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The Optimization of Operational Variables of Electrochemical Water Disinfection Using Response Surface Methodology

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Abstract: The electrochemical treatment of canal water was investigated in a batch-wise system in the presence of stainless steel 316-grade electrodes. Three effective process parameters, including current density, reaction time, and electrode spacing, were evaluated in the range of 0.25–2.5 mA/cm^2, 1–10 min, and 0.5–2.5 cm, respectively. Operational variables of electrochemical disinfection are optimized in response surface methodology (RSM) using Box–Behnken design. Before electrochemical disinfection, a pretreatment process of coagulants mixing for turbidity removal was conducted. Results revealed that a 10 ppm dosage of Ferric chloride ($FeCl_3.6H_2O$) and alum ($Al_2(SO_4)_3{\cdot}16H_2O$) at neutral pH is appropriate. Furthermore, the RSM analysis shows that interelectrode spacing is the most prominent factor affecting the disinfection performance, and increasing electrode spacing inversely affects the disinfection efficiency. Results revealed that 1.52 mA/cm^2 current density, 6.35 min reaction time, and 1.13 cm of electrode spacing are the optimum conditions, resulting in a statistically 98.08% disinfection of the total coliform. The energy required for electrochemically disinfection of water at optimum conditions was 0.256 kWh/m^3.

Keywords: current density; electrochemical disinfection; response surface methodology; treatment time

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1. Introduction

The supply of an adequate amount of pure water is essential for human existence. Urbanization has placed incredible stress on natural water resources [1]. The fastest growing population and economic revolutions also have a significant impact on the sustainability of water [2]. Water is a scarce natural resource, and around 40% of the global population has been affected by this scarcity [3]. Global water consumption is about 4600 billion cubic meters per year, which will increase by 1% annually [4]. Water sources such as lakes, rivers, and groundwater are running dry due to vast utilization and climate change. Natural water's quality is mostly linked with climate change, the state of the soil, and human activities [5].

As a universal solvent, water is contaminated with unwanted components that change water quality and cause water pollution [6]. Undeniably, the available resources are continuously being polluted with a wide range of chemical, physical, and biological pollutants from industrial wastewater, municipal effluent discharge, the agricultural sector, and leachate leakage from landfills and other natural sources [7]. Among these pollutants, pathogenic and nonpathogenic microorganisms are considered at high risk as compared to other physical or chemical pollutants because of the high number of illnesses and deaths that they could cause [8]. For example, waterborne diseases related to pathogenic microorganisms, such as diarrhea and gastrointestinal upset, are responsible for an estimated 2 million deaths every year. The World Health Organization (WHO) finds that about 80% of diseases are waterborne, and the death rate is 3.1% due to unhygienic conditions of drinking water in various countries [9].

Therefore, it is necessary to eliminate the contaminants before human consumption [10]. Over the period, numerous disinfection techniques have been developed, including (1) chemical systems based on chlorine and ozone, (2) photocatalysis and photodynamic disinfection, (3) physical methods such as ultraviolet irradiation, and (4) electrochemical disinfection [11]. Compared to alternatives, ozonation and ultraviolet (UV) radiation have gained acceptance in commercial water treatment systems. UV is an efficient and widely utilized disinfection technology; yet, it is not without technological restrictions due to the transmission limits of ultraviolet radiations within the water sample [12]. The chemical treatment processes produce strong oxidants that may react with natural organic matter and produce toxic disinfection by-products. Due to their carcinogenic characteristics, these by-products have public health concerns [13]. Recently, efficient electrochemical disinfection methods for water treatment are developed [14]. The electrochemical techniques are known to inactivate various microorganisms, including bacteria, viruses, and algae. In addition, the technology does not significantly impact the environment and is inexpensive and simple to run [15]. Researchers are trying the address the challenges that electrochemical disinfection is facing for commercialization. For instance, during electrochemical disinfection, fouling at the electrode surface and scavenging hydroxyl ions by inorganic ions or natural organic matter can occur [16]. At low potentials, the fouling of electrodes has been observed as a result of the polymerization of phenolic compounds present during the treatment time. The fouling at the electrode surface may result in reduced reaction rates of the substrate. A periodic backwash treatment can overcome the fouling of electrodes, and the deposits could be flushed to areas with higher potential-containing hydroxyl ions to remove the foulants [17]. The microbial inactivation through electrochemical disinfection methods largely depends on the electrolytic cell configuration, electrode material, and other operational parameters such as electrode spacing, treatment time, current density, and flow rate [18].

Different optimization methods are employed to determine the optimum conditions for water disinfection. Response surface methodology (RSM) has been used to optimize and model various operational variables for the treatment of wastewater. RSM is a collection of statistical and mathematical techniques used to build models for evaluating several variables and producing desirable values for response [19]. The methodology consists of stages, including selecting independent variables, experimental design, model selection, model adequacy checking, graphical representation of the model, and parametric optimization [20].

The choice of suitable electrode materials is a critical factor in the electrolysis process because it affects not only the process efficiency but also the selectivity of disinfection by-products [21]. The anode materials studied for electrochemical disinfection include boron-doped diamond (BDD), dimensionally stable anode, and platinum. In a study, platinized titanium mesh electrodes are used for the deactivation of E-coli from tap water. Results highlight that at a maximum current density of 13.5 mA/cm^2, 4 log of E-coli are disinfected after a reaction time of 10 min. Similarly, in another study, ordinary steel and aluminum electrodes are employed [22]. Riyanto et al. conducted an experimental study in which water is artificially contaminated with a known microbial load of 190 MPN/100 mL and carbon electrodes are used for disinfection. Results show that after 40 min of reaction time, the percentile decrease in the coliform was 88% at the current density of 10 mA/cm^2 [23].

Electrode materials surveyed in the literature are expensive and are not easily available. Moreover, there is a lack of research on stainless steel electrodes of specific grades for electrochemical disinfection. Stainless steel of grade 316 offers promising advantages such as being cost-effective and corrosion-resistive even at high temperatures [24]. In this study, the electrochemical disinfection of canal water was carried out in the presence of stainless steel plates of 316-grade. Moreover, the operational parameters are optimized in design expert software by using response surface methodology. The experimental runs were designed following Box–Behnken design (BBD) and performed in a batch-type electrochemical reactor. Three factors, namely, current density, electrode spacing, and treatment time, were selected as independent process variables, while disinfection efficiency was chosen as the response variable.

