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RESEARCH ARTICLE-CHEMISTRY

Treatment of Slaughterhouse Wastewaters with $Ti/IrO_2/RuO_2$ Anode and Investigation of Energy Consumption

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Abstract

In the present study, the treatment of slaughterhouse wastewater (SWW) by electro-oxidation method and the energy consumption of the system during the process were investigated. The removal of chemical oxygen demand (COD), color, turbidity, and suspended solids (SS) from wastewater is emphasized. $Ti/IrO_2/RuO_2$ was used as the anode material and uncoated titanium was used as the cathode material in the experiments. The experiments were carried out in a glass reactor; the supporting electrolyte (SE) type, concentration, initial pH value, and current density were studied under different conditions. Na_2SO_4, $NaNO_3$ NaCl, and KCl salts have been used as SE. Salt studies were carried out at 0.1, 0.2, 0.3 and 0.4 M concentrations. The current density was studied at 4.06 mA/cm^2, 6.09 mA/cm^2, 8.12 mA/cm^2, 10.15 mA/cm^2, 12.18 mA/cm^2. Studies have been carried out at pH 3–9–11 and natural wastewater pH values. The optimum conditions to be used in electro-oxidation of SWW with high organic content have been determined. The results have revealed that as the current density increased, the efficiency increased, and the highest removal was achieved at the natural pH value of the NaCl salt. Studies have shown that color, turbidity, and SS removal for SWW is approximately 90%. COD was found to be 66.46% at the lowest current density. The energy consumption in the selected optimum conditions was found to be 183.6 kWh/m^3.

Keywords Electro-oxidation · Electrochemical treatment · Oxidation · Slaughterhouse wastewater treatment · $Ti/IrO_2/RuO_2$ anode

1 Introduction

Today, industrialization, technological developments, and rapid population growth affect our world negatively. Water resources are being consumed rapidly with the increasing population, which is risky for all living creatures. Traditional methods used for the treatment of wastewater delivered to the receiving environment require a lot of chemicals and high costs. Electrochemical treatment methods are used as an alternative to those methods. Recent use of electrochemical treatment methods in both water and wastewater treatment is becoming common because of their easy availability, simple operating conditions, and high activity. Electro-oxidation was based on the principle of indirect oxidation, using an insoluble anode material such as graphite [1, 2] coated titanium [3, 4] platinum [5, 6] boron-coated diamond [7, 8]. Organic pollutants can be split by the oxidation process directly or indirectly oxidation on the anode surface [9]. The United States Environmental Protection Agency has identified as one of the most hazardous wastewaters slaughterhouse wastewaters [10, 11]. Slaughterhouse wastewaters (SWW) are rich in organic content (blood, skin, manure) and heavy metals, so they must be managed properly. Among different components of SWW, FOG (fats oil and grease) comprises 2–20% of total COD in SWW [12]. According to the 2007 data of the World Bank, 2.5–40 m^3 wastewater per ton is generated as a result of the process in meat production in a slaughterhouse [13]. Studies have demonstrated that the wastewater from the meatpacking sector, which has a high level of oil, SS, COD, nitrate, and chloride, has considerably contributed to water contamination [14]. It has also been reported that these wastewaters are quite rich in terms of nitrogen and phosphorus. Besides, SWW pH was found to be in the range of 6.5–7.6 with high turbidity, red in color, and offensive odor [15, 16]. The reactions occurring at the anode

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Table 1 General characteristics of slaughterhouse wastewater [13]

Parameter	Range	Mean
TOC (mg/L)	700–1200	546
BOD5 (mg/L)	150–4635	1249
COD (mg/L)	500–15,900	4221
TN (mg/L)	50–841	427
TSS (mg/L)	270–6400	1164
pH	4.90–8.10	6.95
TP (mg/L)	25–200	50
Orto-PO_4 (mg/L)	20–100	25
Orto-P_2O_5 (mg/L)	10–80	20
K (mg/L)	0.01–100	90
Color (mg/L Pt scale)	175–400	290
Turbidity (FAU)	200–300	275

FAU formazine attenuation units

and cathode during the process are shown in the following Eqs. (1–6) [17].

