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Application of Electrochemical Disinfection Process Using Aluminum Electrodes for Efficient Removal of Coliforms from Wastewater

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ABSTRACT

In this work, it was attempted to evaluate and demonstrate disinfection effectiveness of an electrochemical process to entirely remove coliform from wastewater effluent following secondary treatment. For the tests, an experimental bench-scale batch electrochemical cell was constructed, and aluminum electrodes were employed in the electro-disinfection reactor. In the electric disinfection phase, wastewater samples were put in the reactor/disinfector and a direct current (DC) was applied to it. According to findings, a significant decrease occurred in the total number of coliforms in the treated wastewater, and a high improvement occurred in the effluent properties. At a contact time of 15 min and a current density of 5.5 mA/cm2, led to a bacterial killing effectiveness of 97.7% or above. As the current density and contact time increased, a general increase occurred in the bacterial killing efficiency, and the effect of the two above-mentioned factors was much greater than the effect of salinity. Moreover, according to the experimental data, the removal efficiency of chemical oxygen demand (COD) and total suspended solids (TSS) by the aluminum electrodes were 78.50% and 99.93%, respectively. The findings indicate the applicability of the proposed electrochemical treatment to wastewater effluent. Nevertheless, to be able to apply this system at an industrial scale in the future, it is necessary to conduct more research into the optimum operation conditions and make an in-depth comparison of energy consumptions between the electrochemical treatment and the conventional approaches.

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INTRODUCTION

Disinfection is defined as the deactivation and/or elimination of disease-causing (pathogenic) microorganisms so that the spread of waterborne diseases can be prevented (Bakheet et al., 2020). Various approaches are employed to disinfect wastewater and enhance the quality of water for downstream uses, including crop irrigation crops, shellfish cultivation, public water supply, or swimming purposes (Gil et al., 2019; Martínez-Huitle & Brillas, 2021). A body of water that receives wastewater effluents with inadequate disinfection is a definite place for the spread of diseases (Zand & Abyaneh, 2019).

The end stage of municipal wastewater treatment is disinfection (Hand & Cusick, 2021). The most common approach for disinfecting wastewater is still chlorination. Nevertheless, although it is effective at present, important safety issues and ecological concerns are related to its application (Thostenson et al., 2018; Wang et al., 2020). Storing, handling, and shipping of different types of chlorine is a threat to public health, which has led to the issuance of more safety guidelines (Li et al., 2020). There are some other defects related to the employment of

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this compound, which include the production of unwanted carcinogenic agents (Mazhar et al., 2020); damage to aquatic ecosystems by chlorine residues (Xu et al., 2018); increase in the amount of total dissolved solids in the effluent after treatment (Du et al., 2017); resistance of a number of parasitic species to small amounts of chlorine (Li et al., 2017); established long term impacts of the disposal of dechlorinated chemicals into the ecosystem (Léziart et al., 2019). Therefore, a lot of wastewater treatment plants that consume huge volumes of chlorine gas per year have decided to replace chlorine with an alternative disinfectant.

It has been attempted to avoid a number of these issues by developing alternatives to chlorine gas and hypochlorite disinfection. Although techniques including ultraviolet radiation (Sullivan et al., 2017), ozonation (Wei et al., 2017), and ClO_2 treatment (Rougé et al., 2018) have shown efficient removal of pathogenic microorganisms from water and wastewater, they are normally costly or more complicated than chlorination (Fu et al., 2019; Zand & Abyaneh, 2020). Thus, there is an urgent need for novel disinfection methods showing reliability, effectiveness, cost-effectiveness, and eco-friendliness.

According to the test results, the electro-disinfection method is able to eliminate a broad range of microorganisms, encompassing bacteria and viruses, from raw water, tap water, milk, liquid food, and treated wastewater effluents over a short contact period (Zheng et al., 2017). As its primary benefit, the electro-disinfection method produces in situ disinfected conditions in treatment system; hence, it lacks the shortcomings of conventional chlorination including the storage and transportation of dangerous chlorine (Ghernaout et al., 2020).

Wastewater disinfection approaches must be able to address various site-specific challenges. Adherence to specific limitations on the quality of discharging or entering water is set by the national pollutant discharge removal system is usually required (Hadi et al., 2019). Based on regulations in different states and the features of the specific receiving body of water, these limitations can vary (Muddemann et al., 2019). In this study, this need is addressed by evaluating an innovative electro-disinfection technology experimentally to disinfect wastewater effluent, examine the impacts of important operational parameters, e.g., energy consumption, residence time, and salinity of the wastewater, on the performance of the process.

MATERIALS & METHODS

Experimental program and setup

The experimental phase was carried out by assembling and operating a bench-scale batch electrochemical cell. According to the literature, aluminum electrodes were used in the electro-disinfection reactor (Devlin et al., 2019; Shahedi et al., 2020). The cell is in the shape of a cylinder and consists of a tubular column as external electrode, and a similar tube with a smaller diameter inserted in the middle in a concentric manner. Both tubes are supported by a plastic section that seals the central opening and provides an annular gap between the tubes for the placement of the water sample. Table 1 gives the dimensions of the tube, together with the total anode/cathode/water contact area. Figure 1 gives the experimental setup.

The rectifier was capable of automatic polarity reversal, which enables the electrochemical reaction at average electrochemical potentials determined by surface groups-electrolytic solution equilibrium. Moreover, the periodic current reversal leads to a self-cleaning impact and in turn alters the composition of compounds formed at the electrodes; therefore, deposition and other undesirable cumulative impacts are prevented. The direct current (DC) output could be adjusted by changing either the current in from 0 to 40 amp or voltage from of 0 to 20 v. The current values were read by a multimeter (AMPROBE®, model ACDC-400).

