

Investigation Removal Efficiency of Electrocoagulation Process as A Slaughterhouse Wastewater Treatment Technique: Toxicity Assessment.

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Abstract

The wastewater resulting from slaughtering and meat processing in slaughterhouses contains a high amount of organic matter. The discharge of these wastewaters into rivers, sewage networks or soil creates significant environmental pollution. In the present study, the treatment of wastewater from Erbil cattle and poultry slaughterhouse through an electrochemical coagulation method has been investigated. In experimental studies, the effects of current density, initial pH and support electrolyte concentration (Na_2SO_4) on chemical oxygen demand (COD) removal efficiency have been investigated. Through numerous treatment investigations with main slaughterhouse wastewater samples, the aluminum electrodes exhibited a removal efficiency of 82.43% at an electric current density of 20.00 mA.cm^{-2} and pH 5. While by using iron electrodes a COD removal efficiency reached 92.52% at an electric current density of 20 mA.cm^{-2} and pH 9. Despite the electrocoagulation (EC) lower COD, the system's effectiveness cannot be evaluated without taking the toxicity into account. For such cases, Microtox evaluations were done for the most efficient COD removal level. The electrocoagulation technique was shown to lower both toxicity and COD. As a result, it has been found that EC with iron and aluminum electrodes is an appropriate technique for treating slaughtering wastewater containing high levels of organic contaminants in terms of COD reduction and toxicity minimization.

1. Introduction:

The observed increase in domestic and industrial water use due to population growth puts the recovery and reuse of wastewater on the agenda. Industrial wastewater can be used as post-treatment, process water, or cooling water. The use for cooling water purposes is known as the most common use. In the water recycling system, it has been adopted as the main objective to remove pollutants at a sufficient level. The wastewater generated in the meat industry originates from slaughterhouses and rendering plants. The mentioned slaughterhouse wastewater is water with high COD and BOD levels,

in addition, contain blood, fats, protein and carbohydrate [1].

Slaughterhouses generate wastewater mostly during the slaughtering process, as well as during the cleaning of equipment and buildings and the generation of by-products. Consumption of water per decapitated animal differs according to the animal and employee used in the industry and ranges from 2 to 8.3 m^3 . Most of this amount is disposed of as wastewater, the volume of which is from 0.3 to 3.5 m^3 Per slaughtered animal reported in the literature [2].

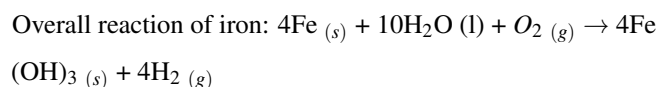
In recent years, there has been a rise in interest in the use of electrochemical techniques for phosphate extraction. [3] and boron removal from water, heavy metals destruction of poisonous and bio-refractory industrial wastewater [4].

Electrons are used as the primary reagent in a heterogeneous reaction in electrochemical procedures. When there are insufficient ionic components in the wastewater, the support-



ing electrolyte must be applied to increase ionic conductivity. Electrochemical treatment processes are economically and environmentally suitable alternatives, simply operable and eco-friendly in nature. In addition, treated water is clear, colorless potable and odorless with minimum production of sludge. These methods provide little to no risk of secondary water contamination.[5].

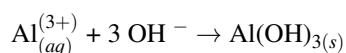
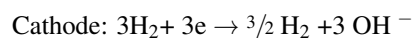
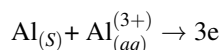
Among these processes, electro-coagulation (EC) relies on the in situ coagulant formation on the anode, such as aluminum and/or iron, oxidation of the anode by electrical current leads to generate metal ions which in turn reacts in an aqueous solution with water to form metal hydroxide or metal oxyhydroxide. Such species in turn help to destabilize suspended particles by reducing surface charge and promoting their aggregation. Also, the hydrogen gas formation at a cathode helps to eliminate pollutants through buoyancy [6]. Overall chemical reactions for both iron and aluminum electrodes in the media are:



The $\text{Fe}(\text{OH})_n$ (s) which are generated stay as the gelatinous suspension in the media, and have the potential to eliminate the pollutant from the solution via electrostatic methods, accompanied by a coagulation process [6].