Cathodic reactions:

$$2H_2O + 2e^- \rightarrow 2OH^- + H_2 \tag{1}$$

$$OCl^- + H_2O + 2e^- \rightarrow Cl^- + 2OH^- \tag{2}$$

Anodic reactions:

$$2Cl^- \rightarrow Cl_2 + 2e^- \tag{3}$$

$$6HOCl + 3H_2O \rightarrow 2ClO_3^- + 4Cl^- + 12H^+ + 1.5O_2 + 6e^-$$
$$2H_2O \rightarrow O_2 + 4H^+ + 6e^- \tag{4}$$

Solution reactions:

$$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^- \tag{5}$$

$$HOCl \rightarrow H^+ + OCl^- \tag{6}$$

The general characteristics of the slaughterhouse wastewater and the properties of the water used in this study are shown in Table 1 [13].

In this study, the treatment of slaughterhouse wastewater containing high organic pollution with mixed coating anode ($Ti/IrO_2/RuO_2$) was investigated. In the studies, the removal efficiencies of COD, color, turbidity and SS parameters were studied. The study measured the mixed coating anode efficiency in the removal of turbidity and SS, as well as the removal of color parameter, which is an important problem in SWW. In this study, a comparison of the treatment efficiency of slaughterhouse wastewater made by using different anode types and different electrochemical processes was also made. Data for these comparisons are shown in Table 2 [18–21].

2 Material and Method

2.1 Experimental Setup

During the experiments, a jacketed glass reactor with a volume of 2000 mL at a depth of 15 cm was used. Eight electrodes, four anodes and four cathode plates were used in the experiments. The electrode dimensions are 70 × 100 × 2 mm and the distance between the electrodes is 5 mm. The wet surface area of all plates was calculated as approximately 2464 cm^2. To increase the surface area, the electrodes are connected parallel to each other. DC-Power Supply brand KXN-3050D model power supply was used to provide the required electric current. With this power source, electricity was supplied to the system and the wastewater was mixed with the help of a magnetic stirrer during the experiment. With the Thermo Orion 5 star brand device, the pH, conductivity and temperature of the wastewater were measured at regular intervals from the start of the experiment. Analytical measurements were made using standard methods for wastewater samples. COD measurements were made using the dichromate open reflux method [22]. Color measurements were made at 430 nm wavelengths, turbidity measurements were made at 530 nm and SS measurements at 660 nm wavelengths with a Lovibond Multi Direct brand photometer. COD analysis results were measured with a Hach Lange DR5000 brand spectrophotometer at 600 nm wavelength. $Ti/IrO_2/RuO_2$ was used as the anode material, and sieve type uncoated titanium was used as the cathode material. 1200 mL of wastewater sample was used for the experiments. The effects of SE type and concentration, initial pH value of wastewater and flow density were investigated as treatment parameters. The experimental setup is given in Fig. 1. All analyzes in these trials were performed in Balıkesir University Environmental Engineering Laboratory. Indirect oxidation of organic compounds on the anode surface is shown in Fig. 2. In addition, the characteristic properties of the wastewater used in the studies are shown in Table 3.

Table 2 Comparison of some studies with different processes and different types of anodes

Wastewater type	Process	Optimal operating conditions	Pollutant removal (%)	References
Raw SWW (poultry)	Electro-Fenton process (batch operation mode)	Iron electrodes, interelectrode gap 3 cm pH 4.38 and 3.4, current density 74.1 and 67.9 mA/cm^2, molar ratio (H_2O_2/Fe^{2+}) 3.7 and 3.6, H_2O_2/SWW ratio 1.6 and 1.4 mL/L, treatment time 55.6 49.2 min for COD and color removal, respectively	COD (92.4%), color removal (88.1% at 340 nm)	[18]
Raw SWW (cattle)	Electrochemical oxidation process (batch operation mode)	pH 7, Ti/Pt mesh type anode, Ti mesh type cathodes, interelectrode gap 3 mm, supporting electrolyte 0.025 M NaCl, current density 4.73 mA/cm^2, treatment time 4 h	TOC (88%), COD (92.2%), TN (93.5%), color (99.9%), TSS (99.5%)	[19]
Raw SWW (pig)	Electrochemical oxidation-related process (semi-continuous operation mode)	pH 7.5, BDD anode, platinum cathode, interelectrode gap 3.7 mm, specific charge 7.3 A h/L, treatment time 8 h	COD (48.3–95.9%), color (100%), TSS (61.5%)	[20]
Synthetic and raw SWW (cattle)	Aerobic–anoxic process (batch operation mode)	Temperature 37 °C, treatment time 6 and 1 day, respectively, Scenedesmus obliquus bacteria, pH 6.8	COD (48–70%), Nitrogen (> 95%), phosphorous (63–99%)	[21]

