a registered trademark and a trade name of Research Triangle Institute.

<https://doi.org/10.3768/rtipress.2017.rr.0031.1704>

www.rti.org/rtipress

Contents

About the Authors	i
Acknowledgments	ii
Abstract	ii
Introduction	1
Materials and Methods	2
Bacterial Culture and Excreta Collection	2
Experimental Methods and Apparatus	3
Analytical Measurements	4
Microbial Enumeration	4
Results	4
Model Wastewater Characteristics and Running Conditions	4
Energy Comparison for <i>E. coli</i> Inactivation by BDD and MMO	5
Oxidant Generation in Model Wastewater	6
Optimization of the MMO Electrode System	7
Impact of Fecal Material	8
Discussion	10
References	11

About the Authors

Katelyn L. Sellgren, PhD, is a chemical engineer in the Engineered Materials, Devices and Systems Division at RTI.

Christopher W. Gregory, MSEE, was an electron engineer in the Electronics and Applied Physics Division at RTI (now the Engineered Materials, Devices and Systems Division) at the time of data collection. He is currently a research engineer at Micross Components.

Michael I. Hunt, BS, was an electrical engineer in the Electronics and Applied Physics Division (now the Engineered Materials, Devices and Systems Division) at RTI at the time of data collection. He is currently an electronic hardware maintenance engineer with the Northrop Grumman Corporation.

Akshay S. Raut, PhD, was a postdoctoral associate at Duke University in Durham, NC, at the time of data collection. He is now a process technology engineer at the Intel Corporation.

Brian T. Hawkins, PhD, is a research biologist in the Engineered Materials, Devices and Systems Division at RTI.

Charles B. Parker, PhD, is the director of the nanomaterials and thin films laboratory at Duke University in Durham, NC.

Ethan J. D. Klem, PhD, is a research scientist in the Engineered Materials, Devices and Systems Division at RTI.

Jeffrey R. Piascik, PhD, is a senior research electron engineer in the Engineered Materials, Devices and Systems Division at RTI.

Brian R. Stoner, PhD, is an RTI Distinguished Fellow in materials and electronic technologies.

Acknowledgments

E. coli K12 (ATCC #10798) source plates were generously provided by Dr. Jeffrey Glass, Duke University.

Funding was provided by a grant from the Bill & Melinda Gates Foundation, Reinvent the Toilet Challenge.

Abstract

Electrochemical disinfection has gained interest as an alternative to conventional wastewater treatment because of its high effectiveness and environmental compatibility. Two and a half billion people currently live without improved sanitation facilities. Our research efforts are focused on developing and implementing a freestanding, additive-free toilet system that treats and recycles blackwater on site. In this study, we sought to apply electrochemical disinfection to blackwater. We compared commercially available boron-doped diamond (BDD) and mixed metal oxide (MMO) electrodes for disinfection efficiency in *E. coli*-inoculated model wastewater. The MMO electrodes were found to be more efficient and thus selected for further study with blackwater. The energy required for disinfection by the MMO electrodes increased with the conductivity of the medium, decreased with increased temperature, and was independent of the applied voltage. Fecal contamination considerably increased the energy required for blackwater disinfection compared to model wastewater, demonstrating the need for testing in effluents representing the conditions of the final application.

Introduction

An estimated 2.5 billion people live without improved sanitation facilities and as a consequence resort to open defecation, creating dangerous health situations (Wardlaw, Salama, Brocklehurst, Chopra, & Mason, 2010, p. 871; World Health Organization & UNICEF, 2014). In a study conducted in 2002, researchers concluded that without intervention, as many as 135 million people could die from preventable water-related diseases by 2020 (Gleick, 2002, pp. 1, 6). Gleick projected that even if the UN Millennium Development Goals were achieved, as many as 76 million people could still die of these diseases by 2020 (Gleick, 2002, pp. 1, 7, 8). Although 2.1 billion people have gained access to improved sanitation since 1990, the Millennium Development Goal for sanitation was missed by almost 700 million people (World Health Organization & UNICEF, 2015, pp. 13, 20-21, 90). The most underserved populations are those in rural areas and areas where implementation of solutions is complicated by lack of access to dependable electrical and clean water infrastructures, which are required for modern sanitation facilities (World Bank, 2013). For these reasons, the Bill & Melinda Gates Foundation

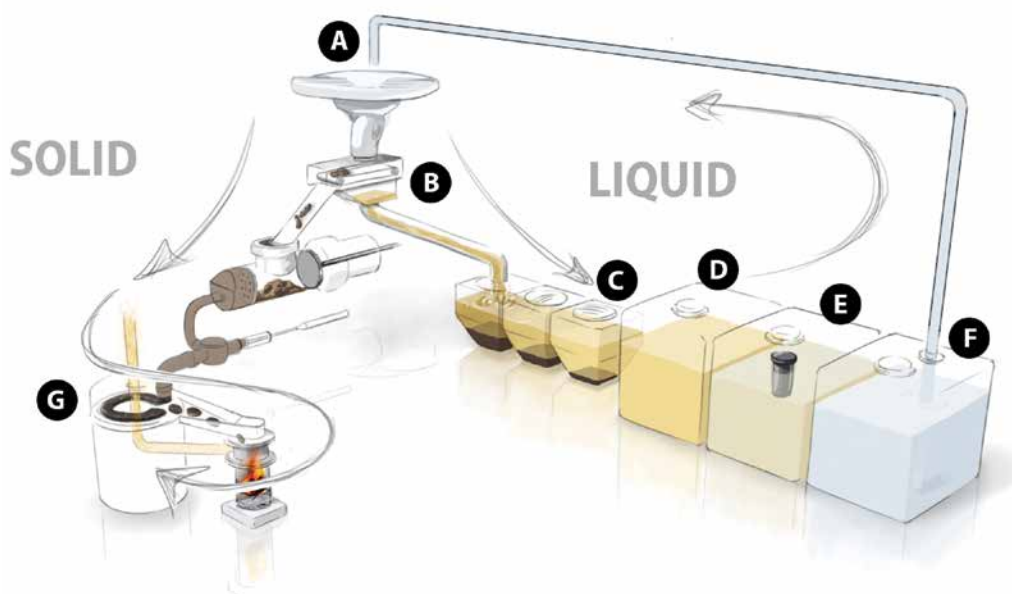
launched the “Reinvent the Toilet Challenge,” aimed at developing and implementing sustainable sanitation facilities that provide hygienic and affordable methods of waste disposal by treating excreta in unconnected, decentralized locations (Bill & Melinda Gates Foundation, n.d.). The challenge calls on grantees to develop a standalone toilet requiring no piped-in water, sewer connections, or outside electricity, and having a facility cost of less than 5 cents per person per day. In response to this challenge, we sought to apply electrochemical disinfection to blackwater (urine, feces, and flush water). This study is intended to support the continued development of an electrochemical blackwater disinfection system, which will ultimately be integrated into a modular toilet system designed and developed by RTI International researchers for use in low-income countries and rural areas (Figure 1). Development and testing of the other components of this system (solids processing and energy recovery) are described elsewhere (Stokes, Baldasaro, Bulman, & Stoner, 2014).