Following are the primary chemical processes that take place during the electrocoagulation process on aluminum electrodes.

At the anode, the oxidation of aluminum happens.



By considering this study analyzes the effectiveness of iron and aluminum and electrodes for the EC treatment of Slaughterhouse wastewater effluent. Numerous studies used iron and aluminum electrodes for the treatment of different industrial wastewater [7], [8], [9] and [10]. However, the literature on the electrocoagulation technique for the treatment of slaughterhouse wastewater is limited [11], [12], [13] and [14]. Furthermore, except for a few studies on distinct wastewater types, concurrent assessment of toxicity after treatment was not a prevalent research approach.

The current study aims to investigate the treatability of Slaughterhouses that produce wastewater through EC using iron and aluminum electrodes, as well as the impacts of var-

ious effective parameters such as current density, initial pH and support electrolyte amount (Na_2SO_4). Iron and aluminum electrodes are cross-examined and compared within such parameters. The high removal efficiency has been obtained. Furthermore, toxicity assessments demonstrate that the electrocoagulation treatment technique has a significant effect on toxicity minimization. The results of the experiment are expected to inform the slaughterhouse wastewater treatment community about the importance of concurrent toxicity and COD analysis.

2. Materials and Methods:

2.1 Characterization and Sampling of Slaughterhouses Wastewater:

Real slaughterhouse wastewater was utilized in our studies to achieve realistic and acceptable findings. The slaughterhouse wastewater samples were obtained from local slaughterhouses located in the city of Erbil (Kurdistan region of Iraq). Producing approximately 250 tons of wastewater daily. The samples were kept in a refrigerator at 4 °C to maintain their original properties during experimental studies. Before starting the investigation, the collected samples were filtered through a 2 mm membrane to separate solids and feathers, lipids and other contaminants. The chemical analysis of Erbil slaughterhouses is specified in Table 1.

Table 1. Slaughterhouse wastewater Parameters before and after the treatment process.

parameters	Raw wastewater	Electrocoagulation	
		Using Fe	Using Al
pH	5	9	7.5
Conductivity ms/cm	2.45	4.8	3.4
BOD mg.L ⁻¹	960	60	74
COD mg.L ⁻¹	2000	149.6	351.4
BOD/ COD	0.48	0.40	0.21
Suspended solid mg.L ⁻¹	160.35	58.82	49.74

2.2 Experimental Setup:

The electrolysis reactor consists of a 500 ml beaker made from a magnetic stirrer. The electrolysis electrodes consist of six parallel (three anode and three cathode) electrodes made from iron or aluminum. The total immersed anode surface area is 100 cm². The power supply is a DC power supply (Sunshine P3005A, 0–8 A/0–70 V, Chinese). A pole changer was utilized to exchange the electrode's polarity at a specific setting period to ensure homogeneous erosion at the electrode sides, and the coagulants are evenly uniformly by symmetric electrode consumption. Various process parameters were carefully modified to provide optimal working conditions at room temperature. The effluent pH was initially adjusted with

0.1N NaOH and H₂SO₄. The conductivity levels and pH have been measured with a conductivity and pH meter (labForce M1000).

2.3 Analysis:

The COD levels of the samples were evaluated with a COD reactor using open reflux methods (HI-839800 25 Vial Thermo-Reactor). All COD evaluation steps were repeated to increase measurement accuracy, and average values were provided.

$$\text{Removal efficiency } \% = (C^0 - C_t) / C^0 \times 100 \quad (1)$$

Where C_0 denotes to initial concentration of COD, and C_t is the concentration at any time.

In this study, a standard toxicity evaluation method was used with a Microtox® Model 500 Analyzer provided by AZUR Environmental (Carlsbad, CA, USA). Microtox bioassay assays were performed on the treated slaughterhouse wastewater at predetermined time intervals to assess toxicity. The relative toxicity index is calculated as the equation below:

$$\text{RTI} = \frac{\%EC_{50at}(t_0)}{(\%EC_{50}(t))} \quad (2)$$

In which $EC_{50\%}$ at t_0 and $EC_{50\%}$ at t are the 5,15 minutes Microtox toxicity at (0) and (t) time intervals.