2. Materials and Methodology

2.1. Sample Collection and Experimental Setup

The water sample was collected from BRB Canal near Jallo Mor Lahore (31°36′02.8″ N 74°29′44.4″ E). The main components of the experimental setup include a flocculation tank, electrochemical chamber, and solar-powered DC power supply. The batch-type electrochemical reactor with a volume of 4000 mL that consists of two plates of stainless steel 316-grade as electrodes was used in this study. Each plate has an exposed height and width of 214 mm and 145 mm, respectively. The process flow diagram of the treatment unit is shown in Figure 1. Raw canal water is pumped to the flocculation tank for settling of the silt and dirt. Coagulants are used to accelerate the settling process. The most commonly used metal coagulants are of two types: aluminum-based and iron-based. The aluminum-based coagulants include sodium aluminate ($AlNaO_2$), aluminum chloride ($AlCl_3$), and aluminum sulfate ($Al_2(SO_4)_3$). The iron-based coagulants include ferrous sulfate ($FeSO_4$), ferric chloride ($FeCl_3$), and ferric sulfate ($Fe_2(SO_4)_3$). In this study, aluminum sulphate and ferric chloride were used as coagulants. The flocculation tank functions both as a mixer as well a settler and therefore requires batch mode operation. After the flocculation process, clear water from the tank is then pumped to an intermediate storage tank to provide water for the downstream treatment process even when the flocculation tank is in the mixing or setting phase. Water is then pumped to a micron-sized prefilter (polypropylene filter or commonly known as PPF) to remove the unsettled/suspended particles having a particle size greater than 3 microns. The prefiltered water is then subjected to the electrochemical reactor. The redox reactions occurring at the electrode surface generate reactive species (ROS or RCS), which disinfect the microorganisms. Besides disinfection, the redox process also removes various suspended and dissolved solids through the electrocoagulation phenomenon, which are afterward filtered out in the postfilter. In the postfiltration process, water is passed through a 0.3-micron size filter membrane to remove particles and a bed of activated carbon to remove organic contaminants causing color, odor, and taste. A solar system has been installed to power the pumps and electrochemical reactor.

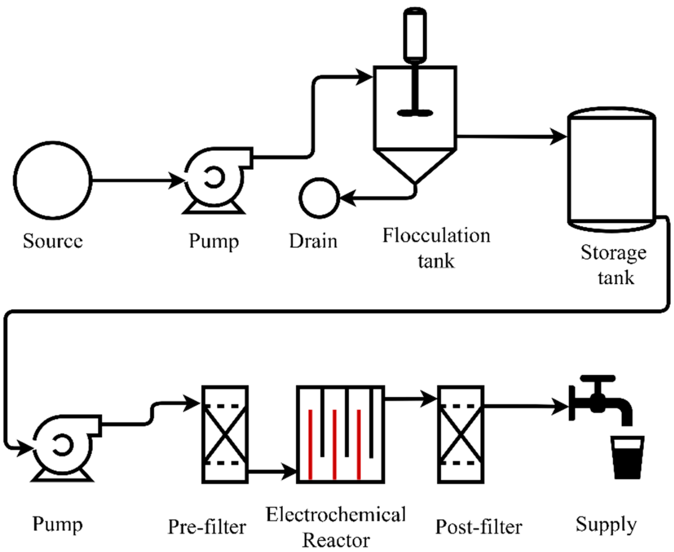

Figure 1. Process flow diagram of the proposed water treatment unit.

2.2. Response Surface Methodology

Experiments must be designed with reliable and suitable measurements of the responses under consideration. Among several factors affecting the system performance, current density, operating time, and electrode spacing were chosen as model parameters. The experimental runs were planed according to Box–Behnken design (BBD) with lower and upper bounds of three process parameters. BBD was selected because it is more efficient and involves fewer experiments compared to three-level full factorial design and center composite design (CCD) [25].

The process parameters, including current density, operating time, and electrode spacing, were evaluated in the range of 0.25–2.5 mA/cm^2, 1–10 min, and 0.5–2.5 cm, respectively. The factor labels and corresponding levels are given in Table 1. The complete model was developed, simulated, and analyzed in the Design-Expert statistical software. The BBD model suggested 17 experimental runs with 5 replicates, as provided in Table 2.

Table 1. Design variables with lower and high-level in Box–Behnken design (BBD).

		Coded Factors, *x*			
		−1	**0**	**1**	
Variables	**Units**	**Minimum**	**Mean**	**Maximum**	**Std. Dev.**
Current density (*i*)	mA/cm^2	0.25	1.38	2.50	0.77
Treatment time (*t*)	min	1.00	5.50	10.0	3.09
Interelectrode spacing (*d*)	cm	0.50	1.50	2.50	0.69

Table 2. Experimental design for electrochemical disinfection efficiency with independent variables and experimental and predicted response values.

Run	**Independent Variables**			**Response Disinfection Efficiency (%)**		
	***i* (mA/cm^2)**	***t* (min)**	***d* (cm)**	**Experimental**	**Predicted**	**Residual**
1	1.375	10	0.5	84.0	79.88	4.13
2	1.375	1	0.5	75.0	75.63	−0.63
3	2.5	5.5	0.5	68.0	68.38	−0.38
4	0.25	5.5	0.5	79.0	82.13	−3.13
5	1.375	5.5	1.5	93.0	93.50	−0.5
6	1.375	5.5	1.5	92.0	93.50	−1.50
7	1.375	5.5	1.5	95.0	93.50	1.5
8	1.375	5.5	1.5	93.5	93.50	0.00
9	1.375	5.5	1.5	94.0	93.50	0.50
10	2.5	10	1.5	79.0	82.75	−3.75
11	0.25	1	1.5	76.0	72.25	3.75
12	0.25	10	1.5	58.0	59.00	−1.0
13	2.5	1	1.5	69.0	68.00	1
14	1.375	1	2.5	28.0	32.13	−4.13
15	0.25	5.5	2.5	12.0	11.63	0.38
16	2.5	5.5	2.5	48.0	44.88	3.13
17	1.375	10	2.5	30.0	29.38	0.63

The response was correlated with process variables using a second-order model. The model is customarily stated in Equation (1):

$$Y = f(x) = \beta_0 + \sum_{i=1}^{n} \beta_i \, x_i + \sum_{i=1}^{n} \beta_{ii} {x_i}^2 \sum_{j} \sum_{i=2}^{n} \beta_{ij} x_i x_j \quad (1)$$

where Y is the expected response, β_0 is the intercept or the constant regression coefficient, x_i is the independent process factor, β_i and β_{ii} represent the linear and quadratic regression coefficients, respectively, and β_{ij} is the interaction coefficient in relation to the factors x_i and x_j where k is the number of independent variables (k = 3 in the current study). The analysis of variant (ANOVA) technique was used to analyze the second-order model's fitness statistically.