Electrochemical water treatment is a versatile technology capable of mineralizing organic matter, eliminating nitrogen species, and neutralizing pathogens, including *E. coli* (Bergmann & Koparal,

Figure 1. Modular toilet designed and developed by RTI researchers for low-income countries

Note: (A) Squat plate user interface (B) Solid-liquid separator (C) Series of baffle tanks through which liquids are diverted to allow any remaining solid particulate to settle out (D) Pre-process tank where the settled liquid is sent until the desired volume is collected (E) Process tank where liquid volume is electrochemically disinfected (F) Holding tank where treated liquid is stored until needed for recycling (G) Solids are processed and burned. The heat from burning is used to generate electricity through thermo-electrics.

Source: ©RTI International



2005; Drees, Abbaszadegan, & Maier, 2003; Huang et al., 2016; Jeong, Kim, Cho, Choi, & Yoon, 2007). Electrochemical processes do not require elevated temperatures (though they may benefit from them) or the addition of potentially hazardous chemical components, and they are considered less dangerous than non-electrochemical processes (Kuhn & Mortimer, 1973; Schmalz, Dittmar, Haaken, & Worch, 2009). Moreover, they can be tailored to minimize power losses and are well-suited for process automation and control, reducing the need for user intervention (Dominguez-Ramos, Aldaco, & Irabien, 2010). Electrochemical disinfection is driven by the production of oxidants, such as chlorine and hypochlorite, from naturally occurring chloride in the process solution. Chloride is the second-most abundant solute in urine (Putnam, 1971). Chlorine has been shown to effectively reduce incidence of diarrheal disease by 29 percent when used as point-of-use treatment of drinking water through the inactivation of enteric pathogens (Arnold & Colford, 2007, p. 357; Jeong, Kim, & Yoon, 2006). Disinfection and mineralization of organics takes place either directly on the electrode surface or indirectly in the bulk solution depending on the electrode material, oxidants produced, and oxygen overvoltage (Jeong, Kim, & Yoon, 2009; Jeong et al., 2006; Kraft et al., 1999). For a more detailed review on these parameters, see Anglada, Urriaga, and Ortiz (2009).

To date, electrochemical disinfection has been shown to be effective in killing a variety of microorganisms in modelled wastewaters (Bergmann & Koparal, 2005; Drees et al., 2003; Jeong et al., 2009; Jeong et al., 2007; Jeong et al., 2006; Li et al., 2004), saline and ballast waters (Li, Ding, Lo, & Sin, 2002; Nanayakkara, Alam, Zheng, & Chen, 2012), and secondary treated water from wastewater treatment plants (Schmalz et al., 2009). Electrochemical disinfection has also been used in the remediation of pollutants in municipal and industrial wastewaters (Adhoum & Monser, 2004; Mohan, Balasubramanian, & Basha, 2007; Pulgarin, Adler, Peringer, & Comninellis, 1994; Szpyrkowicz, Kaul, Neti, & Satyanarayan, 2005). However, research has not described, to the authors' knowledge, the disinfection efficacy of and energy requirements for electrochemical disinfection of concentrated blackwater, the material a freestanding

toilet system will receive and need to process. In this study, we aim to demonstrate the feasibility of electrochemical disinfection in blackwater and describe the initial phases of testing. Testing goals were (1) to evaluate easily attainable electrode materials with an accessible supply chain, requiring no post-purchase modifications, that can be used and implemented by unskilled workers; (2) to define the electrical energy input needed for *E. coli* disinfection in modeled wastewater; and (3) to investigate the impact of fecal material on disinfection kinetics and energy requirements in a reaction volume relevant to implementation in a freestanding toilet system.

Materials and Methods

Bacterial Culture and Excreta Collection

E. coli K12 (American Type Culture Collection [ATCC] #10798) source plates were maintained at 4°C on lysogeny broth (LB) agar substrates. The LB agar was a mixture of 10 g/L tryptone, 5 g/L yeast extract, 10 g/L NaCl, and 15 g/L agar in deionized water, sterilized at 121°C for 15 minutes. The sterilized LB agar was then cooled to a temperature allowing handling and sterilely dispersed into sterile 100 mm × 15 mm petri dishes to a depth of 5 mm. The agar-filled petri dishes were cooled, solidified, and stored sterilely upside-down at 4°C until use. *E. coli* were expanded on the above agar substrate for 24 hours at 37°C by spreading one colony from the source plate. After expansion, two average colonies were selected and added to 0.5 L of LB. LB consisted of 10 g/L tryptone, 5 g/L yeast extract, and 10 g/L NaCl in deionized water. The broth was sterilized at 121°C for 15 minutes and then cooled to a temperature allowing handling before addition of *E. coli* colonies. The *E. coli*-inoculated broth was incubated for 24 hours at 37°C before addition to the testing apparatus, ensuring that *E. coli* were in the exponential growth phase (DNA Learning Center, n.d.). Urine and fecal samples were collected from healthy volunteers 20–50 years of age. Urine was collected in sterile 1 L bottles and stored at 4°C until use. Feces was collected in plastic toilet hats and stored at 4°C until use. Collection methods were approved by RTI's institutional review board.

Experimental Methods and Apparatus

Two different volume (10 L or 60.5L) polyethylene containers were used as reaction vessels. Each had an 8-inch sample port on the top wall. The containers were housed in a fume hood. Each model wastewater experiment utilized a well-mixed composition of 68 percent deionized water, 25 percent urine, and 7 percent *E. coli*-inoculated broth.

Blackwater was obtained from the prototype toilet system (Figure 1). For this pilot study, initial assumptions of toilet use patterns were made for 10 users per day and included two urinations and one defecation (with urination) per person per day. On the basis of an approximate 24-hour urine volume of 1.5–2 L per person (Curhan, Willett, Speizer, & Stampfer, 2001, p. 2293), each urine flush contained 1.5 L of flush water (treated recycled water) and 0.5 L of urine. Each fecal flush contained approximately 200 g of wet feces, 1.5 L of flush water, and 0.5 L of urine (Rose, Parker, Jefferson, & Cartmell, 2015, p. 1833). Thus, the wastewater requiring treatment contained 25 percent urine in recycled flush water contaminated with feces. This was also the basis for the composition of the model wastewater described above.

The BDD electrochemical cell was inserted in a flow-through configuration in which the sample solution was continuously pumped across the electrodes at 2 L/min. This speed was based on the capabilities of commonly available small form factor DC-powered pumps that fit within the expected power and cost budget of the integrated waste processing system. The MMO electrochemical cell was inserted into the bulk solution, and a pump circulated the fluid. The electrochemical cells were connected to a power source capable of delivering up to 32 V and 6 A and were run in constant voltage mode.

Table 1 lists the characteristics of the two cells used in this study. BDD electrodes were acquired from Advanced Diamond Technologies (Romeoville, IL). A niobium substrate with a 2 μm -thick boron-doped ultrananocrystalline diamond film served as the anode. A tungsten plate served as the cathode. The electrodes were 42 cm^2 and separated by a 2 mm gap. MMO electrodes containing ruthenium oxide

Table 1. Electrode characteristics

Specifications	BDD	MMO
Intended disinfection purpose	Industrial water treatment	In-ground pools (up to 30,000 gallons)
Electrode type	Boron Doped Diamond	Mixed Metal Oxide
Oxidant production	Mixed	Chlorinator
Expected lifetime (yr)	Unknown	1+
Cost	\$2,000	\$200
Electrode area (cm^2)	42	64
Voltage (V) used	12	12, 24, 32
Current (A) used	4	0.4, 1.5, 4

were obtained from Hayward (Elizabeth, NJ) in the form of a Salt & Swim 3C electrochemical cell. The cell consists of 13 dual-sided electrodes that are 64 cm^2 in area and separated by a 3 mm gap. The electrochemical cells were used as provided from the manufacturer.