The efficiency of energy consumption (kWh.m^{-3}) has been calculated using the below equation:

$$\text{Energy Consumption} = \frac{V \times I \times t}{Vol.} \quad (3)$$

Where I is applied electrical current (A), v is the voltage, at t time (min) and vol is the sample volume (m^3).

3. Results and Discussion:

In the following part, the results of experiments for the electrochemical treatment of slaughterhouse wastewater by applying iron and aluminum electrodes across different electrochemical setting parameters are described. The efficiencies were determined at first based on the COD removed percentage throughout the treatment procedure. Once the optimal COD removal was determined, the appropriate results of treatment were examined for toxicity, which it is also plays a vital role in the long-term treatment of slaughterhouse wastewater. The values of energy consumption were also measured during the treatment process. The results reveal that the proposed treatment technique is an effective method of treating slaughterhouse wastewater in terms of COD removal and reduction of toxicity.

3.1 Effects of Current Density:

The electrical current density was considered to be a crucial parameter influencing the performance of electrochemical systems. The quantity of Fe^{2+} and Al^{3+} ions coagulants formed in the EC system is controlled by the current used in the system [15]. As expected, increasing the applied current density improved removal effectiveness due to the increase in metal ions production, which has an essential significant impact on precipitation [16]. Due to the power supply and working environment limitations, current density values of 18, 20 and 22 mA.cm^{-2} have been applied to the reactor to investigate their impact, and current density investigation was initially achieved at pH 5 (original pH) with both electrodes.

Figure 1 (a) demonstrates the current density influence on COD reduction by iron electrodes. The figure showed that a current density of 22 mA.cm^{-2} had much greater effect on COD reduction. For current densities of 18, 20 and 22 mA.cm^{-2} , the COD reduction efficiency was 50%, 59.5% and 77.8%, respectively. After 60 minutes, the COD removals came to a dead point.; accordingly, the results are given at a time interval of 60 min output of a process. Energy consumption for current densities of 18, 20 and 22 mA.cm^{-2} at the end of the electrolysis was 52.46, 60.64 and 79.27 kWh.m^{-3} respectively for treated wastewater. Because of the prominent COD reduction advantage, 22 mA.cm^{-2} operation was recommended for the rest of the research studies.

Figure 1 (b) depicts the effect of current density on COD reduction using aluminum electrodes. Current density experiments have been carried out at the pH 5 (original pH) values, the final pH values have been found to range between 7 and 8. COD removal efficiency values of 56 %, 79.3 % and 80.2% were obtained for current densities of 18 mA.cm^{-2} , 20 mA.cm^{-2} and 22 mA.cm^{-2} , respectively. At current densities of 20 and 22 mA.cm^{-2} , COD removal efficiency didn't differ significantly. Since current density has a direct effect on energy consumption, any rise in current density results in an increase in consumed energy per meter cubic of wastewater. [17]. Energy consumption amounts for current densities of 18, 20 and 22 mA.cm^{-2} were determined to be 41.9, 56.7 and 67 kWh.m^{-3} respectively for treated wastewater. A time interval of the experiment was also determined by COD removal efficiency versus the energy. Because non-significant change has been observed in COD removal rates after 70 minutes of electrolysis, the results for the experiment are reported within the time interval (0–70) min range.

3.2 Effect of Initial PH:

To observe the impacts of the initial pH of slaughterhouse wastewater on the EC for both electrodes (iron and aluminum), laboratory experiments were conducted by differing the starting point pH value.