3 Results

3.1 Effect of Supporting Electrolyte Type

The effect of SE type on the removal efficiency for $Ti/IrO_2/RuO_2$ anodes was investigated. At the same concentrations (0.2 M), NaCl KCl, $NaNO_3$, and Na_2SO_4 were studied at 400 rpm stirring speed, 4.06 mA/cm^2 current density, and wastewater natural pH (≈ 7). The results are shown in Fig. 3. As a result of the experiments, each of which lasted 3 h, the highest COD removal efficiency was NaCl with 66.46%. Afterward, the COD removal efficiencies were found to be 54.50%, 30.88%, 25.52%, and 20.88% in the trials KCl, $NaNO_3$, Na_2SO_4 and without SE, respectively. In experiments without NaCl, KCl, $NaNO_3$, Na_2SO_4 and SE, the color yields were 98.67%, 98.26%, 94.95%, 91.35% and 81.96%, respectively. The turbidity efficiencies were 98.47%, 94.81%, 87.89%, 83.25%, 73.95%, and SS yields were 98.91%, 98.30%, 95.65%, 91.35%, and 85.61%, respectively. The obtained results showed that the salts used in electro-oxidation had a significant effect on the purification efficiency by increasing the conductivity. Chloride ion has a positive effect on purification in electro-oxidation trials and consequently increase efficiency. The most common electrolyte used is sodium chloride (NaCl) as it is inexpensive and readily available [23]. NaCl has the highest removal efficiency, which is similar to observations reported in the literature. $NaNO_3$ and Na_2SO_4 electrolytes were not as effective as NaCl [19]. SE are key factors during electro-oxidation of wastewater as they increase the electrolyte conductivity and hence decrease the energy consumption of the electrochemical cell [24]. It was decided to use the NaCl salt, which reached the highest yield, in subsequent trials.

$$\text{Removal efficiency } \eta(\%) = \left(\frac{C_0 - C_e}{C_0}\right) \times 100 \qquad (7)$$

C_0 and C_e refer to "initial and final pollutant concentrations".

3.2 Effect of Supporting Electrolyte Concentration

The type and concentration of the supporting electrolytes utilized have a significant impact on electrochemical processes [25]. NaCl, which was seen to be the most effective SE in the trials, was decided to be the electrolyte to be used in the next trials. Studies were carried out at 0.1–0.4 M concentrations for 400 rpm stirring speed and 4.06 mA/cm^2 current density (Fig. 4). COD obtained in the results of 0 M, 0.1 M, 0.2 M, 0.3 M, and 0.4 M trials the removal efficiencies are as follows, respectively. 20.88%, 52.25%, 66.46%, 70.14% and 74.48%. As seen in the results shown in Fig. 4, the removal efficiency increases as the SE concentration is increased. Because the increase in the concentration means the increase in the chlorine ions given into the water and with

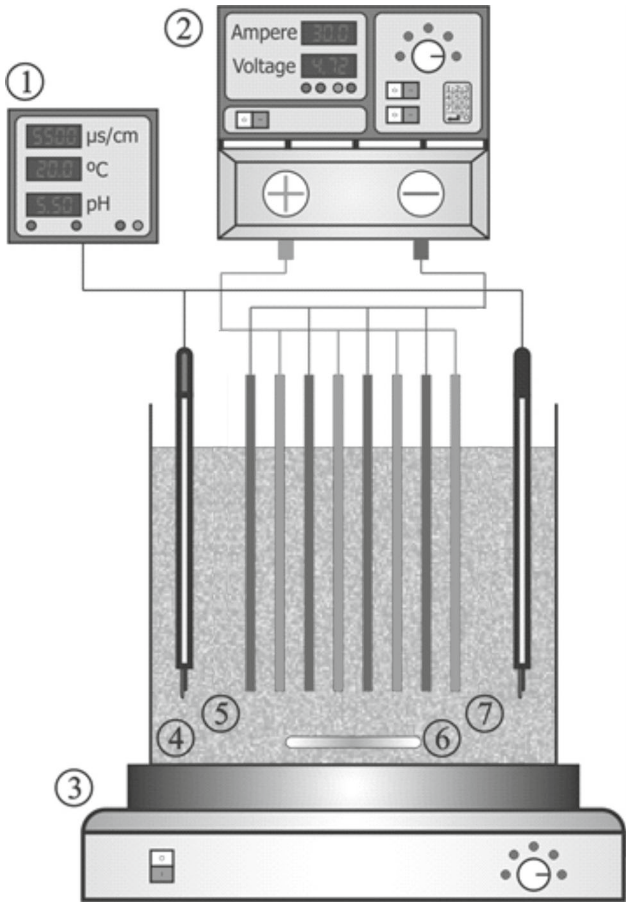


Fig. 1 Schematic diagram of the electro-oxidation process (1-Multi-parameter meter, 2-Direct current power supply, 3-Magnetic stirrer, 4-Wastewater, 5-Steel cathode, 6-Magnet, 7-Ti/IrO_2/RuO_2 Anode)