For heated tests, a $\frac{1}{4}$ " inner diameter copper coil attached to a Haake 80 refrigerated/heating circulator (Thermo Scientific, Waltham, MA) was wound through the reaction vessel, heating the effluent without exposing it to the heating liquid. The circulator was set to 90°C. Wastewater was heated for 1 hour before the start of electrochemical experiments. The temperature on the heated bath was adjusted as needed to maintain the experimental effluent at the desired temperature.

Experiments comparing the reaction rates of total chlorine, ammonia, and monochloramine for the MMO and BDD electrodes, as presented in Figure 3, were performed with the cell completely submerged in a 4 L beaker containing a model salt solution consisting of a 0.15 M NaCl solution in deionized water. For experiments involving urea, a concentration of 0.5 M was used. Experiments comparing chlorine production by the MMO electrode in model water, urine, and blackwater were sampled directly from the reaction vessel. Results for the MMO cell are from a minimum of two independent trials, whereas BDD results are from a single trial.

Analytical Measurements

Redox potential (ORP), pH, and conductivity were all measured before disinfection runs using a Myron L 6PFC^E Ultrameter II (Myron L Company, Carlsbad, CA) per the manufacturer's instructions. The conductivity cell and pH/ORP sensor wells were rinsed three times with sample effluent at each time point, before data recording. Reported data are from at least two individual trials.

Chlorine was measured using the N,N Diethyl-1,4 Phenylenediamine Sulfate (DPD) method (HACH method 8167) and a HACH DR 900 colorimeter (HACH, Loveland, CO). Samples were run per the manufacturer's instructions. Free or total chlorine pillow packs (HACH) were added to 10 mL of sample effluent, collected in the appropriate sample cell (HACH), allowed to react for 1 minute for free chlorine and 3 minutes for total chlorine, and then run using program 87 Chlorine, F&T PP MR. Results were recorded in mg/L. Blanks consisting of effluent without the addition of the DPD reagent were run before each sample. Free ammonia and monochloramine were measured in the same way, except that HACH method 10200 was used.

Microbial Enumeration

Samples for a three-tube most probable number (MPN) assay were collected using clean sterile pipettes before *E. coli* spiking of the model wastewater to ensure that the system was not contaminated before the start of the experiment. The first sample after *E. coli* addition and in fecal-contaminated effluent was drawn with a sterile pipette before applying a voltage ($t = 0$). Five 1 mL samples were taken from different places in the treatment tank, then mixed to ensure a representative MPN at each time point. The voltage was applied at the desired value, and samples were drawn in the same manner

using a new sterile pipette for each sample. All experiments were performed at least twice. For MPN quantification, the Food & Drug Administration (FDA) method for MPN from serial dilutions was used (Blodgett, 2010). Triplicate samples were serially diluted ($10^{-1} - 10^{-8}$) to 1 mL in LB and cultured in sterile 48-well plates. Samples were incubated at 37°C for 48 hours before being analyzed. Reported values are the average of at least two individual trials unless otherwise specified.

Results

Model Wastewater Characteristics and Running Conditions

Initial model wastewater characteristics for electrode comparison tests (BDD vs. MMO) are listed in Table 2, and initial conditions for energy evaluation tests using the MMO electrode are listed in Table 3. Conductivity of the urine solution was used as collected and not adjusted to a constant value for each trial. The variation in solution conductivity accounts for the differences in current between experiments. All trials were driven with a constant voltage condition to mimic field implementation of the unit. A fixed voltage of 12 V was used for electrode comparison tests, and voltage was varied from 12 V to 32 V for energy evaluation tests. The 32 V upper limit was selected on the basis of the recommended operating conditions provided by the cell manufacturer. In model wastewater tests, no *E. coli* was detected in the solution before inoculation. After inoculation, initial MPNs were on the order of 10^7 MPN/mL for electrode comparison tests and varied between 10^5 and 10^8 MPN/mL for energy optimization tests. The initial concentrations of *E. coli* used for testing are in line with those previously reported for sewage (Edberg, Rice, Karlin, & Allen, 2000).

Table 2. Initial model wastewater conditions for data presented in Figure 2 (n = 2)

Trial ID	Cell Type	Voltage (V)	Current (A)	ORP (mV)	Conductivity ($\mu\text{S}/\text{cm}$)	pH	Initial MPN (<i>E. coli</i> /mL)* 10^7
BC	BDD	12	3.5 ± 0.2	-237 ± 8	5112 ± 430	6.1 ± 0.1	8.1 ± 4.1
MC	MMO	12	0.4 ± 0.1	-187 ± 23	4602 ± 963	6.2 ± 0.1	6.7 ± 3.0

Notes: BDD = boron doped diamond; MMO = mixed metal oxide; MPN = most probable number.

Table 3. Initial model wastewater conditions for data presented in Figure 4 (n = 2)

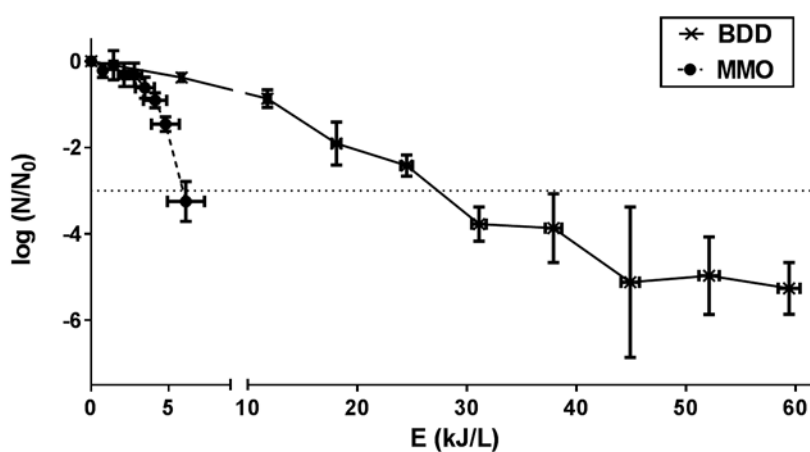
Trial ID	Voltage (V)	Current (A)	ORP (mV)	Conductivity ($\mu\text{S}/\text{cm}$)	pH	Initial MPN (<i>E. coli</i> /mL)* 10^7
MD1	12	0.4 \pm 0.0	-222 \pm 31	4721 \pm 990	6.3 \pm 0.2	2.0 \pm 1.1
MD2	24	1.6 \pm 0.1	-227 \pm 13	2801 \pm 163	5.9 \pm 0.1	5.5 \pm 5.5
MD3	24	3.3 \pm 0.7	-56 \pm 31	6144 \pm 628	5.9 \pm 0.0	3.1 \pm 1.6
MD4	32	3.2 \pm 0.6	-74 \pm 20	3304 \pm 427	6.0 \pm 0.2	6.3 \pm 4.8
MD(50°C)	24	3.3 \pm 0.8	-92 \pm 78	3915 \pm 485	6.2 \pm 0.1	2.0 \pm 0.5

Note: MPN = most probable number.