Treatment of wastewater sample

The samples were extracted from the wastewater effluent of secondary clarifier following

Table 1. Dimensions of the electrochemical cell

Aluminum electrochemical cell		Dimensions
Inner pipe	Inside diameter (cm)	5.26
	Outside diameter (cm)	6.03
	Thickness (cm)	0.40
	Length (cm)	71.00
Outer pipe	Inside diameter (cm)	7.74
	Outside diameter (cm)	8.89
	Thickness (cm)	0.55
	Length (cm)	66.00
Anode/cathode/water contact area (cm^2)		1828

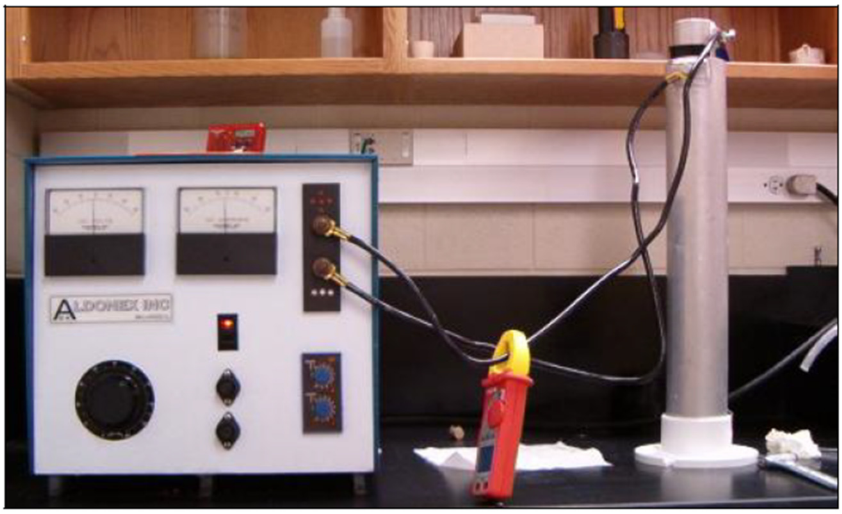

Fig. 1. Electrochemical cell- power supply connection

Table 2. Principal features of wastewater effluents

Property	Average values	Ranges
Coliforms (MPN/100 ml)	4.78×10^6	1.68×10^6-7.13×10^6
COD (mg/l)	69	45-106
TSS (mg/l)	16	5-40
Cl^- concentration (mg/l)	141	120-163
pH	7.12	6.77-7.32
Temperature (°C)	21.0	16.9-24.7

treatment with activated sludge at Tehran Eastern Wastewater Treatment Plant. Following sample extraction, it was immediately transferred to laboratory for disinfection in the electrochemical cell. An acceptably consistent quality was observed for the quality of the treated wastewater during the tests. Table 2 summarizes the principal water quality features. By adding sodium chloride, water salinity (chloride content) was increased in a number of experiments.

Disinfection efficiency of electrochemical cell was evaluated by using an orthogonal array in empirical setup. This was done to address the three principal parameters believed to affect the disinfection efficiency: current density, chloride concentration, and contact time. By employing the full-factorial design, many factors can be taken into account at the same time to discover their interactions. The actual maximum and minimum values of each factor considered for the experimental design with the cell are provided in Table 3.

Table 3. Experimental parameters and associated values

Parameter	Unit	Minimum value	Maximum value
Current density	mA/cm^2	1.5	5.5
Contact time	min	5	15
Cl$^-$ concentration	mg/L	130	1000

Table 4. The layout of experimental design

Run	Current density (mA/cm^2)	Contact time (min)	Chlorides (mg/l)
1	5.5	5	130
2	5.5	15	130
3	3.5	10	500
4	1.5	5	130
5	1.5	15	130
6	5.5	5	1000
7	1.5	5	1000
8	1.5	15	1000
9	5.5	15	1000
10	5.5	5	130
11	3.5	10	500
12	5.5	15	130

Based on the test design achieved using Design-Ease® 6.0 software from Stat-Ease®, 12 runs were carried out. Table 4 presents the design matrix, consisting of all tested operational conditions (which include two central points and replicates). By conducting the runs in a randomized manner, the effect of any underlying variables including temperature, sampling time, or humidity was offset. To consider variability and lower test errors, each test was carried out in triplicates.

Laboratory analyses

At different combinations of the test conditions, a pre- and post-disinfection analysis was conducted on the water for total coliform bacteria, chlorides, total chlorine, total chemical oxygen demand (COD), aluminum concentration, and total suspended solids (TSS). With the exception of the total coliform count, all the other analytical methods were conducted by following the guidelines for the standard examination methods of water and wastewater (APHA, 2005) or guidelines provided in analytical procedures manual of HACH direct reading spectrophotometer DR/2010 (HACH, 1999).

To further simplify and reduce the time demand, total coliform bacteria were considered as the indicator microorganism for disinfection investigation. To catalogue them, 3M Petrifilm™ E. coli/coliform count plates were used. Each run was followed by the dechlorination of a sample of about 200 ml with extra $Na_2S_2O_3$ to avoid potential killing by residual chlorine before enumeration. By considering the dilution factors, the total coliform bacteria concentration of the initial samples was measured.