2.3. Electrochemical Disinfection

After the design of experiments in RSM, the experiments are carried out in batch type electrochemical reactor according to the design model. The experimental unit consist of two main parts. A batch type electrochemical chamber and second component is the DC power supply with a current intensity of 0 to 5 A and an adjustable electrical voltage of 0 to 30 V. Power is supplied from photovoltaic solar system installed on the roof of chemical engineering department. After each experimental run, the disinfected samples are collected in sterilized glass vials.

2.4. Microbial Analysis

The electrochemically disinfected samples are tested through microbial analysis. The plate count method is the most commonly used method for bacterial count [26]. In this study, the bacterial determination was carried out through the plate count method, in which Lysogeny Broth (LB) agar medium was used for bacterial growth. LB agar media was prepared from yeast extract, NaCl, Tryptone, and agar of known concentration. Initially, the equipment and prepared LB agar media were sterilized in a medical autoclave unit at 121 °C for 25 min. Afterward, the diluted samples were poured into the petri dishes that contain a known volume of cultured media. At last, these petri dishes were incubated in a microbiological incubator at a temperature of 37 °C for 24 h. The bacteria colonies were counted following the incubation period. Finally, the colony forming unit (CFU) per ml was calculated using Equation (2):

$$CFU = \left(\frac{no\ of\ colonies * dilution\ factor}{Volume\ of\ the\ cultured\ plate} \right) \quad (2)$$

3. Results and Discussion

3.1. Pretreatment Experiment

Canal water contains highly suspended particles (typically above 100 NTU). The turbidity should be less than 5 NTU for effective downstream treatment to avoid fouling filter membranes and deposition on the electrode plates. Silt, characterized as particles that have a size above 10 µm, can settle down (assuming a particle density of 2650 kg/m^3) in 33 min. However, smaller particles suffer repulsive forces and remain suspended for longer. Mixing appropriate coagulants can reduce the repulsive forces and assist floc formation, which can settle rapidly [27]. Ferric chloride ($FeCl_3.6H_2O$) and alum ($Al_2(SO_4)_3 \cdot 16H_2O$) are commonly used in water treatment to minimize turbidity in drinking water [28–31].

The first set of experiments aimed to determine the appropriate coagulant dosage, pH, and conditions of mixing and settling to obtain residual turbidity of less than 5 NTU. The pH of the water sample was adjusted by using NaOH and H_2SO_4 [32,33]. The optimum coagulant dosage and pH value were tested in the jar experiment. At the start of the experiment, rapid mixing (350 rpm for 1 min) was performed to enhance the coagulation, followed by gradual mixing (30 rpm for 20 min) for flocculation. After the mixing profile was completed, the sample was poured into the Imhoff sedimentation cone for settling. The flocs that formed were allowed for 45 min to settle down. The volume of sediments at the bottom and turbidity of the clear water at the top was recorded as a time function. For turbidity measurements, supernatant samples were obtained 20 mm below the surface of the water. Residual turbidity was used as the performance parameter. The reactor was filled with 3500mL of raw water in each experiment, and experiments were performed according to the design model through RSM.

Turbidity in water is induced by suspended particles and measured in terms of nephelometric turbidity units (NTU). The fine sand particles, having a particle size of more than 100 µm, easily settle down in less than 1 min. However, small particles characterized as silt require extended settling time or the addition of a coagulant to accelerate the settling rate. The turbidity of the raw water sample collected from the canal was 92.8 NTU. After unassisted settling of around 15 min, the turbidity reduced to less than 50 NTU.

The impact of coagulant dosage and pH on the residual turbidity of the raw water sample, with initial turbidity of 48 NTU, is shown in Figures 2 and 3 for aluminum sulfate and ferric chloride, respectively. As observed, a 10 ppm dosage of both coagulants is suitable to reduce the turbidity level to GDWQ standards. For instance, the residual turbidity of 3.5 and 1.5 was obtained for aluminum sulfate at pH = 7 and ferric chloride at pH = 6, respectively. Ferric chloride performs better in turbidity removal than aluminum sulfate, but both meet the quality standards desired for post-treatment. The pH affects the coagulation/flocculation process, which is well documented in the literature. High coagulant dosing (above 30 ppm) produces countereffects that might be due to the charge reversal and instability of the colloidal. These results are in good agreement with the literature [32].

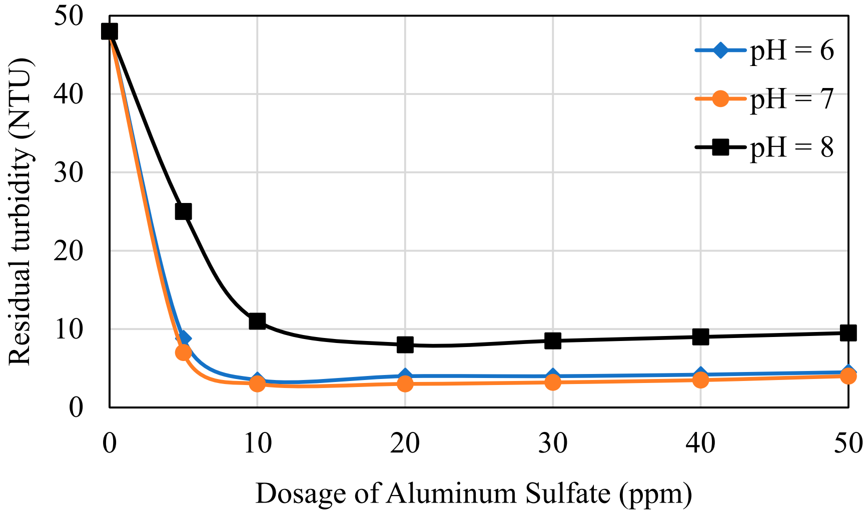

Figure 2. Residual turbidity as a function of aluminum dose at three pH levels.