Energy Comparison for *E. coli* Inactivation by BDD and MMO

The World Health Organization's (WHO's) standard for using recycled wastewater for crop irrigation requires a three-log reduction in pathogen concentration (Mara, 2008, p. 11). For comparison, drinking water is expected to have no *E. coli*/100 mL (World Health Organization, 2011, p. 149). Because the final treated water in our system is not expected to be potable, but is expected to come into contact with humans when recycled for flushing or hand washing, we compare the BDD and MMO electrodes using the energy required for a three-log reduction (99.9 percent inactivation). A three-log reduction in *E. coli* was observed after 100 minutes with the BDD electrode system and 180 minutes with the MMO electrode system. In disinfection with the MMO system, there was an initial lag phase between 0 and 3 kJ/L, which was not seen in the BDD system (Figure 2). However, there is a slower initial slope of *E. coli* inactivation seen with the BDD electrode (Figure 2). This is most likely due to the difference in the chlorine production between the two electrode material types (Figures 3A and 3B) and the

Figure 2. Comparison of boron doped diamond (BDD) and mixed metal oxide (MMO) electrodes



Note: Shown are mean log reductions in most probable number (MPN) (\pm standard deviation [SD]) for 180-minute disinfection runs in 8 L model wastewater with the BDD (n = 2) and MMO (n = 3) electrodes, plotted against mean energy consumption (\pm SD). The horizontal dotted line indicates three-log reduction from the initial MPN and was used as the criterion for comparison.

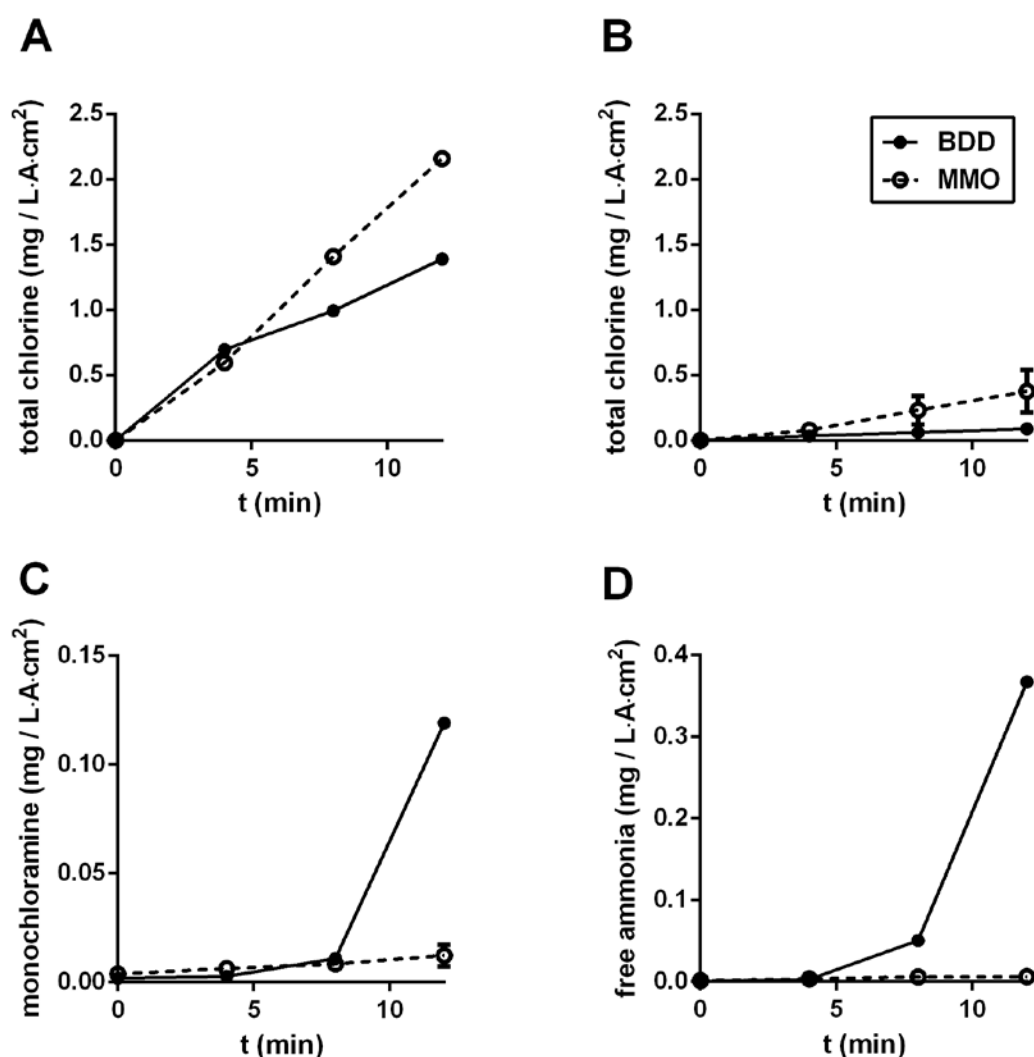
minimum contact time needed for chlorine to disinfect a population. The total energy requirement for the BDD electrode was found to be five times greater than for the MMO electrode system in achieving a three-log reduction in *E. coli* (Figure 2) because of the higher current draw at comparable conductivity.

Oxidant Generation in Model Wastewater

The MMO cell was more efficient at producing chlorine, generating 1.6 times more chlorine per current density after 12 minutes compared with the BDD cell (Figure 3A). In the presence of urea, the main solute in urine, chlorine production in both systems decreased. However, in the presence of urea, the MMO cell was not only more efficient at producing chlorine species but also produced more overall total chlorine than the BDD electrode system, generating more than four times the amount of total chlorine in 12 minutes (Figure 3B). The BDD cell was more proficient at producing monochloramine and ammonia in the presence of urea (Figure 3C and D).

Previous literature has shown oxidant generation to be the main mechanism for inactivation of bacteria in electrochemical systems (Bergmann & Koparal, 2005; Jeong et al., 2006; Li et al., 2004; Schmalz et al., 2009). Therefore, the higher energy requirement of the BDD electrochemical cell could be attributed to differing compositions of oxidants produced by the different electrode materials. BDD electrodes have high overvoltages, making them very efficient at producing electrochemical oxidants, particularly reactive oxygen species such as the hydroxyl radical ($\bullet\text{OH}$) (Jeong et al., 2009; Schmalz et al., 2009). Active electrodes with low overpotential, such as the MMO, allow the $\bullet\text{OH}$ radicals to favorably interact with the electrode surface, forming a higher-oxide surface.

Figure 3. Oxidant generation in model wastewater



Note: Oxidant generation is corrected for electrode area and process current. (A) Total chlorine generation in NaCl solution. (B) Total chlorine generation in NaCl solution containing urea. (C) Monochloramine generation in model water with urea. (D) Free ammonia generation in model water with urea.