RESULTS AND DISCUSSION

By selecting two levels for each variable, the three principal operation conditions, namely contact time, salinity, and current density, were evaluated simultaneously, as shown in Table 3); in doing so, 12 runs were carried out in total, as can be seen in Figure 2. Given the somewhat smaller initial total concentration of coliform in the wastewater than that on the remaining

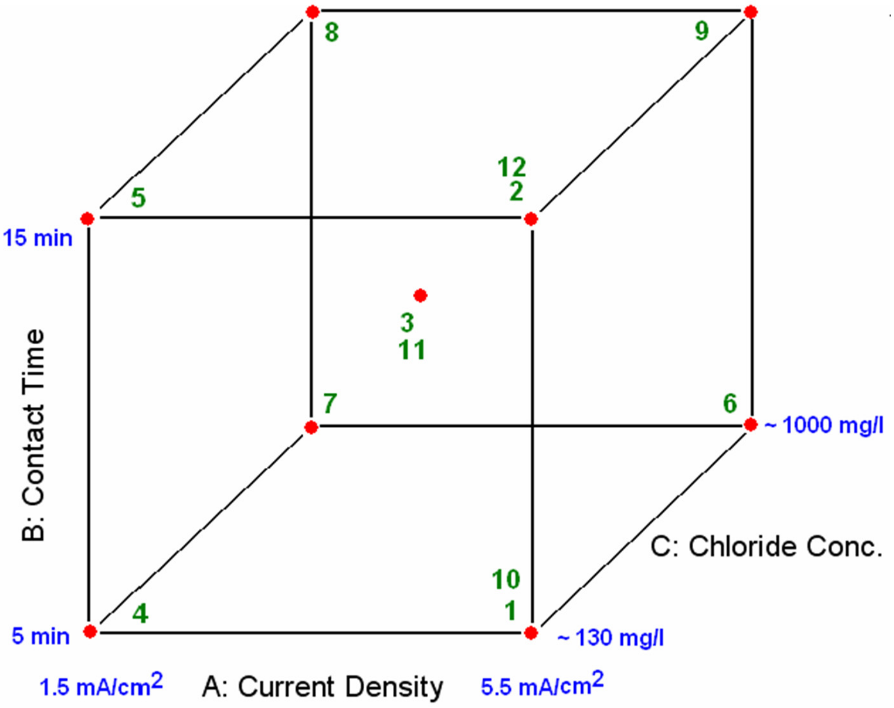

Fig. 2. Empirical parameters in each run

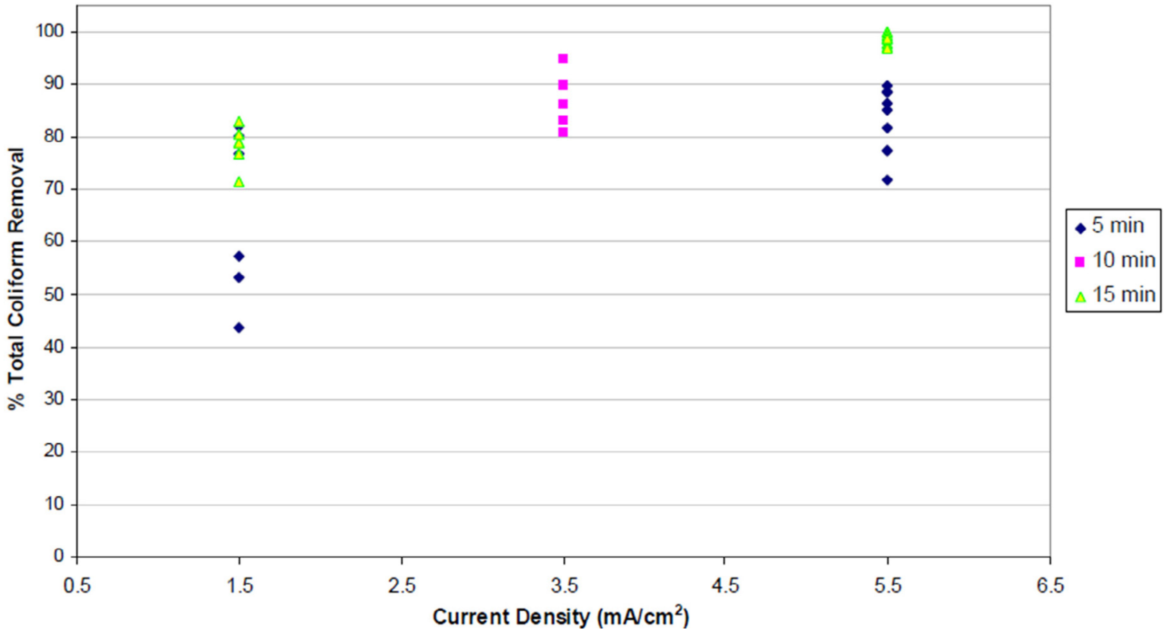

Fig. 3. Coliform removal efficiency vs. current density at different contact times

days, the first two test conditions were repeated. Moreover, the center point conditions were conducted two times.

According to the findings, using the aluminum reactor, led to a total coliform elimination efficiency of 97.7% or above at 15 min of contact time combined with the proper current input (Figure 3). According to expectations and as reported by Hashim et al. (2020), the bacteria elimination efficiency in general rose as the contact time and current density increased, and the

Table 5. Average test data

Run	Current density (mA/cm^2)	Contact time (min)	Chlorides (mg/l)	Coliform log removal	COD removal (%)	Total residual chlorine (mg/l)
1	5.5	5	137	0.60	-	0.36
2	5.5	15	135	2.20	64.1	0.91
3	3.5	10	580	0.95	64.0	0.80
4	1.5	5	142	0.33	64.7	0.33
5	1.5	15	130	0.64	75.8	0.51
6	5.5	5	866	0.91	61.1	0.42
7	1.5	5	1001	0.68	53.9	0.35
8	1.5	15	1027	0.71	66.5	0.16
9	5.5	15	1076	1.75	69.5	1.07
10	5.5	5	124	0.84	45.9	0.39
11	3.5	10	581	0.86	72.5	0.34
12	5.5	15	164	1.78	74.6	0.62

effect of the above parameters was much greater than that of salinity.