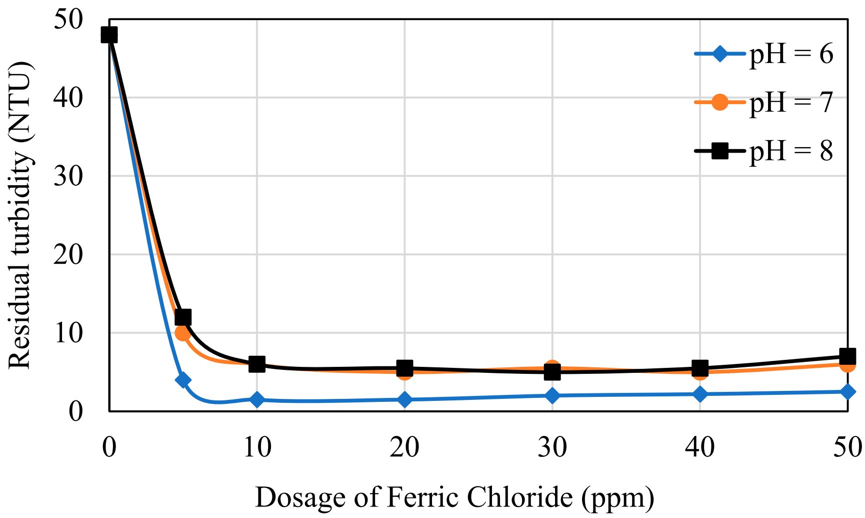

Figure 3. Residual turbidity as a function of ferric chloride dose at three pH levels.

3.2. Optimization of Designed Parameters through RSM

The experimental domain of current density, treatment time, and interelectrode spacing was previously determined during the preliminary experiments [33]. To further investigate the effect of these parameters on the treatment performance and determine optimum conditions, Box–Behnken design (BBD) methodology was followed, suggesting 17 experiments with five replicas, as given in Table 2. The experiments were performed under these designed conditions. Table 2 lists measured and predicted responses of the electrochemical disinfection at each set of independent variables. As observed, the disinfection efficiency ranged from 12% to 95%, corresponding to runs # 15 and 7, respectively. The highest efficiency does not lie at terminal values, suggesting that the variable domain chosen for the experiments is appropriate. Notice that when using a current density of as low as 0.25 mA/cm^2, it is still possible to eliminate 79% of the coliforms. Moreover, it can also be observed that the predicted response from the model is nearly close to the experimental response, which represents a good relationship between predicted and actual response.

These results imply the possibility of disinfection without the need to generate a large amount of electricity, implying lower operating costs and higher reaction time. However, the interelectrode spacing dominates the response as large spacing causes high ohmic resistance and expectedly lowers the generation of disinfectant concentration, evident from the low disinfection efficiency at 2.5 cm spacing (run # 14–17). Therefore, high disinfection efficiency would make it possible to narrow electrode spacing and low current density value [34].

In order to find out the main and double-interaction effects of independent variables, an analysis of variance (ANOVA) was carried out. Table 3 presents the regression coefficients, F-ratio, and *p*-value of the system performance obtained through ANOVA. The *p*-values were used to identify the independent variables that have a significant statistical influence on the disinfection. The value of $p < 0.05$ indicates that the variable is statistically significant with a confidence level of 95%. As presented in Table 3, the current density and interelectrode spacing have *p*-values < 0.05, suggesting that they directly affect the disinfection efficiency. The significance of all three variables is in the order of C: interelectrode spacing > A: current density > B: treatment time. The double interaction parameters also fulfill the statistical constraints, implying that their corresponding coefficients have significant weight in the polynomial equation.

Table 3. Regression coefficients of the proposed model.

Factor	Coefficient Estimate	Standard Error	F-Ratio	*p*-Value
Intercept (β_0)	93.5	1.60	90.48	<0.0001
A: Current density (β_1)	4.87	1.27	14.83	0.0063
B: Treatment time (β_3)	0.38	1.27	0.088	0.7757
C: Interelectrode spacing (β_2)	−23.5	1.27	344.58	<0.0001
AB (β_{12})	7	1.79	15.29	0.0058
AC (β_{13})	11.75	1.79	43.07	0.0003
BC (β_{23})	−1.75	1.79	0.96	0.3609
A^2 (β_{11})	−12.75	1.75	53.39	0.0002
B^2 (β_{22})	−10.25	1.75	34.50	0.0006
C^2 (β_{33})	−29	1.75	276.18	<0.0001
R^2	0.991	–	–	–
Adjusted R^2	0.981	–	–	–

The appropriateness of the developed model was assessed based on the determination coefficients (R^2 and adjusted R^2). The results achieved by using the quadratic model (Equation (3)) were better compared to other fits, with R^2 = 0.991 and adjusted R^2 = 0.981 (Table 3). Bashir et al. [35] remarked that high values of R^2 suggest great accordance between the estimated data of the model and experimental data. Since both of these coefficients are very close to unity, it indicates that the model developed is suitable for describing electrochemical disinfection. The appropriateness of the model is evident from a good agreement among the experimental and predicted values, as shown in Figure 4.

$$\begin{aligned}\eta_{TC}(\%) = 51.52 + 8.77\,A + 4.33\,B + 51.28\,C + 1.38\,AB \\ + 10.44\,AC - 0.39\,BC - 10.07\,A^2 - 0.51\,B^2 \\ - 29.00\,C^2\end{aligned} \quad (3)$$

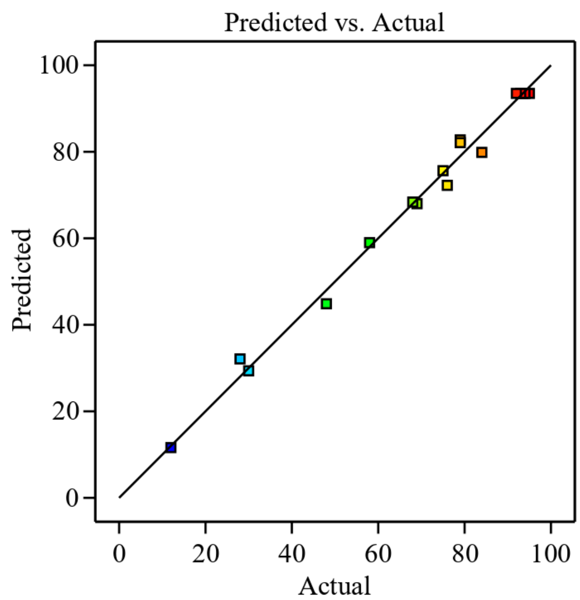

Figure 4. Adequacy of model for electrochemical disinfection efficiency.