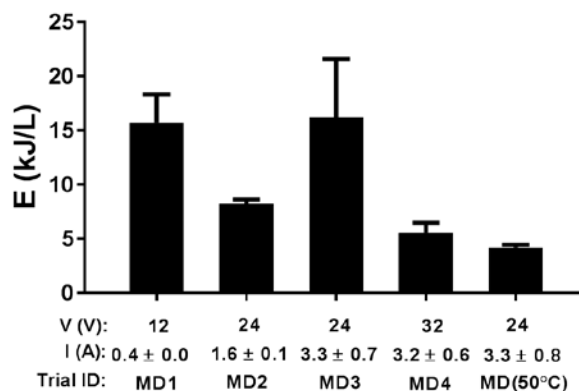
MMO electrodes show greater electrocatalytic activity toward the generation of chlorine due to direct oxidation on the electrode surface (Jeong et al., 2009; Jeong et al., 2006; Schmalz et al., 2009). Moreover, reactive oxygen species are less-potent inactivators of *E. coli* than the free chlorine species and do not provide residual disinfection (Jeong et al., 2009; Kerwick, Reddy, Chamberlain, & Holt, 2005; Schmalz et al., 2009). BDD electrodes do produce free chlorine, but only from indirect reactions in the bulk solution. We hypothesized that the addition of urea to the BDD system resulted in the $\bullet\text{OH}$ radical preferentially reacting with urea to form ammonia (World Health Organization, 2006). The ammonia could then react with free chlorine species to form monochloramine. Consequently, the oxidants formed in the BDD electrode system are significantly less potent than those produced in the MMO electrode system. Because of the potential for better production of a more-potent mix of oxidants with less energy demand, all further experiments were conducted with the MMO electrode system.

Optimization of the MMO Electrode System

For energy optimization tests, we examined the effect of varying initial model wastewater conditions and electrochemical cell running conditions. Table 3 summarizes the conditions tested in this study. First, we examined the time and energy differences needed to achieve full sterilization under the same conditions as comparison tests for the MMO cell (Trials MC vs. MD1). We found that full sterilization required 2.3 times as much time and 2.7 times as much energy compared to the three log reduction in trial MC (Figures 2 and 4). When the voltage was increased from 12V (MD1) to 24 V (MD3) while keeping conductivity consistent, we found that the current increased from 0.4A to approximately 3A (Table 3). This increase in voltage reduced the kill time by almost 95 percent; however, total energy remained constant (Figure 4). When voltage was again increased from 24 V (MD2) to 32 V (MD4) while maintaining a constant conductivity, we found that the kill time was again reduced by 75 percent, and the total energy decreased slightly (Figure 4). When the conductivity was decreased while the voltage was increased (MD1 v MD2), the time and energy needed

for disinfection were decreased. When voltage was maintained but conductivity was decreased (MD3 vs MD2), the kill time increased by 33 percent, and the total energy was reduced by half (Figure 4). These results indicate that simultaneously reducing the conductivity and increasing the voltage decreases the energy requirement while reducing the time needed for disinfection. Increasing the voltage alone reduces time, but not the energy requirement, and reducing the conductivity decreases the energy requirement but increases the time for disinfection. Thus, energy consumption will be lower in systems handling more-dilute (lower conductivity) effluents, regardless of the voltage applied.

Figure 4. Effects of voltage, conductivity, and temperature on disinfection energy



Note: Shown are the energies required for complete disinfection (most probable number [MPN] ≤ 3 / ml) of model wastewater with the MMO electrode at the voltages, currents, and temperatures indicated. Bars are mean \pm standard deviation. n = 2 per condition.

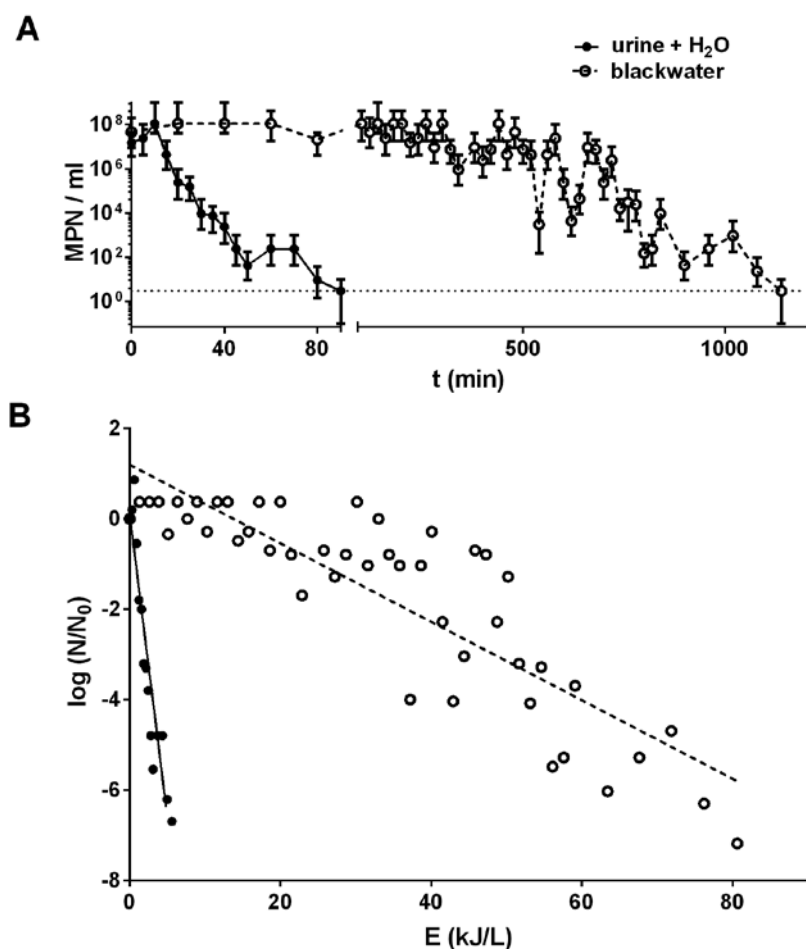
Given the apparent independence of energy consumption from the voltage applied, we explored the feasibility of lowering the energy needed for electrochemical disinfection by raising the temperature of the model wastewater to be treated. Previous studies have shown that a temperature of 60°C is needed to disinfect *E. coli* in wastewater by heat alone (Feachem, Bradley, Garelick, & Mara, 1983, p. 232). Separately, other studies have shown that increasing the liquid temperature accelerates electrochemical disinfection and decreases the overpotential at a given current density on metal oxide surfaces (Kuhn & Mortimer, 1973; Schmalz et al., 2009). The electrochemical process explored in this manuscript is intended to be used in a self-

contained toilet apparatus where solid waste (feces) will also need to be disinfected. Burning the solid waste could mitigate this problem, and the exhaust stream from the combustion unit could be used to heat the liquid disinfection process in addition to driving the electrochemical process through conversion to electrical energy (Stokes et al., 2014). On the basis of our assumptions for toilet usage, we estimate we will have enough heat energy to heat the liquid being treated to approximately 50°C (this modeling is a part of a different study and not shown here). We therefore decided to test how much electrochemical energy would be needed to achieve a full kill at this elevated temperature (MD3 v MD[50°C]). The model wastewater was heated from room temperature to 50°C over approximately 1 hour, without affecting starting MPNs, before electrochemical treatment. We found that the energy and time needed for disinfection by the electrochemical cell were reduced by 66 percent and 75 percent, respectively, from the comparable room temperature condition (Figure 4).