Table 5 presents the calculated log removal efficiency values, as well as COD removal efficiency values and total residual chlorine determined once the disinfection phase terminated. The average results of the three tests conducted based on each set of parameters are provided in Table 5. The impact of the three parameters (current density, contact time, and salinity) on the system was assessed based on the log removal values using Design-Ease® 6.0 software from Stat-Ease®.

To identify which factors or factors combinations had a more statistically significant effect on the disinfection efficiency of the system, a half-normal plot was drawn, as shown in Figure 4. In this figure, y axis represents normal cumulative likelihood of achieving a value equal to or less than any point of interest, while x axis gives, on an absolute value scale, impact of the interaction of variables. By obtaining the averages of the achieved responses (log removal efficiency values) at the corresponding maximum and minimum levels and calculating the difference between them, these effects are determined. To calculate an effect, is the following mathematical expression is used.

$$\text{Effect} = (\Sigma Y_{\text{high level}}/n) - (\Sigma Y_{\text{low level}}/ n) \tag{1}$$

In the above, Y and n are the response and the number of responses at the level of interest.

According to Figure 4, three points experience a drop considerably more distant than the rest; this shows that these three variables (or their combinations) have a significant impact on the removal efficiency of coliform. Impacts falling on the line indicate a normal scattering; thus, they are considered to change only because of normal causes, making them insignificant. Figure 4 indicates the negligible impact of chloride content on disinfection efficiency, which necessitates a focus on the contact time and current density parameters.

It was attempted to validate significance of the three larger factors (A, B, and AB in Figure 4) by incorporating them in a prediction formula for log removal effectiveness and performing an analysis of variance (ANOVA). Table 6 gives ANOVA results of considered factorial model. Considering that the obtained mean square for the empirical error is only 1.10, analysis data of the proposed empirical factorial model is considered reliable.

The obtained F results indicate the significant statistical effect of current density and contact time on disinfection efficiency; however, the effect of chloride concentration can be regarded as negligible. It is seen that the actual F value is 30.92, which is higher than the critical value

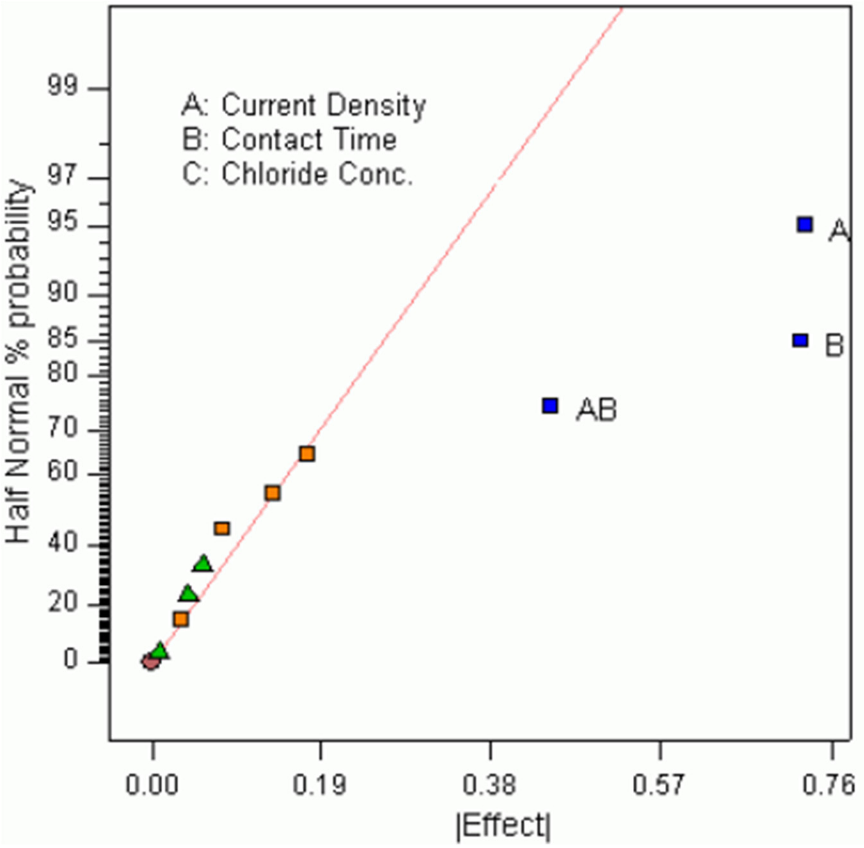

Fig. 4. Half-normal plot of impacts for log removal effectiveness

Table 6. The ANOVA of empirical model (Log Removal)

Source	Sum of squares	DF	Mean square	F value	Prob > F
Model	3.32	3	1.11	32.90	0.0001
A	1.38	1	1.38	35.88	0.0003
B	0.99	1	0.99	24.87	0.0011
AB	0.55	1	0.53	17.05	0.0058
Residual	0.24	7	0.02		
Cor total	3.58	11			

for 0.1% risk (18.77). Hence, reaching an F as large as the one obtained, only by chance, has a probability of smaller than 0.1%. This means that one or more of the selected factors in model have a significant effect on the removal efficiency of coliform with a confidence of greater than 99.9%.