3.2.1. Adequacy of Mathematical Model

The diagnostic plots such as actual vs. predicted values are constructed to evaluate the adequacy of the developed mathematical model. These plots help figure out the relationship between the experimentally conducted values and the predicted response values. Figure 5 illustrates the analytical plot of predicted versus actual values for electrochemical disinfection efficiency. It can be observed that the data points lie very close to the diagonal line, which represents the good relationship between the data predicted from the developed model and the experimental data.

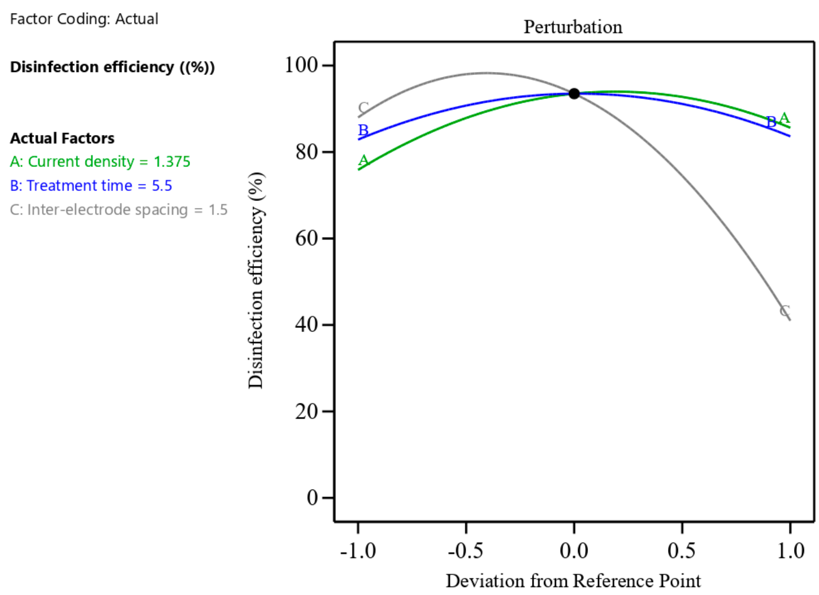

Figure 5. Partial effect of process variables on electrochemical disinfection efficiency.

3.2.2. Effect of Process Variables on the Disinfection Efficiency

The partial effect of process variables is shown in Figure 5. The partial effect plot illustrates the variation in disinfection efficiency with the various operating factors under the optimum condition. The reference point on the plot represents the mean values of process variables. It can be observed that all three variables nonlinearly affect the disinfection efficiency. An increase in current density and treatment time mildly affects the efficiency.

The observation shows that disinfection increases as the current density increases; however, an opposite trend may also be observed by a further increase in current density due to the polarization and passivation of electrodes. Oxides are formed on the electrode surface, which hinders the disinfection rate, yet this leads to an increase in potential. Furthermore, results reveal that the disinfection rate increases by increasing the reaction time. The percentage decrease in the coliform count is 93.5% by increasing the reaction time from 1 to 5.5 min.

However, a significant behavior was observed for interelectrode spacing. Increasing spacing reduces efficiency. Moreover, the energy consumption also increases as the electrode spacing increases. This is expected as an increase in the spacing also increases the ohmic resistance and decreases the generation of electrochemically generated disinfecting agents, which subsequently lowers the disinfection efficiency. These results are in agreement with [36]. GilPavas et al. [37] remarked that an increase in current density increases the disinfection of E. coli, and the effect is independent of the electrode type used. The disinfection has been related to the high generation of oxidizing species, particularly hydroxide (OH), which was accountable for coliform membrane disruption. Qi et al. [38] reported that increasing the current density from 15 to 20 mA/cm^2 eliminates the microbes and results in reducing the operating time from 30 to 20 min.

Nevertheless, it has a less significant effect than interelectrode spacing as it directly affects the medium resistance. In the literature, the medium resistance is commonly reported in terms of conductivity [39]. To improve the disinfection efficiency, medium conductivity can be increased by adding NaCl and Na_2SO_4. It implies that the generated anions and radicals improve the conductivity and disinfection efficiency.

3.2.3. Combined Effect of Process Variables on Disinfection Efficiency

The additive or nonadditive effect of the process variables can be represented by combined interaction terms of a mathematical model. The additive impact of two-factor effects indicates that the influence of one variable on the response is independent of the level of the other parameter. Figure 6 demonstrates the effect of change in current density and electrode spacing on disinfection efficiency; as can be seen, the disinfection percentage increases with a raise in current density. At the same time, a reverse trend was observed in the case of increasing electrode spacing [23,34]. The combined effect of current density and treatment time on the disinfection efficiency is presented in Figure 7. This figure illustrates that reaction time and current density positively affect disinfection efficiency. However, this ascending trend is predicted for up to 6 min of operating time and current density of up to 1.4 mA/cm^2 [34]. Figure 8 illustrates the effect of reaction time and interelectrode spacing on disinfection efficiency. In the 3D surface plot, the disinfecting efficiency increase from the blue to the red color region.

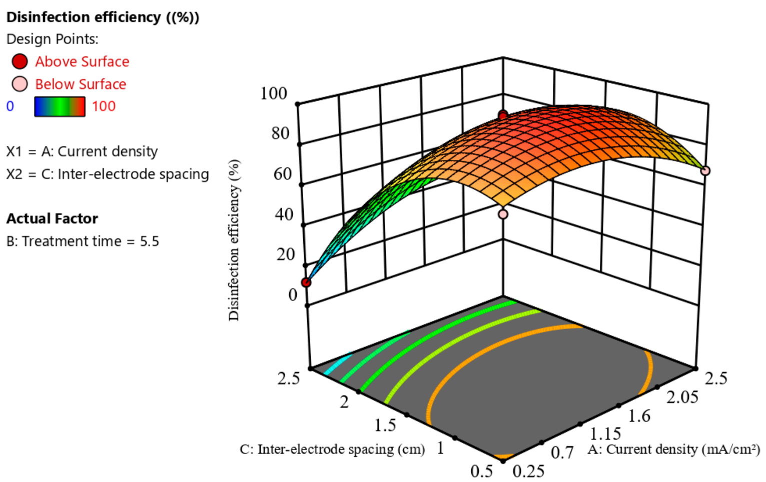

Figure 6. Three-dimensional graphical surface optimization of disinfection efficiency vs. current density and interelectrode spacing.