Impact of Fecal Material

Fecal contamination tests were run at 24 V and performed at a volume of 60 L. This volume represents the projected volume of 10 users in a single day, based on assumptions stated in the materials and methods section. Initial MPN values varied between 10^5 and 10^8 MPN/mL. Initial conditions for fecal contamination tests are listed in Table 4. Fecal contamination of the model wastewater increased

Figure 5. Effect of fecal matter on disinfection



Note: 60 L of model wastewater (urine + H₂O) or blackwater were disinfected using the mixed metal oxide (MMO) electrode system. (A) most probable number (MPN) \pm 95% confidence intervals plotted against active run time. (B) Log reduction plotted against energy. Data in A and B are from the same representative runs. The model wastewater condition was repeated twice at 8 L and 16 L with comparable results on a per liter basis, and the blackwater condition was repeated once at 60 L with comparable results.

both the time and energy needed for disinfection (Figure 5A and B). Interestingly, both disinfection curves show an initial shoulder, logarithmic decay, and final long tail phase, but the duration of each phase in the fecal-contaminated blackwater is greatly exaggerated compared to the model wastewater (Fig 5A).

Table 4. Initial conditions for fecal-contaminated blackwater and model water comparison tests

Trial ID	Voltage (V)	Current (A)	ORP (mV)	Conductivity ($\mu\text{S}/\text{cm}$)	pH	Initial MPN (<i>E. coli</i> /mL)* 10^7
Urine n = 1	24	2.6	103	4182	6.24	2.4
Feces n=2	24	2.1 \pm 0.6	-219 \pm 17	4659 \pm 1609	7.27 \pm 1.28	2.5 \pm 2.1

Because sodium and chloride ions make up almost 60 percent of the mass of the total salts present in normal urine (Putnam, 1971, p. 40), many previous studies on wastewater disinfection have used model wastewater systems containing no or low organic content (Bergmann & Kopalal, 2005; Drees et al., 2003; Jeong et al., 2009; Jeong et al., 2007; Jeong et al., 2006; Li et al., 2004). Our results show that modeling blackwater electrochemical processes in media without fecal contamination—even those with high chemical oxygen demand, such as urine—are not representative of fecal contaminated effluents, even though feces is composed almost entirely of water and organic material (Pierce, 1962).

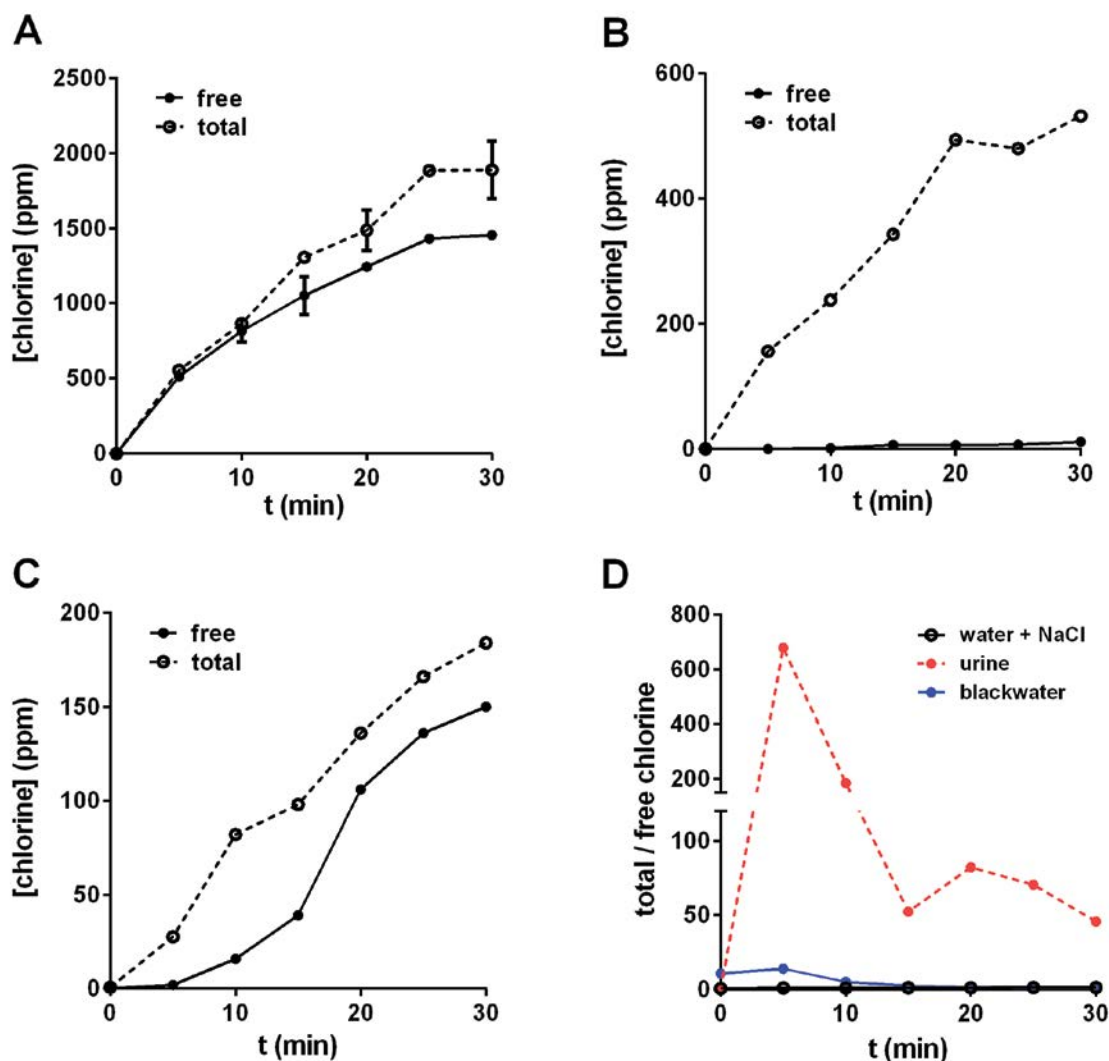
Several factors may contribute to the increased time and energy demands for electrochemical disinfection with the introduction of fecal contamination: (1) the diversity of human fecal bacteria (Newton et al., 2015) allows for the possibility of chlorine-resistant subpopulations, compared with the (presumably) homogenous population of cultured *E. coli*; (2) fecal particulate matter could shield bacteria from inactivation by chlorine (Winward, Avery, Stephenson, & Jefferson, 2008); (3) the association of organic chlorine demand with fecal particulate matter competing for free chlorine with particulate matter-associated bacteria (Dickenson & Sansalone, 2012); or (4) some combination of these factors.

Because it is assumed that chlorine is the main disinfecting oxidant, we compared chlorine production among a salt solution, pure urine, and sterilized blackwater (Figure 6A–C). The conductivities of the salt solution and blackwater were approximately 18,000 $\mu\text{S}/\text{cm}$, whereas that of the urine was 13,000 $\mu\text{S}/\text{cm}$. Total chlorine was more

than three times greater in the salt solution than in pure urine and an order of magnitude greater than in blackwater. Production of free chlorine was greatest in the salt solution. Free chlorine in blackwater was an order of magnitude lower than in the salt solution, and almost no free chlorine was measured in the urine solution.