The program yielded the mathematical formula below as the model based on the actual factors.

$$\text{Log Removal} = 0.485 - 0.046667{*}CD - 0.0185{*}CT + 0.023667{*}CD{*}CT \quad (2)$$

In the above, CD and CT are current density (mA/cm^2) and contact time (min), respectively.

A comparison is made between the log removal efficiency obtained from Eq. (2) and the experimental log removal efficiency in Figure 5. Given the relatively suitable obtained correlation ($R^2 = 0.91$), the model is reliable for the empirical range of conditions considered

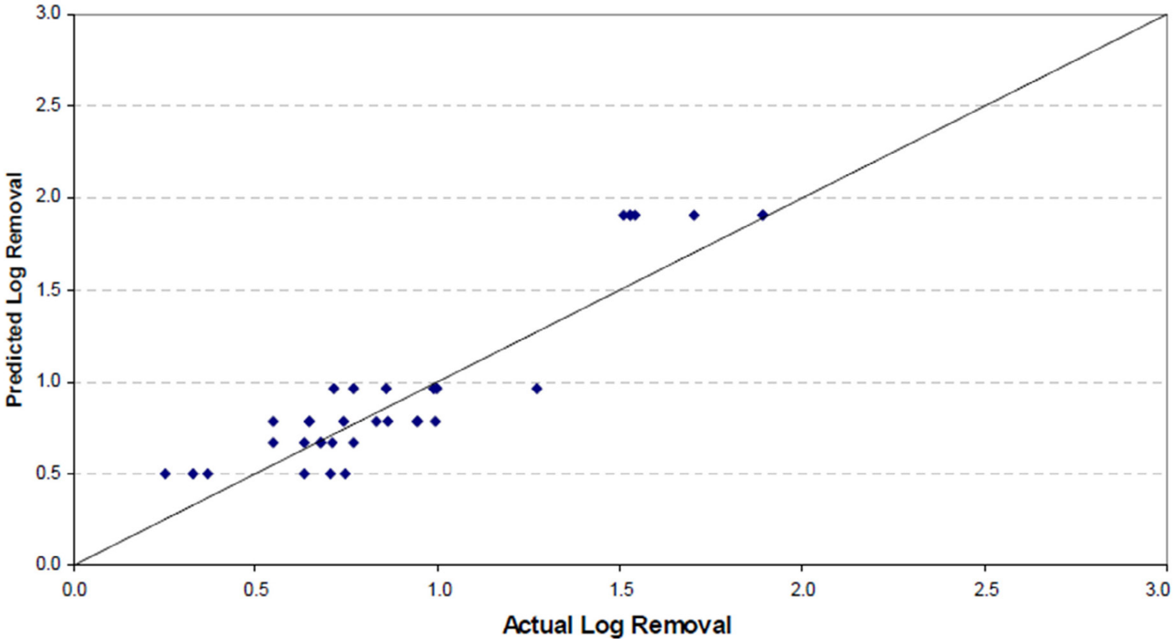

Fig. 5. Predicted vs. actual log removal efficiency values

here.

In Figure 6, it is seen how contact time and current density interact and affect log removal of coliform. Two lines are seen in the figure, which are bracketed at both ends by least significant difference (LSD) bars. As can be seen, for the low contact time of 5 min, a small change occurs in log removal by changing the current density. On the other hand, the log removal sees a considerable increase at the high contact time of 15 min, which indicates a significant positive impact as a result of the supplemented current density. Furthermore, at the low current of 1.5 mA/cm^2, overlapping of LSD bars occurs at that end of the interaction plot, suggesting an insignificant discrepancy in log removal at the low current density.

Figure 7 also shows how these two factors interact by depicting a system contour. This 2D illustration of log removal efficiency in terms of contact time and current density shows factor values beyond the experimental ranges. Nevertheless, it can serve as an applicable means to predict optimum combinations of contact time and current density. As an example, by using a current density equal to 5 mA/cm^2 as well as a residence time equal to 25 min, a significant bacterium killing efficiency of around 3 log removal is achieved. These are proper conditions for implementation in a wastewater treatment plant.

The total residual chlorine was determined shortly following different test runs; this was done to ensure that during the electrodisinfection, the oxidization of chloride ions to produce chlorine gas and hypochlorite ions at the anode occurred (see Table 5 listing average values). The time interval from the start of work on a sample and chlorine analysis was not exactly the same for the tests. Nevertheless, as shown in Table 5 and Figure 8, the effluent samples contained total residual chlorine. By increasing the operational current density, the chlorine/hypochlorite formation experienced a general improvement. This empirical finding supports the assumption that chlorine gas, hypochlorous acid, and hypochlorite are some of the lethal species (Nidheesh et al., 2021).

By analyzing effluent samples pre- and post-treatment (following sludge sedimentation/flotation) for total COD, the electro-disinfection by Al electrodes showed the ability to remove up to 76% of COD (see Table 5 listing average values). Indeed, this result was expected as a number of studies report that electrocoagulation is superior to traditional coagulation in terms