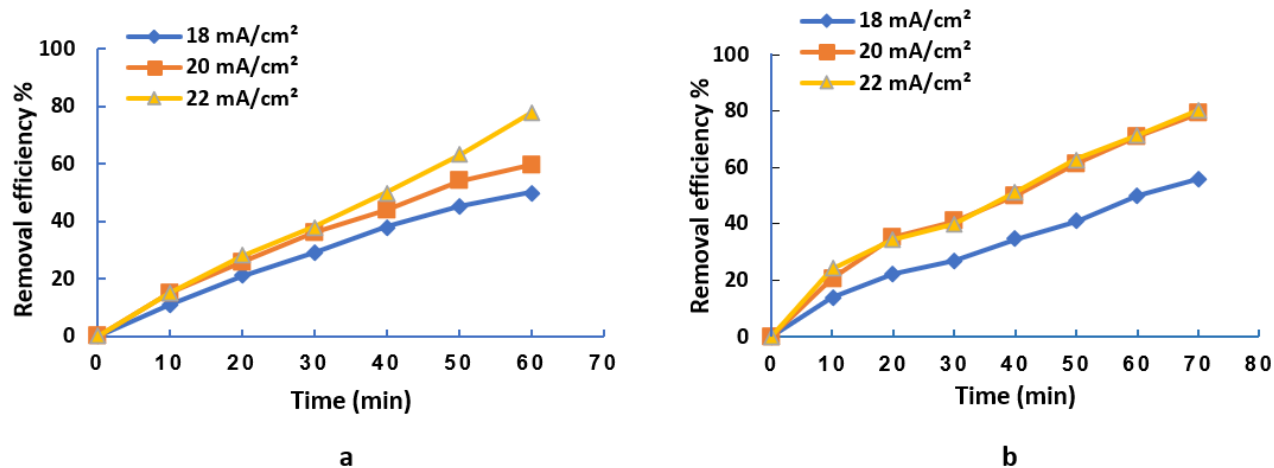


Figure 1. COD removal efficiency with time as a function of density. (a) Fe-electrode and (b) Al-electrode; ($C_o = 2000 \text{ mg L}^{-1}$, $\text{pH} = \sim 5$).

The recommended current density (22 mA.cm^{-2}) was used in the experiment using an iron electrode and the initial pH values were altered. Figure 2(a) exhibits the fluctuation in COD reduction efficiency as an influence of starting pH values. The iron electrode studies provide removal efficiencies of 72.63%, 78.12% and 92.52%, respectively, for the initial pH of 5, 7 and 9. According to removal levels, iron electrodes give greater COD reduction capabilities at comparatively higher pH levels. The gained result is consistent with the literature, which suggests that most iron coagulants are produced with higher pH levels. [18]. We noticed that the initial pH values that facilitate greater removal of COD, additionally facilitate minimum consumption of energy. When iron electrodes are applied, a high starting pH (9) level gives greater COD reduction with the minimum consumption of energy of 45.26 kWh.m^{-3} but at the low initial pH value consumes about 78.50 kWh.m^{-3} .

In the same manner, as the Fe-electrode experiment, the optimal current density (20 mA.cm^{-2}) has been used in treatment with the Al electrode. Three different values of pH were chosen 5 (original pH), 7 and 9. The COD removal efficiencies of 82.43 %, 64.12% and 50.2%, for pH 5, 7 and 9 were investigated, respectively. Figure 2(b). This indicates that the original pH (slightly acidic) can be regarded as appropriate for the treatment. This was also a cost-effective and efficient choice for the process because no additional chemicals were needed to regulate the pH. When the experiment started with pH 5 (original), the pH value was investigated to increase during the electrolysis period until reached pH 7.5. This increase in pH might be due to the electrolysis process, which produces hydrogen and hydroxyl ions [19]. At high concentrations and high starting pH, a gel layer occurs on

the anode surface, which is likely to interfere with aluminum hydrolysis. For this reason, a polar changer was used in the experiment to avoid gel formation. The acidic pH 5 was a favorable reduction with the minimum energy consumption of 53.56 kWh.m^{-3} whereas the highest initial (pH 9) level consumed about 80.12 kWh.m^{-3} . Based on the observations made above, the original pH 5 was chosen as the starting point in the remaining experiments.

3.3 Effects of Na_2SO_4 :

Because conductivity is directly connected to an electron transport speed in the media, it may possess an impact on electrochemical treatment. As the dosage of Supporting electrolytes increases, the conductivity of the media increases. Na_2SO_4 has been added as a supporting electrolyte to the media to improve its electrical conductivity and also to evaluate the impact of their dosage on COD reduction efficiency.