Figure 6D shows the ratios of total to free chlorine in all three media. The ratio of total to free chlorine represents how much of the electrochemically generated chlorine is free and available for disinfection. Values near one suggest that all produced chlorine is in the free available state. In the salt solution, the ratio of total to free chlorine remained constant at around 1.2 throughout the 30-minute experiment, indicating that most of the chlorine produced was free and available for disinfection. Even though the blackwater had been pre-sterilized (with chlorine) and a small chlorine residual (0.07 ppm) remained, the ratio of total to free chlorine in the first 15 minutes was approximately 2.5, indicating a substantial organic chlorine demand in blackwater, even in the absence of live bacteria. Interestingly, the ratio of total to free chlorine production in pure urine was orders of magnitude greater than in both blackwater and the salt solution because very little free chlorine was measured over the course of the experiment (Figure 6B). This was consistent with the total and free chlorine levels observed during the model wastewater (25 percent urine in water) disinfection experiments, where free chlorine never exceeded 0.3 mg/L (data not shown).

Figure 6. Effect of media on chlorine production



Note: Total and free chlorine production are shown as a function of time in (A) salt water, (B) urine, and (C) disinfected blackwater. (D) Ratios of total to free chlorine in all three media.

Discussion

In this study, we examined the energy efficiencies of two electrode materials, BDD and MMO, in the inactivation of cultured *E. coli* added to a solution consisting of ~25 percent human urine in water. In initial tests, the MMO system was found to require less energy for disinfection than the BDD system, and processing conditions were further examined to determine minimal energy usage conditions with the MMO electrode system. Energy consumption was found to be less in lower-conductivity media regardless of voltage applied, but increasing the temperature of the medium to 50°C reduced

the energy required for disinfection at a given conductivity and applied voltage.

We also showed that the addition of fecal material to a urine solution significantly affects both the time and the energy needed for disinfection. The current paradigm for evaluating the disinfection capabilities of new and existing electrochemical technologies is based on the generation of oxidants, the main bacterial inactivators produced in wastewater disinfection systems (Kraft et al., 1999; Li et al., 2004; Schmalz et al., 2009). However, the results of this study suggest that the ability of a solution to produce large quantities of free chlorine is not necessarily

predictive of disinfecting capacity in urine or fecal-contaminated (blackwater) effluents, as *E. coli* were readily inactivated in diluted urine with minimal free chlorine generation.

Our research, a response to the Bill & Melinda Gates Foundation Reinvent the Toilet Challenge, is focused on developing a freestanding disinfection system for human waste that requires no added water, chemical reagents, or energy in excess of what can be produced at the point of use. In this report, we have shown MMO electrochemical water

disinfection systems to be effective at inactivating enteric pathogens, such as *E. coli*, in blackwater effluents, qualifying these systems as a promising avenue for further investigation toward the Challenge goals. Furthermore, we have demonstrated the critical importance of testing electrochemical disinfection processes in the wastewater that they are intended to treat, rather than relying on assumptions based on model wastewaters. Future investigations will focus on the effect of recycling treated water, water discharge standards, and the energy concerns related to these factors.

References

- Adhoum, N., & Monser, L. (2004). Decolourization and removal of phenolic compounds from olive mill wastewater by electrocoagulation. *Chemical Engineering and Processing: Process Intensification*, 43(10), 1281-1287. <https://doi.org/10.1016/j.cep.2003.12.001>
- Anglada, A., Urtiaga, A., & Ortiz, I. (2009). Contributions of electrochemical oxidation to waste-water treatment: Fundamentals and review of applications. *Journal of Chemical Technology and Biotechnology*, 84(12), 1747-1755. <https://doi.org/10.1002/jctb.2214>
- Arnold, B. F., & Colford, J. M., Jr. (2007). Treating water with chlorine at point-of-use to improve water quality and reduce child diarrhea in developing countries: A systematic review and meta-analysis. *American Journal of Tropical Medicine and Hygiene*, 76(2), 354-364.
- Bergmann, M. E. H., & Koparal, A. S. (2005). Studies on electrochemical disinfectant production using anodes containing RuO₂. *Journal of Applied Electrochemistry*, 35(12), 1321-1329. <https://doi.org/10.1007/s10800-005-9064-0>
- Bill & Melinda Gates Foundation. (n.d.). *What we do—Reinvent the Toilet Challenge—Strategy overview*. Retrieved November 18, 2016, from <http://www.gatesfoundation.org/What-We-Do/Global-Development/Reinvent-the-Toilet-Challenge>
- Blodgett, R. (2010). *BAM appendix 2: Most probable number from serial dilutions*. Retrieved from <http://www.fda.gov/Food/FoodScienceResearch/LaboratoryMethods/ucm109656.htm>
- Curhan, G. C., Willett, W. C., Speizer, F. E., & Stampfer, M. J. (2001). Twenty-four-hour urine chemistries and the risk of kidney stones among women and men. *Kidney International*, 59(6), 2290-2298. <https://doi.org/10.1046/j.1523-1755.2001.00746.x>
- Dickenson, J. A., & Sansalone, J. J. (2012). Distribution and disinfection of bacterial loadings associated with particulate matter fractions transported in urban wet weather flows. *Water Research*, 46(20), 6704-6714. <https://doi.org/10.1016/j.watres.2011.12.039>
- DNA Learning Center. (n.d.). *Bacterial culture techniques*. Retrieved November 18, 2016, from http://labprotocols.dnalc.org/files/017_bacteria_culture_techniques.pdf
- Dominguez-Ramos, A., Aldaco, R., & Irabien, A. (2010). Photovoltaic solar electrochemical oxidation (PSEO) for lignosulfonate waste water treatment. *Journal of Chemical Technology and Biotechnology*, 85, 821-830. <https://doi.org/10.1002/jctb.2370>
- Drees, K. P., Abbaszadegan, M., & Maier, R. M. (2003). Comparative electrochemical inactivation of bacteria and bacteriophage. *Water Research*, 37(10), 2291-2300. [https://doi.org/10.1016/S0043-1354\(03\)00009-5](https://doi.org/10.1016/S0043-1354(03)00009-5)
- Edberg, S. C., Rice, E. W., Karlin, R. J., & Allen, M. J. (2000). *Escherichia coli*: The best biological drinking water indicator for public health protection. *Journal of Applied Microbiology*, 88, 106s-116s.
- Feachem, R., Bradley, D., Garelick, H., & Mara, D. D. (1983). *Sanitation and disease health aspects of excreta and wastewater management*. New York, NY: John Wiley & Sons.