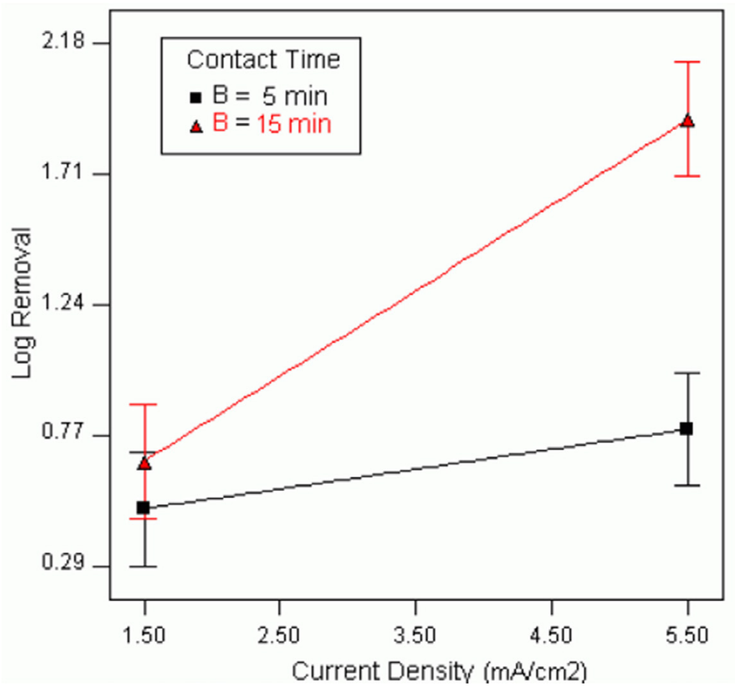

Fig. 6. Log removal vs. current density in terms of contact time

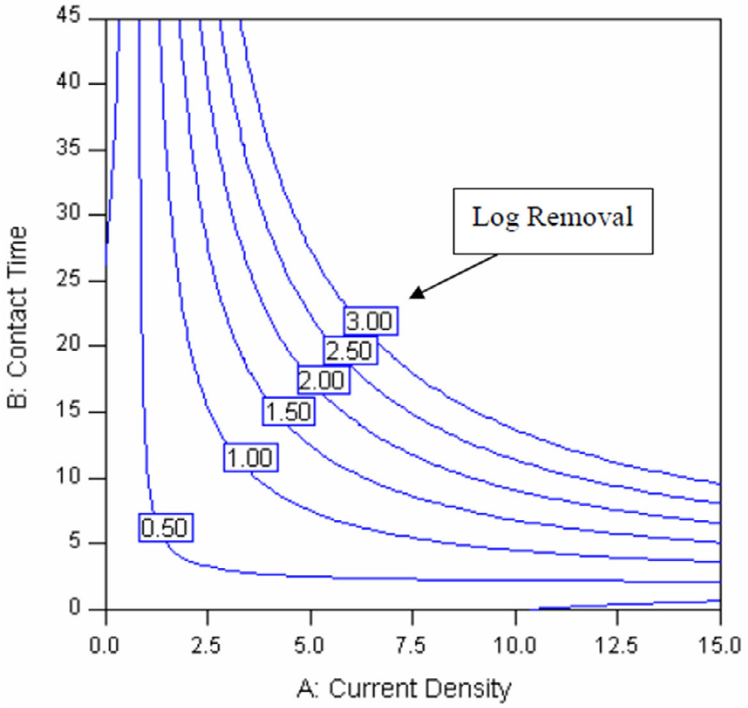

Fig. 7. Current density vs. contact time in terms of log removal efficiency

of the removal of COD and suspended solids (Li et al., 2020). The main reason for this is the indirect oxidative impact of species that are formed in the reaction and coagulation/absorption by aluminum hydroxide floc (Kourdali et al., 2018).

In the tests, the current of 2 to 10 amps associated with a current density ranging from 1.5 to 5.5 mA/cm^2 was used, and voltage demand was around 2 to 14 volts. Eq. (3) was used to calculate the rate of energy consumption (kWh/m^3) per different cases.