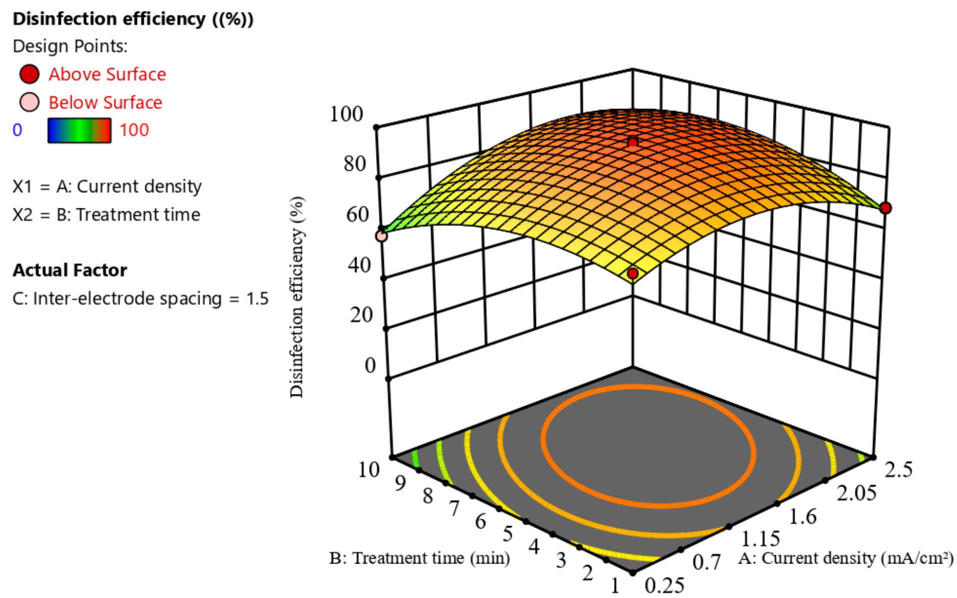

Figure 7. Three-dimensional graphical surface optimization of disinfection efficiency vs. current density and treatment time.

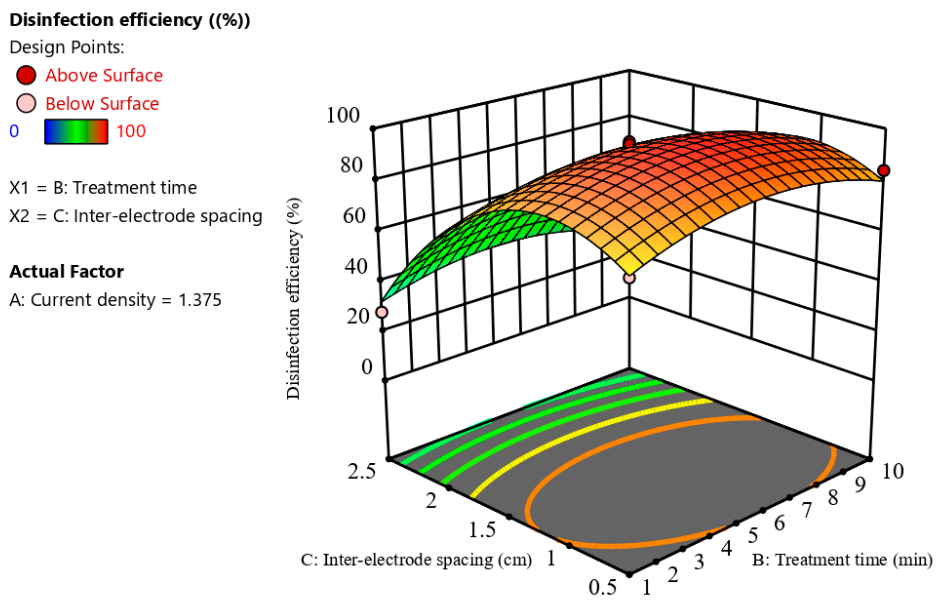

Figure 8. Three-dimensional graphical surface optimization of disinfection efficiency vs. interelectrode spacing and treatment time.

3.2.4. Optimization

In order to determine the optimum operating conditions for treating the canal water by using the electrochemical disinfection method, simultaneous optimization of disinfection efficiency is performed through Derringer's desirability function method [40]. Here, the response disinfection efficiency was fixed as maximizing while the operating variables (A, B, and C) were chosen within the range. A point is evaluated via numerical optimization, which maximizes the desirability function. Table 4 lists optimum operating conditions to obtain the maximum disinfection efficiency. The optimum value of current density, operating time, and interelectrode spacing was found to be 1.52 mA/cm^2, 6.35 min, and 1.13 cm, respectively, resulting in a statistically 98.08% elimination of the total coliform. Triplicate experiments were performed on the suggested optimum conditions, and the total coliform was tested. Coliform was not detected in any of these samples, suggesting that the treated water is biologically safe to consume.

Table 4. Optimum conditions for disinfection efficiency.

Current Density (mA/cm^2)	Interelectrode Spacing (cm)	Treatment Time (min)	Disinfection Efficiency (%)	Desirability
1.52	1.13	6.35	98.08	0.98

4. Conclusions

The electrochemical technique is an efficient, environment-friendly, and economical method for treating biological contaminants in drinking water. The use of stainless steel 316-grade electrodes was found suitable to serve the purpose. The canal water's turbidity and biological loads were 92.8 NTU and 10,000 CFU/mL, respectively. To reduce the turbidity to less than 5 NTU, as per the Guidelines for Drinking-Water Quality (GDWQ) [41], two different coagulants were tested using a jar test: ferric chloride ($FeCl_3$) and alum ($Al_2(SO_4)_3 \cdot 16H_2O$). It was observed that a 10ppm dosage of both coagulants is appropriate to lower the turbidity level to GDWQ standards. Ferric chloride appears to perform better in turbidity removal than aluminum sulfate; however, both meet the quality standards desired for post-treatment.