In the treatment using an iron electrode, additional experiments have been conducted with the addition of Na_2SO_4 as supporting electrolytes (0, 5 and 10 mM) at optimum pH level (pH 9) and 22 mA.cm^{-2} current density. For 0, 5 and 10 mM Na_2SO_4 , COD removal efficiency of 92.52%, 90.33 and 92.21% were obtained. This lack of relationship between extra Na_2SO_4 quantity and removal effectiveness was clearly noticed in the literature [20]. Because of the uneven performance of Na_2SO_4 , an additional energy consumption study has been conducted to clearly determine which support electrolyte dosage was recommended. At the end of electrolysis, the energy consumption for (0 mM, 5 mM and 10 mM) Na_2SO_4 were 48.12, 40.27 and 35.58 kWh.m^{-3} respectively. considering the limited variation in the cost of energy and the previously mentioned COD reduction efficiencies data, we concluded that additional Na_2SO_4 dosage has no clear im-

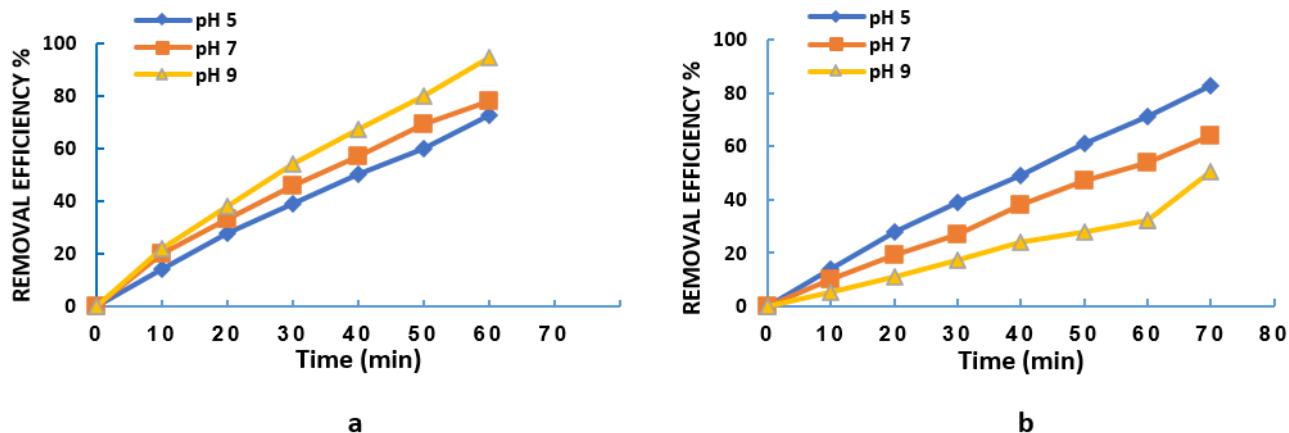


Figure 2. COD removal efficiency with time as a function of pH. (a) Fe-electrode ($i = 22 \text{ mA.cm}^{-2}$) and (b) Al-electrode (20 mA.cm^{-2}); ($C_0 = 2000 \text{ mg L}^{-1}$).

provement effect during electrolysis. This once again can be due to, the conductivity of the sample being enough high for electrolysis.

During electrocoagulation using an Al electrode, Additional studies were conducted with the addition of different amounts of supporting electrolyte (0 mM, 5 mM and 10 mM) within previously adjusted conditions (pH 5) and a current density (20 mA.cm^{-2}). The conductivities of slaughterhouse wastewater were measured to be (2.45, 4.27 and 6.75) mS cm^{-1} for 0, 5 and 10 $\text{mM Na}_2\text{SO}_4$, respectively. Results of the experiments reveal that the addition of Na_2SO_4 had no valuable effect. This might be because the electrical conductivity of raw slaughterhouse wastewater was strong enough to undertake electrochemical reactions. As a result, the remaining investigations were conducted without Na_2SO_4 .