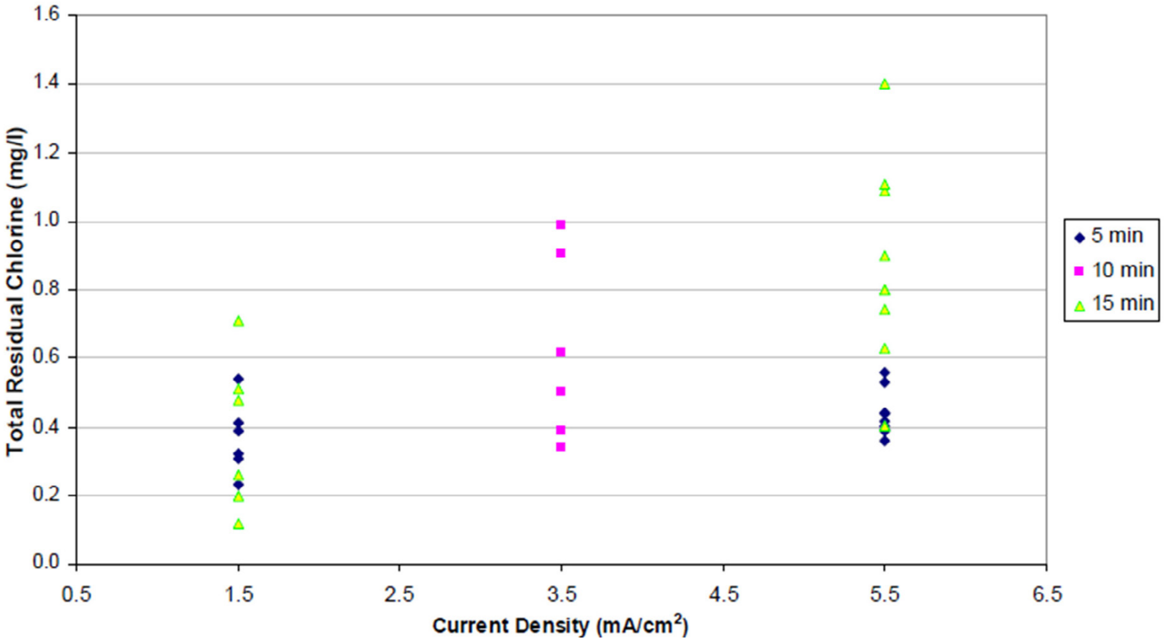

Fig. 8. Total residual chlorine vs. current density in term of contact time

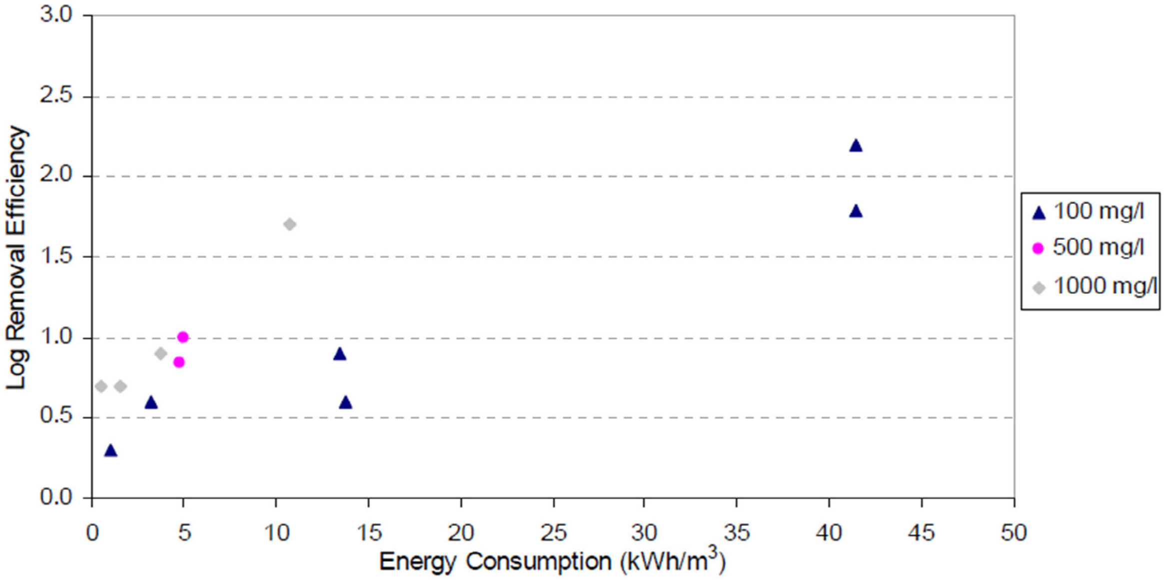

Fig. 9. Log removal efficiency vs. energy consumption for different chloride concentrations in the wastewater effluent

$$P = (I \cdot V \cdot t)/Vol \tag{3}$$

In the above, I, V, t, Vol is current (amps), applied voltage (volts), contact time (h), and sample volume (liters), respectively.

Figure 9 demonstrates the variation of log removal efficiency with energy consumption at different chloride concentrations in the wastewater influent. As can be seen, in the two cases with the highest disinfection efficiency values, the energy demands were very high. However, adding salts (e.g. sodium chloride) lowers the consumption of energy, which enables an improvement of the system from the energy consumption perspective.

Table 7. Total aluminum and TSS following electric disinfection

Run	Current density (mA/cm^2)	Contact time (min)	Total Al (mg/l) prior to settlement	Total Al (mg/l) supernatant	TSS (mg/l) prior to settlement	TSS (mg/l) supernatant
1	5.5	5	687	-	1976	<4
2	5.5	15	1154	-	6785	<4
3	3.5	10	1032	1.3	2720	<4
4	1.5	5	176	4.8	366	<4
5	1.5	15	411	6.2	1121	<4
6	5.5	5	803	15.9	2922	<4
7	1.5	5	197	4.1	481	<4
8	1.5	15	553	9.4	1422	<4
9	5.5	15	3179	8.5	5915	<4
10	5.5	5	867	20	1956	<4
11	3.5	10	808	>8.0	2538	<4
12	5.5	15	1548	17.0	5756	<4