For electrochemical disinfection, preliminary experiments were performed to identify the factors affecting the system performance and their domains. Current density, treatment time, and interelectrode spacing are vital factors in the system design. The experiments were designed statistically using the Box–Behnken design (BBD) model. Seventeen experiments were performed at designed conditions with five replicates. The conditions were then optimized using analysis of variance (ANOVA). A mathematical relationship defined disinfection efficiency as a function of operating variables such as current density, treatment time, and interelectrode spacing. The optimum value of current density, treatment time, and interelectrode spacing was found to be 1.52 mA/cm^2, 6.35 min, and 1.13 cm, respectively, resulting in statistically 98.08% elimination of the total coliform. The current study was limited to the use of specific electrodes material used in a batch type electrochemical reactor. For future research, a flow arrangement is recommended for continuous water treatment. Moreover, a comparative analysis for performance prediction with different electrode material is also suggested. Optimizing the geometry of electrochemical reactor to have uniform potential distribution is also recommended for future work.

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References

1. Plappally, A.K.; Lienhard, J.H.V. Energy requirements for water production, treatment, end use, reclamation, and disposal. *Renew. Sustain. Energy Rev.* **2012**, *16*, 4818–4848. [CrossRef]
2. Hoekstra, A.Y.; Chapagain, A.K. The water footprints of Morocco and the Netherlands: Global water use as a result of domestic consumption of agricultural commodities. *Ecol. Econ.* **2007**, *64*, 143–151. [CrossRef]
3. Czarny, J.; Präbst, A.; Spinnler, M.; Biek, K.; Sattelmayer, T. Development and simulation of decentralised water and energy supply concepts—Case study of rainwater harvesting at the angkor centre for conservation of biodiversity in cambodia. *J. Sustain. Dev. Energy Water Environ. Syst.* **2017**, *5*, 626–644. [CrossRef]
4. Liu, Y.; Chen, B.; Wei, W.; Shao, L.; Li, Z.; Jiang, W.; Chen, G. Global water use associated with energy supply, demand and international trade of China. *Appl. Energy* **2020**, *257*, 113992. [CrossRef]
5. Abu Hasan, H.; Muhammad, M.H.; Ismail, N.I. A review of biological drinking water treatment technologies for contaminants removal from polluted water resources. *J. Water Process Eng.* **2020**, *33*, 101035. [CrossRef]
6. Sonone, S.S.; Jadhav, S.V.; Sankhla, M.S.; Kumar, R. Water Contamination by Heavy Metals and their Toxic Effect on Aquaculture and Human Health through Food Chain. *Lett. Appl. NanoBioSci.* **2020**, *10*, 2148–2166. [CrossRef]
7. Saleh, I.A.; Zouari, N.; Al-Ghouti, M.A. Removal of pesticides from water and wastewater: Chemical, physical and biological treatment approaches. *Environ. Technol. Innov.* **2020**, *19*, 101026. [CrossRef]
8. Garcia-Segura, S.; Nienhauser, A.B.; Fajardo, A.S.; Bansal, R.; Conrad, C.L.; Fortner, J.D.; Marcos-Hernández, M.; Rogers, T.; Villagran, D.; Wong, M.S.; et al. Disparities between experimental and environmental conditions: Research steps toward making electrochemical water treatment a reality. *Curr. Opin. Electrochem.* **2020**, *22*, 9–16. [CrossRef]
9. Bharadwaj, K.K. Water Pollution: A Threatening Alarm to The Mankind. *Innov. Res. Thoughts* **2018**, *4*, 204–209.
10. Ogbo, F.A.; Agho, K.; Ogeleka, P.; Woolfenden, S.; Page, A.; Eastwood, J.; Homaira, N.; Burrett, S.; Zwi, K.; Schaefer, M.; et al. Infant feeding practices and diarrhoea in sub-Saharan African countries with high diarrhoea mortality. *PLoS ONE* **2017**, *12*, e0171792. [CrossRef]
11. Martínez-Huitle, C.A.; Brillas, E. Electrochemical alternatives for drinking water disinfection. *Angew. Chem. Int. Ed.* **2008**, *47*, 1998–2005. [CrossRef] [PubMed]
12. Guerra, O.J.; Reklaitis, G.V. Advances and challenges in water management within energy systems. *Renew. Sustain. Energy Rev.* **2018**, *82*, 4009–4019. [CrossRef]
13. Kraft, A. Electrochemical water disinfection: A short review. *Platin. Met. Rev.* **2008**, *52*, 177–185. [CrossRef]
14. Chakrabarti, M.H.; Saleem, M.; Irfan, M.F.; Raza, S.; Hasan, D.B.; Daud, W.M.A.W. Application of waste derived activated carbon felt electrodes in minimizing NaCl use for electrochemical disinfection of water. *Int. J. Electrochem. Sci.* **2011**, *6*, 4470–4480.
15. Cossali, G.; Routledge, E.J.; Ratcliffe, M.S.; Blakes, H.; Fielder, J.E.; Karayiannis, T.G. Inactivation of E. coli, Legionella, and Pseudomonas in Tap Water Using Electrochemical Disinfection. *J. Environ. Eng.* **2016**, *142*, 04016063. [CrossRef]
16. Chaplin, B.P. *Advantages, Disadvantages, and Future Challenges of the Use of Electrochemical Technologies for Water and Wastewater Treatment*; Elsevier Inc.: Amsterdam, The Netherlands, 2018; ISBN 9780128131602.
17. Kerwick, M.I.; Reddy, S.M.; Chamberlain, A.H.L.; Holt, D.M. Electrochemical disinfection, an environmentally acceptable method of drinking water disinfection? *Electrochim. Acta* **2005**, *50*, 5270–5277. [CrossRef]
18. Jeong, J.; Kim, C.; Yoon, J. The effect of electrode material on the generation of oxidants and microbial inactivation in the electrochemical disinfection processes. *Water Res.* **2009**, *43*, 895–901. [CrossRef] [PubMed]
19. Nair, A.T.; Makwana, A.R.; Ahammed, M.M. The use of response surface methodology for modelling and analysis of water and wastewater treatment processes: A review. *Water Sci. Technol.* **2014**, *69*, 464–478. [CrossRef]
20. Darvishmotevalli, M.; Zarei, A.; Moradnia, M.; Noorisepehr, M.; Mohammadi, H. Optimization of saline wastewater treatment using electrochemical oxidation process: Prediction by RSM method. *MethodsX* **2019**, *6*, 1101–1113. [CrossRef]
21. Jeong, J.; Kim, J.Y.; Cho, M.; Choi, W.; Yoon, J. Inactivation of Escherichia coli in the electrochemical disinfection process using a Pt anode. *Chemosphere* **2007**, *67*, 652–659. [CrossRef]
22. Ndjomgoue-Yossa, A.C.; Nanseu-Njiki, C.P.; Kengne, I.M.; Ngameni, E. Effect of electrode material and supporting electrolyte on the treatment of water containing Escherichia coli by electrocoagulation. *Int. J. Environ. Sci. Technol.* **2015**, *12*, 2103–2110. [CrossRef]
23. Riyanto; Agustiningsih, W.A. Electrochemical disinfection of coliform and Escherichia coli for drinking water treatment by electrolysis method using carbon as an electrode. *IOP Conf. Ser. Mater. Sci. Eng.* **2018**, *349*, 012053. [CrossRef]
24. Samarghandi, M.R.; Nemattollahi, D.; Asgari, G.; Shokoohi, R.; Ansari, A.; Dargahi, A. Electrochemical process for 2,4-D herbicide removal from aqueous solutions using stainless steel 316 and graphite Anodes: Optimization using response surface methodology. *Sep. Sci. Technol.* **2019**, *54*, 478–493. [CrossRef]
25. Casqueira, R.G.; Torem, M.L.; Kohler, H.M. The removal of zinc from liquid streams by electroflotation. *Miner. Eng.* **2006**, *19*, 1388–1392. [CrossRef]
26. Delaedt, Y.; Daneels, A.; Declerck, P.; Behets, J.; Ryckeboer, J.; Peters, E.; Ollevier, F. The impact of electrochemical disinfection on Escherichia coli and Legionella pneumophila in tap water. *Microbiol. Res.* **2008**, *163*, 192–199. [CrossRef]
27. Hyde, K. Turbidity measurement: Its application for water resource recycling in buildings. *Process Saf. Environ. Prot.* **2021**, *146*, 629–638. [CrossRef]