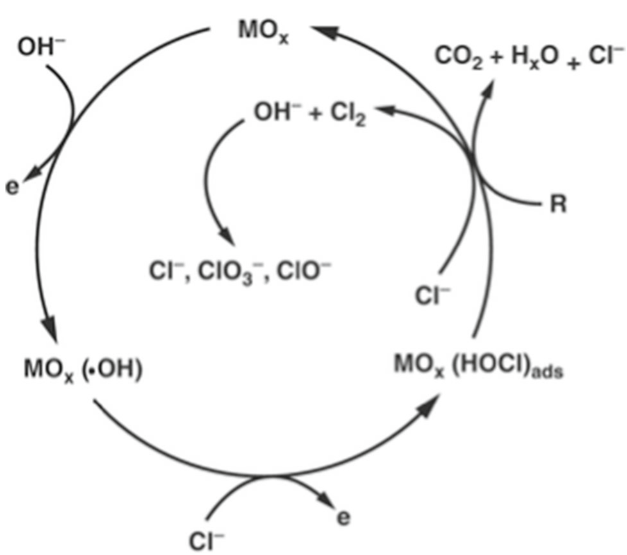


Fig. 2 Content of organic compounds indirect anode oxidation at the anode surface

it the increase in the conductivity. It facilitates the breakdown of chlorine pollutants contained in the structures of NaCl and KCl electrolytes. However, since there is not much change after 0.2 M, the optimum condition was chosen as 0.2 M to avoid excessive chemical consumption. By increasing the concentration of SE, the population of ions in the solution and also in the electrode/electrolyte interface will increase, also in low concentrations of SE the thickness of the diffusion layer increases. For these reasons, it is very important to optimize the concentration of SE [26]. In conclusion, the addition of chloride had a significant effect on the EO treatment of SWW even with both low current density and low chloride concentration [19].

3.3 The Effect of Wastewater Initial pH Value on Removal Efficiency

Initial pH is one of the factors that affect electro-oxidation. It is more important than current density [27]. pH affects the conductivity, zeta potential as well as electrode dissolution in the wastewater solution [23]. In this part of the study, studies were carried out at a constant temperature, 4.06 mA/cm^2 current density, and pH 3.0, 7.0, 9.0, 11.0 using 0.2 M NaCl to examine the effect of the initial pH of the wastewater. COD, color, turbidity, and SS in wastewater were investigated at 400 rpm mixing speed and reaction time of 3 h. The results are shown in Fig. 5. As can be seen from the graphs, the highest treatment efficiency was achieved at approximately 7, where the initial pH value of the wastewater is the highest natural pH. The initial pH of SWW was a very substantial parameter for the treatment of SWW by EO [19]. It was observed that the yield was relatively low in very acidic and very basic conditions. Therefore, the ideal pH value for the studies was determined as natural pH. COD, color, turbidity and SS removal for natural pH are as follows: 66.46%, 98.67%, 98.47% and 98.91%, respectively. Strong oxidants (ClO^-, HClO and Cl_2), which can play an important role in the oxidation of organics through "mediated oxidation" on the anode surface, are the main oxidizing agents. Of these, ClO^- is dominant above pH 7, HClO from pH 7 to 3, and Cl_2 at pH less than 3 [28].

3.4 The Effect of Current Density on Removal Efficiency

In the experiments with Ti/IrO_2/RuO_2 anode, wastewater was examined at a natural pH value with 0.2 M NaCl SE. The results are shown in Fig. 6. At the end of the 3-h reaction time, the COD removal efficiency obtained at 4.06, 6.09, 8.12, 10.15 and 12.18 mA/cm^2 current densities were found to be 66.46%, 74.47%, 82.36%, 94.57% and 98.87%, respectively. Based on these results, we can conclude that the current density affects the treatment efficiency significantly. As the