3.4 Toxicity Assessment:

The continuous observation of the EC treatment process through toxicity characteristics and COD removal efficiency is a significant element in our study. The two different types of electrodes used in electrocoagulation have been examined for their toxicity reduction efficiencies at optimal COD removal. Under optimum conditions for experimentation, Microtox® bioassay methods have been used to evaluate the toxicity level of the slaughterhouse wastewater at specific time intervals of a treatment procedure, including starting time (0). Figures 2(a) and 2(b) show the relative toxicity levels at various time intervals, indicating the clear decline in toxicity levels over time. The decrease in toxicity was therefore demonstrated to be a substantial acquisition and advantage of electrolysis by EC process utilizing iron and aluminum electrodes. A relative toxicity index (RTI) for wastewater treated with iron electrodes clarifies that toxicity reduces during the first 10

min after 20 min exhibiting a constant line below 0.05 RTI Figure 3(a). When compared to the initial toxicity level of slaughterhouse wastewater, this represents a 90% reduction in toxicity. while relative toxicity for wastewater treated with aluminum electrode clarifies that toxicity reduces along the first 10 min and at the end of 30 min exhibits a constant line below 0.4 RTI Figure 3(b). This is equivalent to a 65 % decline in toxicity when compared to the initial toxicity value of slaughterhouse wastewater.

The final toxicity observations indicate that the iron electrode has a particularly beneficial effect, according to the reduction of toxicity. It should be noted that the initial (original) slaughterhouse wastewater toxicity levels had been exceedingly high; hence the graphs merely illustrate a relative decline. The results we obtained show the obvious relationship between toxicity removal and COD reduction in the treatment of mentioned wastewater by EC. Figures 4(a) and 4(b) reveal that the positive inclinations of the toxicity vs. COD plots for both electrodes confirm our assumption of a positive relationship between COD and toxicity.

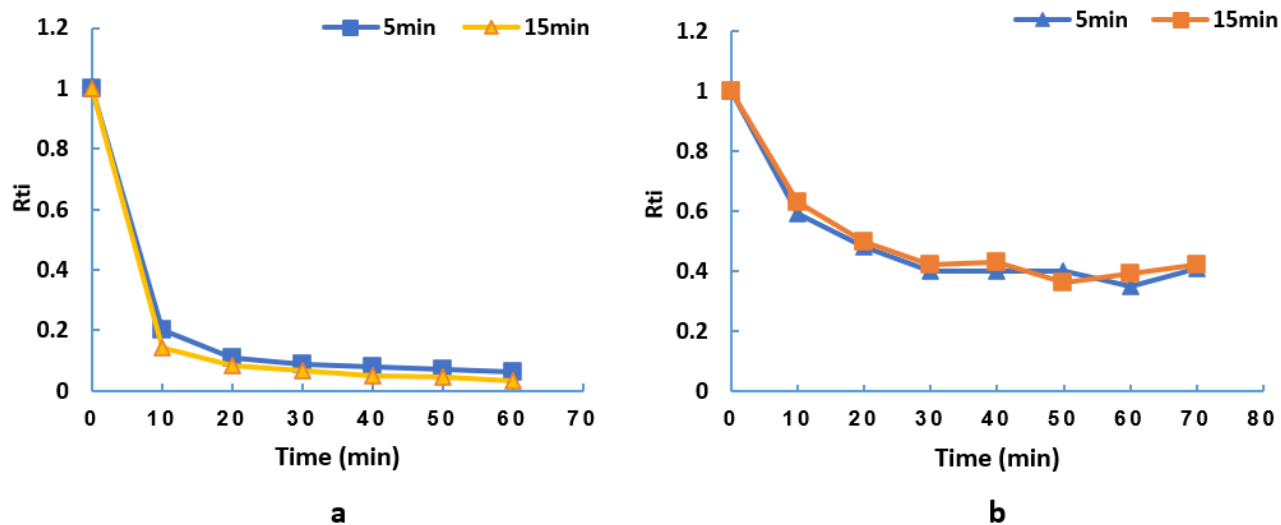


Figure 3. Toxicity variation with time in 5 and 15min slaughterhouse wastewater toxicity for (a) Fe-electrode ($i = 22 \text{ mA.cm}^{-2}$; $\text{pH} \approx 9$) and (b) Al-electrode (20 mA.cm^{-2} ; $\text{pH} = 5$); ($C_0 = 2000 \text{ mg L}^{-1}$).