28. Malik, Q.H. Performance of alum and assorted coagulants in turbidity removal of muddy water. *Appl. Water Sci.* **2018**, *8*, 40. [CrossRef]
29. Agbovi, H.K.; Wilson, L.D. Optimisation of orthophosphate and turbidity removal using an amphoteric chitosan-based flocculant-ferric chloride coagulant system. *Environ. Chem.* **2019**, *16*, 599–612. [CrossRef]
30. Lee, H.J.; Halali, M.A.; Baker, T.; Sarathy, S.; de Lannoy, C.F. A comparative study of RO membrane scale inhibitors in wastewater reclamation: Antiscalants versus pH adjustment. *Sep. Purif. Technol.* **2020**, *240*, 116549. [CrossRef]
31. Baptisttella, A.M.S.; Araújo, A.A.D.; Barreto, M.C.; Madeira, V.S.; da Motta Sobrinho, M.A. The use of metal hydroxide sludge (in natura and calcined) for the adsorption of brilliant blue dye in aqueous solution. *Environ. Technol.* **2019**, *40*, 3072–3085. [CrossRef]
32. Daryabeigi, A.; Baghvand, A.; Zand, A.D.; Mehrdadi, N.; Karbassi, A. Optimizing Coagulation Process for Low to High Turbidity Waters Using Aluminum and Iron Salts. *Am. J. Environ. Sci.* **2010**, *6*, 442–448.
33. Ditta, A.; Ullah, K.S.; Farhat, I.; Zehra, U.; Ayoub, H.; Ahtisham, M.; Imtiaz, H.; Tabish, A.N. The Effect of Operational Variables on the Performance of Electrochemical Water Treatment for Drinking Purposes. In Proceedings of the 2021 4th International Conference on Energy Conservation and Efficiency (ICECE), Lahore, Pakistan, 16–17 March 2021. [CrossRef]
34. Ghasemian, S.; Asadishad, B.; Omanovic, S.; Tufenkji, N. Electrochemical disinfection of bacteria-laden water using antimony-doped tin-tungsten-oxide electrodes. *Water Res.* **2017**, *126*, 299–307. [CrossRef] [PubMed]
35. Bashir, M.J.K.; Aziz, H.A.; Yusoff, M.S.; Adlan, M.N. Application of response surface methodology (RSM) for optimization of ammoniacal nitrogen removal from semi-aerobic landfill leachate using ion exchange resin. *Desalination* **2010**, *254*, 154–161. [CrossRef]
36. GilPavas, E.; Arbeláez, P.; Medina, J.D.; Dobrosz-Gómez, I.; Gómez-García, M.Á. The electrochemical elimination of coliforms from water using BBD/Ti or graphite anodes: A comparative study. *Water Sci. Technol. Water Supply* **2018**, *18*, 408–417. [CrossRef]
37. Gómez-López, V.M.; Gobet, J.; Selma, M.V.; Gil, M.I.; Allende, A. Operating conditions for the electrolytic disinfection of process wash water from the fresh-cut industry contaminated with E. coli o157:H7. *Food Control* **2013**, *29*, 42–48. [CrossRef]
38. Qi, X.; Wang, T.; Long, Y.; Ni, J. Synergetic antibacterial activity of reduced graphene oxide and boron doped diamond anode in three dimensional electrochemical oxidation system. *Sci. Rep.* **2015**, *5*, 10388. [CrossRef]
39. Flox, C.; Cabot, P.L.; Centellas, F.; Garrido, J.A.; Rodríguez, R.M.; Arias, C.; Brillas, E. Electrochemical combustion of herbicide mecoprop in aqueous medium using a flow reactor with a boron-doped diamond anode. *Chemosphere* **2006**, *64*, 892–902. [CrossRef]
40. Thirugnanasambandham, K.; Sivakumar, V.; Maran, J.P. Response surface modelling and optimization of treatment of meat industry wastewater using electrochemical treatment method. *J. Taiwan Inst. Chem. Eng.* **2015**, *46*, 160–167. [CrossRef]
41. WHO. A Global Overview of National Regulations and Standards for Drinking-Water Quality. 2018. Available online: http://apps.who.int/bookorders (accessed on 15 July 2021).