Table 3 Wastewater characteristic used in experiments

Parameter	Concentration (mg/L)			
	Highest	Lowest	Medium	Standard deviation
Total COD	6691.20	626.45	3099.82	1208.05
Dissolved COD	6565.80	605.34	2726.96	1036.59
TCOD/DCOD	1.00	0.57	0.87	0.10
BOD	5145.00	325.00	2173.00	627.12
Suspended solids	520.00	35.00	158.91	94.55
Total solid matter	13,635.00	310.90	2688.30	2156.27
Total nitrogen	632.00	48.00	213.67	163.82
Ammonium nitrogen	144.00	2.30	50.96	44.15
Nitrate nitrogen	44.00	5.00	13.36	9.62
Total phosphorus	80	7	33	18
Oil–Gress	387.50	21.4	124.7	99.84
pH	7.8	5.9	6.3	1.24

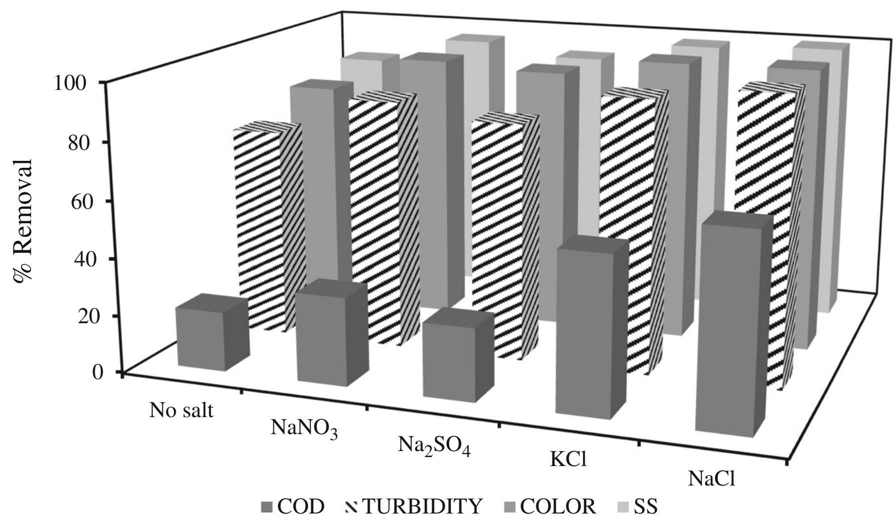


Fig. 3 Effect of SE type on COD, color, turbidity and SS removal (400 rpm, 4.06 mA/cm^2, natural pH (≈ ~ 7), 0.2 M NaCl)

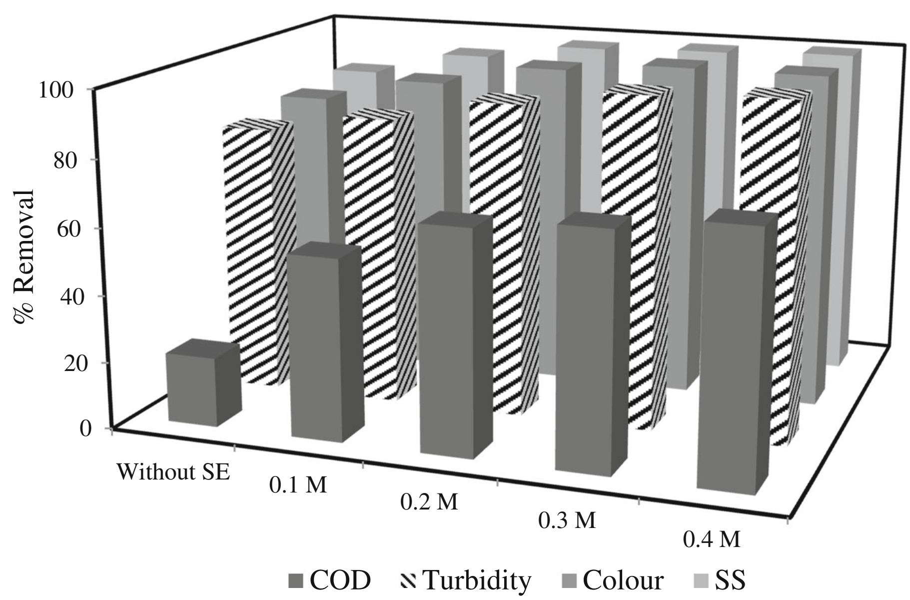


Fig. 4 Effect of SE concentration on COD, color, turbidity and SS removal (400 rpm, 4.06 mA/cm^2, natural pH (≈ 7), NaCl)

Fig. 5 Effect of initial pH value on COD, color, turbidity and SS (400 rpm, 4.06 mA/cm^2, 0.2 M NaCl)