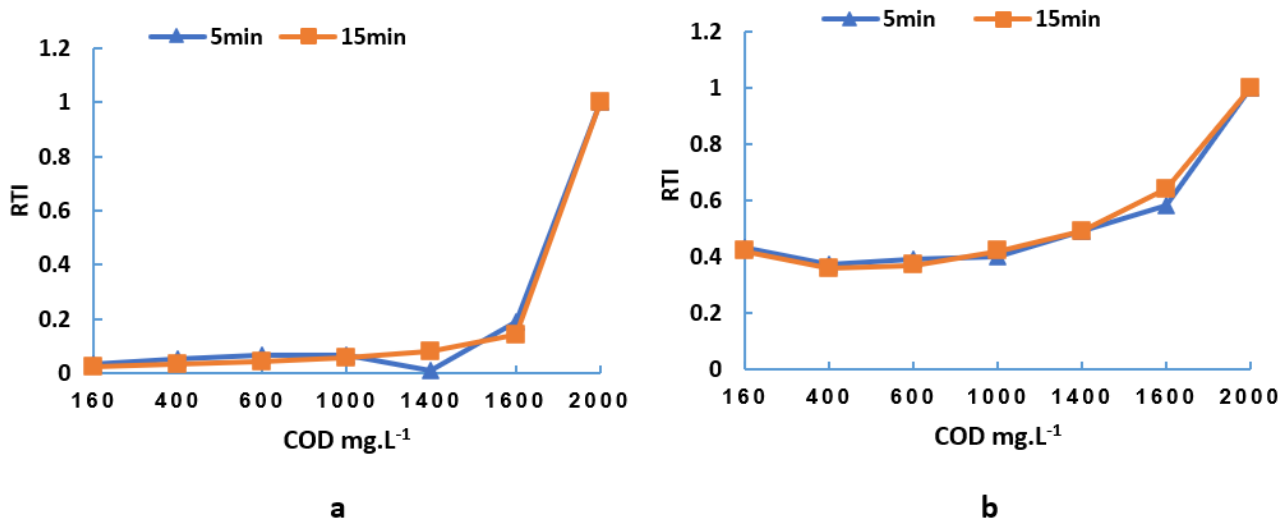


Figure 4. Toxicity variation with COD in 5 and 15min slaughterhouse wastewater toxicity for (a) Fe-electrode ($i = 22 \text{ mA.cm}^{-2}$; $\text{pH} \approx 9$) and (b) Al-electrode (20 mA.cm^{-2} ; $\text{pH} = 5$); ($C_0 = 2000 \text{ mg L}^{-1}$).

4. Conclusion:

This research presents the COD removal and toxicity evaluation results of slaughterhouse wastewater by EC treatment technique (iron and aluminum) for a different parametric such type of electrode, current density, initial pH and addition Na_2SO_4 are presented in this article. The ideal operational parameters for aluminum and iron electrodes for maximal COD elimination efficiency have been determined and these settings were subsequently evaluated for toxicity reduction performance.