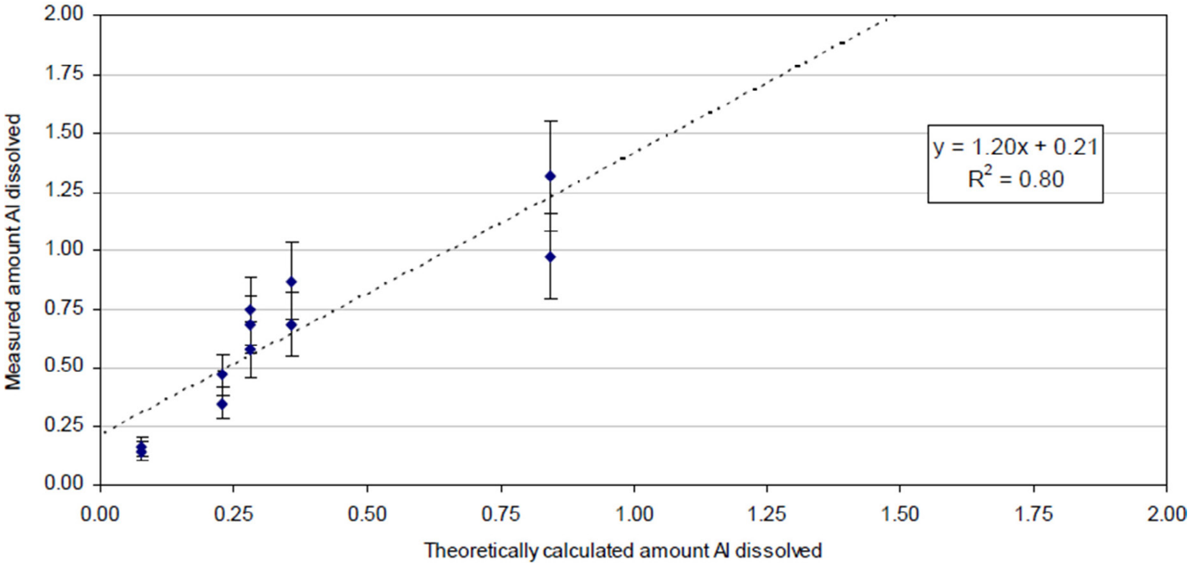

Fig. 10. Relation between theoretical and empirical dissolved aluminum values

The relation of current density (A/cm^2) and the aluminum content incorporated in the solution is described using Faraday's law (Domga et al., 2017). Here, prior and following complete sedimentation, the total aluminum contents of the effluents were measured. The average total aluminum and TSS values, before and after settlement, are provided in Table 7. Figure 10 compares the theoretical dissolved aluminum values using Faraday's Law with the empirical values. To represent the magnitude of empirical errors in different cases, error bars are presented in the plot.

The Al sludge formed in the process significantly increases TSS and total dissolved aluminum (prior to clarification). Per optimum conditions, sedimentation/flotation easily removes this sludge and in turn leaves a supernatant fit for discharge from any wastewater treatment plant.

Figure 10 shows that Al dissolved in the wastewater is about 20% higher than that estimated by Faraday's law. The abnormal Faradaic results of dissolved species of Al can be have multiple explanations, which include atypical pitting corrosion of Al (Trompette et al., 2021) and rapid dissolution of the oxide film formed on the electrode surface (Zini et al., 2020). Thines et al. (2017) demonstrated that in the three-electron oxidation process creating Al(III) species, apparent current efficiency values for aluminum cathode dissolution and aluminum anode dissolution exceed one.

Based on earlier findings, the material of the electrode has a significant effect on the formation of reactive species including OCl^-, Cl_2, ozone, H_2O_2, $\cdot OH$, and $\cdot HO_2$; this in turn affects the disinfection efficiency (Thostenson et al., 2018; Zhang et al., 2023). In all the conducted tests, total chlorine residual in the effluents were obtained. Note that in case the current is on, local concentrations of active disinfected species can become one or two orders of magnitude greater than average concentrations in the fluid coming from the cell (Devlin et al., 2019). Nevertheless, ensuring that chlorine serves as the main cause of bacterial death is not feasible. In terms of technology, this capacity for residual disinfection is interesting because by using this technique for a continuous flow of water, it can be ensured that only a part of the water flow needs to contact the electrodes and consequently be mixed with the rest of the water flow volume. Nevertheless, to discover whether halogenated hydrocarbons are formed, more research is needed.

Other oxidant species can be formed by electrochemical reaction (Shahedi et al., 2020). Due to the instability and short-term life of atomic oxygen, hydroxyl radicals, hydrogen peroxide, and perhydroxyl radicals, monitoring them was not possible (Hashim et al., 2020). However, when the current was active, all of them might be present in the water. These strong oxidation agents not only eliminate the bacteria but also are able to degrade organic pollutants; thus, the process presents a promising strategy to lower disinfection by-products (Mazhar et al., 2020).

Without any regard to the electrode material of interest, the occurrence of electrochemical reactions is usually somewhat not selective and secondary reactions and heat generation consume the majority of the energy of the power source (Kourdali et al., 2018). An electrochemical reaction can have either mass transport- or activation-controlled rates (Ganiyu et al., 2021). Hence, enhancing the rate of these reactions requires (1) the provision of electrodes with high surface areas and (2) the promotion of the conditions of turbulence in the electrolyte via agitation, electrode movement, or turbulence promoters in the cell (Trompette et al., 2021; Zini et al., 2020).

According to the results, the COD and TSS of the wastewater generally declined in all the tests as a result of an electric field that neutralized the surface charges of colloid particles in wastewater and in turn led to the accumulation and sedimentation of these particles (Yang et al., 2019). TSS removal is advantageous for the process due to its potential improving effect on the disinfection of microorganisms considering that high turbidity hampers disinfection (Léziart et al., 2019).

Since this is an early work on the electro-disinfection of very small wastewater effluent volumes, it cannot provide conclusive implications for wastewater treatment. However, the findings are strongly positive and indicate the necessity for further examination of this method.

CONCLUSION

The electrochemical process shows high effectiveness for wastewater effluent disinfection following secondary treatment, particularly in case of the employment of aluminum electrodes. At a contact time and a current density of 15 min and 5.5 mA/cm^2, respectively, the efficiency of bacterial killing reached 97.7%. As the contact time and current density increased, the bacterial killing efficiency experienced a general rise, and these factors had an effect much larger than salinity did. After electro-disinfection, the TSS and COD were found to decrease by 78.50% and 99.93%, respectively; this occurs due to the introduction of a lot of electrons into the water by the electric current, which generates a powerful reducing environment. Based on the results of the conducted preliminary work on electro-disinfection, it is found that this technique can serve as viable replacement for the chlorination of wastewater effluents. In addition to being efficient, this process is more eco-friendly than traditional disinfection methods. However, this method has many notable aspects that require more research before it can be applied in industrial-scale wastewater treatment plants. It is necessary to improve this method in terms of technology and cost-effectiveness.

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The present research did not receive any financial support.

CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/ or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

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