- Gleick, P. H. (2002). *Dirty water: Estimated deaths from water-related diseases 2000-2020*. Oakland, CA: Pacific Institute for Studies in Development, Environment, and Security.
- Huang, X., Qu, Y., Cid, C. A., Finke, C., Hoffmann, M. R., Lim, K., & Jiang, S. C. (2016). Electrochemical disinfection of toilet wastewater using wastewater electrolysis cell. *Water Research*, 92, 164-172. <https://doi.org/10.1016/j.watres.2016.01.040>
- Jeong, J., Kim, C., & Yoon, J. (2009). The effect of electrode material on the generation of oxidants and microbial inactivation in the electrochemical disinfection processes. *Water Research*, 43(4), 895-901. <https://doi.org/10.1016/j.watres.2008.11.033>
- Jeong, J., Kim, J. Y., Cho, M., Choi, W., & Yoon, J. (2007). Inactivation of *Escherichia coli* in the electrochemical disinfection process using a Pt anode. *Chemosphere*, 67(4), 652-659. <https://doi.org/10.1016/j.chemosphere.2006.11.035>
- Jeong, J., Kim, J. Y., & Yoon, J. (2006). The role of reactive oxygen species in the electrochemical inactivation of microorganisms. *Environmental Science & Technology*, 40(19), 6117-6122. <https://doi.org/10.1021/Es0604313>
- Kerwick, M. I., Reddy, S. M., Chamberlain, A. H. L., & Holt, D. M. (2005). Electrochemical disinfection, an environmentally acceptable method of drinking water disinfection? *Electrochimica Acta*, 50(25-26), 5270-5277. <https://doi.org/10.1016/j.electacta.2005.02.074>
- Kraft, A., Stadelmann, M., Blaschke, M., Kreysig, D., Sandt, B., Schroder, F., & Rennau, J. (1999). Electrochemical water disinfection—Part I: Hypochlorite production from very dilute chloride solutions. *Journal of Applied Electrochemistry*, 29(7), 859-866.
- Kuhn, A. T., & Mortimer, C. J. (1973). The kinetics of chlorine evolution and reduction on titanium-supported metal oxides especially RuO₂ and IrO₂. *Journal of the Electrochemical Society*, 120(2), 231-234. <https://doi.org/10.1149/1.2403425>
- Li, X. Y., Diao, H. F., Fan, F. X. J., Gu, J. D., Ding, F., & Tong, A. S. F. (2004). Electrochemical wastewater disinfection: Identification of its principal germicidal actions. *Journal of Environmental Engineering*, 130(10), 1217-1221. [https://doi.org/10.1061/\(Asce\)0733-9372\(2004\)130:10\(1217\)](https://doi.org/10.1061/(Asce)0733-9372(2004)130:10(1217))
- Li, X. Y., Ding, F., Lo, P. S. Y., & Sin, S. H. P. (2002). Electrochemical disinfection of saline wastewater effluent. *Journal of Environmental Engineering*, 128(8), 697-704. [https://doi.org/10.1061/\(Asce\)0733-9372\(2002\)128:8\(697\)](https://doi.org/10.1061/(Asce)0733-9372(2002)128:8(697))
- Mara, D. (2008). *Third edition of the guidelines for the safe use of wastewater, excreta and greywater in agriculture and aquaculture: Guidance note for programme managers and engineers. A numerical guide to volume 2 of the guidelines and practical advice on how to transpose them into national standards*. Geneva, Switzerland: World Health Organization.
- Mohan, N., Balasubramanian, N., & Basha, C. A. (2007). Electrochemical oxidation of textile wastewater and its reuse. *Journal of Hazardous Materials*, 147(1-2), 644-651. <https://doi.org/10.1016/j.jhazmat.2007.01.063>
- Nanayakkara, K. G. N., Alam, A. K. M. K., Zheng, Y.-M., & Chen, J. P. (2012). A low-energy intensive electrochemical system for the eradication of *Escherichia coli* from ballast water: Process development, disinfection chemistry, and kinetics modeling. *Marine Pollution Bulletin*, 64(6), 1238-1245. <https://doi.org/10.1016/j.marpolbul.2012.01.018>
- Newton, R. J., McLellan, S. L., Dila, D. K., Vineis, J. H., Morrison, H. G., Eren, A. M., & Sogin, M. L. (2015). Sewage reflects the microbiomes of human populations. *Mbio*, 6(2), e02574-e02514. <https://doi.org/10.1128/mBio.02574-14>
- Pierce, C. M. (1962). *The detailed composition of feces, sweat, and urine: An annotated bibliography*. Sunnyvale, California: Lockheed Missiles and Space Company.
- Pulgarin, C., Adler, N., Peringer, P., & Comminellis, C. (1994). Electrochemical detoxification of a 1,4-benzoquinone solution in waste-water treatment. *Water Research*, 28(4), 887-893. [https://doi.org/10.1016/0043-1354\(94\)90095-7](https://doi.org/10.1016/0043-1354(94)90095-7)
- Putnam, D. F. (1971). *Composition and concentrative properties of human urine*. Washington, DC: NASA.
- Rose, C., Parker, A., Jefferson, B., & Cartmell, E. (2015). The characterization of feces and urine: A review of the literature to inform advanced treatment technology. *Critical Reviews in Environmental Science and Technology*, 45(17), 1827-1879. <https://doi.org/10.1080/10643389.2014.1000761>

- Schmalz, V., Dittmar, T., Haaken, D., & Worch, E. (2009). Electrochemical disinfection of biologically treated wastewater from small treatment systems by using boron-doped diamond (BDD) electrodes—Contribution for direct reuse of domestic wastewater. *Water Research*, 43(20), 5260-5266. <https://doi.org/10.1016/j.watres.2009.08.036>
- Stokes, C. D., Baldasaro, N. G., Bulman, G. E., & Stoner, B. R. (2014). Thermoelectric energy harvesting for a solid waste processing toilet. In N. K. Dhar, P. Balaya, & A. K. Dutta (Eds.), *Proc. SPIE 9115, Energy Harvesting and Storage: Materials, Devices, and Applications V*.
- Szpyrkowicz, L., Kaul, S. N., Neti, R. N., & Satyanarayan, S. (2005). Influence of anode material on electrochemical oxidation for the treatment of tannery wastewater. *Water Research*, 39(8), 1601-1613. <https://doi.org/10.1016/j.watres.2005.01.016>
- Wardlaw, T., Salama, P., Brocklehurst, C., Chopra, M., & Mason, E. (2010). Diarrhoea: Why children are still dying and what can be done. *Lancet*, 375(9718), 870-872. [https://doi.org/10.1016/S0140-6736\(09\)61798-0](https://doi.org/10.1016/S0140-6736(09)61798-0)
- Winward, G. P., Avery, L. M., Stephenson, T., & Jefferson, B. (2008). Chlorine disinfection of grey water for reuse: Effect of organics and particles. *Water Research*, 42(1-2), 483-491. <https://doi.org/10.1016/j.watres.2007.07.042>
- World Bank. (2013). *ESMAP annual report 2013*. Washington, DC: World Bank.
- World Health Organization. (2006). *Guidelines for safe recreational water environments. Volume 2, Swimming pools and similar environments*. Geneva, Switzerland: World Health Organization.
- World Health Organization. (2011). *Guidelines for drinking-water quality* (4th ed.). Geneva, Switzerland: World Health Organization.
- World Health Organization, & UNICEF. (2014). *Progress on sanitation and drinking-water—2014 update*. Geneva, Switzerland: World Health Organization.
- World Health Organization, & UNICEF. (2015). *25 years progress on sanitation and drinking water: 2015 update and MDG assessment*. Geneva, Switzerland: World Health Organization.

RTI International is an independent, nonprofit research institute dedicated to improving the human condition. Clients rely on us to answer questions that demand an objective and multidisciplinary approach—one that integrates expertise across the social and laboratory sciences, engineering, and international development. We believe in the promise of science, and we are inspired every day to deliver on that promise for the good of people, communities, and businesses around the world.

The RTI Press complements traditional publication outlets by providing another way for RTI researchers to disseminate the knowledge they generate. This PDF document is offered as a public service of RTI International.