The optimal experimental conditions identified for electrocoagulation using iron electrodes were: current density $22 \text{ mA}\cdot\text{cm}^{-2}$; pH 9, no supporting electrolyte used. In these circumstances, the reduction of COD from slaughterhouse wastewater was 92.52 %. The treatment process used $48.12 \text{ kWh}\cdot\text{m}^{-3}$ of energy, and the COD value after one hour of treatment was $149.6 \text{ mg}\cdot\text{L}^{-1}$, subsequently was shown to be even more efficient in terms of COD reduction. With an aluminum electrode, the most effective experimental conditions are shown as shown: current density $20 \text{ mA}\cdot\text{cm}^{-2}$; starting pH= 5, without the addition of supporting electrolyte. At these conditions, COD reduction from wastewater was 82.43 %, after 70 min of electrolysis. In this setting, the treatment used $53.56 \text{ kWh}\cdot\text{m}^{-3}$ of energy. Toxicity assessments for the suggested treatment techniques were performed as a key contribution. Similar to the positive effect of COD removal, the toxicity investigation shows that this system is viable for toxicity reduction, with the iron electrode providing a clear advantage. Our investigations also reveal that COD removal efficacy is directly related to toxicity reduction rate. Because wastewater's original COD and toxicity levels are exceedingly high, it could be considered to use EC coupled with other innovative processes to further decrease toxicity and COD to acceptable limits. these combinations are recommended as additional research to the present study.

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Data Availability Statement: All of the data supporting the findings of the presented study are available from corresponding author on request.

Declarations:

Conflict of interest: The authors declare that they have no conflict of interest.

Ethical approval: The manuscript has not been published or submitted to another journal, nor is it under review.

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التحقيق من كفاءة إزالة عملية التخثير الكهربائي كتقنية لمعالجة مياه الصرف الصحي في المسلخ: تقييم السمية

محمد عزيز عثمان

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الخلاصة

تحتوي مياه الصرف الصحي الناتجة عن الذبح ومعالجة اللحوم في المسلخ على كمية عالية من المواد العضوية. يؤدي تصريف هذه المياه العادمة في الأنهار أو شبكات الصرف الصحي أو التربة إلى تلوث بيئي كبير. في هذه الدراسة، تم التحقيق في معالجة مياه الصرف الصحي من مسلخ الماشية والدواجن في أربيل من خلال طريقة التخثير الكهروكيميائي. في الدراسات التجريبية، وقد تم التحقيق في آثار الكثافة التيار، ودرجة الحموضة الأولية، تركيز الإلكتروليتات (Na_2SO_4) على مطلوب الأوكسجين الكيميائي (COD) كفاءة إزالة. من خلال العديد من تحقيقات المعالجة مع عينات مياه الصرف الصحي الرئيسية للمسلخ، أظهرت أقطاب الألنيوم كفاءة إزالة بنسبة 82.43% عند كثافة تيار كهربائي تبلغ 20.00 مللي أمبير / سم² ودرجة الحموضة 5. أثناء استخدام أقطاب الحديد، وصلت كفاءة إزالة سمك القد إلى 92.52% بكثافة تيار كهربائي تبلغ 20 مللي أمبير / سم² ودرجة الحموضة 9. على الرغم من التخثير الكهربائي يقلل مطلوب الأوكسجين الكيميائي، لا يمكن تقييم فعالية النظام دون أخذ السمية في الاعتبار. في مثل هذه الحالات، تم إجراء تقييمات ميكروتوكس لنتائج لا أكثر كفاءة في تقليل مطلوب أكسجين الكيمياء. وقد تبين أن تقنية التخثير الكهربائي تقلل من السمية و مطلوب أكسجين الكيمياء. ونتيجة لذلك، فقد وجد أن طريقة التخثير الكهروكيميائي مع أقطاب الحديد والألومنيوم هي تقنية مناسبة لمعالجة مياه الصرف الصحي في المذابح التي تحتوي على مستويات عالية من الملوثات العضوية من حيث الحد من س مطلوب أكسجين الكيمياء وتقليل السمية.

الكلمات الدالة: مسلخ؛ مياه الصرف الصحي؛ التخثير الكهروكيميائي؛ مطلوبية الأوكسجين الكيميائية؛ السمية.

التمويل: لا يوجد.

بيان توفر البيانات: جميع البيانات الداعمة لنتائج الدراسة المقدمة يمكن طلبها من المؤلف المسؤول.

اقرارات:

تضارب المصالح: يقر المؤلفون أنه ليس لديهم تضارب في المصالح.

الموافقة الأخلاقية: لم يتم نشر المخطوطة أو تقديمها لمجلة أخرى، كما أنها ليست قيد المراجعة.