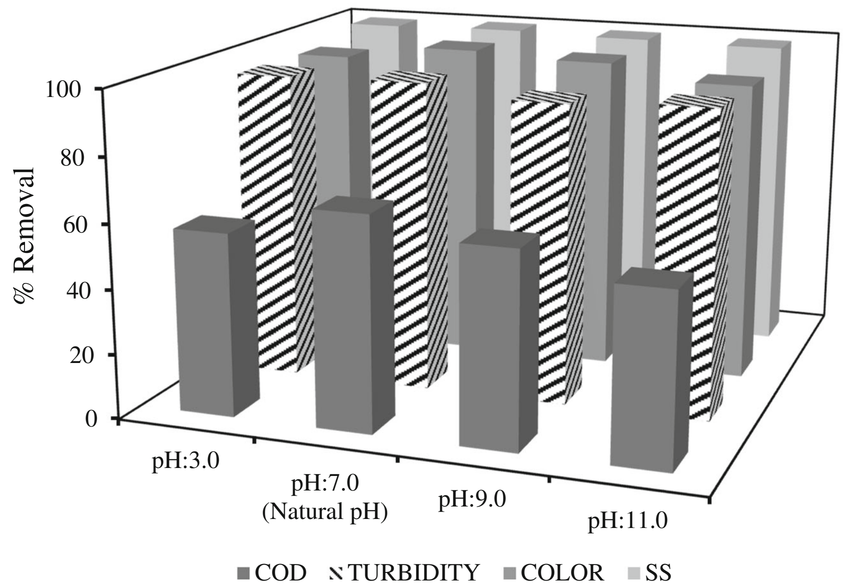
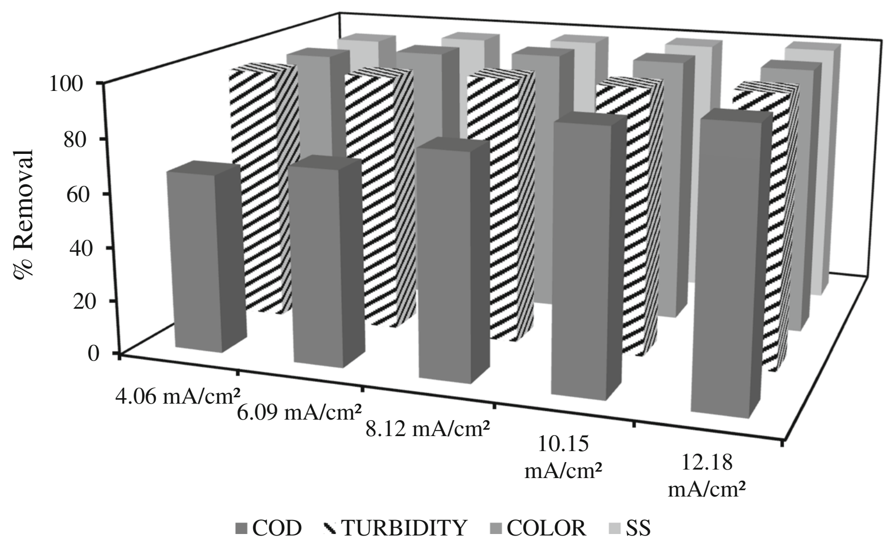


Fig. 6 Effect of current density on COD color, turbidity, and SS removal (400 rpm, natural pH ($\approx$ 7), 0.2 M NaCl)



current density applied increased, the color, turbidity, and suspended solids removal in the wastewater increased, and the time required for these was shortened. These results are attributed to an increase in the rate of generation of oxidants (e.g., OH$^{\cdot}$ radicals and chlorine/ClO^-) at higher current densities [29]. Increasing current density and increasing reaction rate can be attributed to the increase in diffusion rates of chloride species to the anode [30]. It was observed that the results were equally effective for color, turbidity, and flow values.

3.5 Energy Consumption

Energy consumption is one of the important factors for processes. In addition to the removal efficiency, the energy consumed is important in terms of the cost of the process. Energy consumptions for all parameters were found through the studies that were conducted. The energy consumptions in the trials with 1200 mL wastewater are shown in the graphs below.

$$\text{Energy consumption} : i \tag{8}$$

where V (Volt) is the potential difference, I (ampere) is the current intensity, t (hour) is the time, and v (m^3) is SWW the volume.

3.5.1 Effect of Supporting Electrolyte Type on Energy Consumption

Different salts were used at natural pH value, with 400 rpm mixing speed and 0.2 M SE concentration. The results are

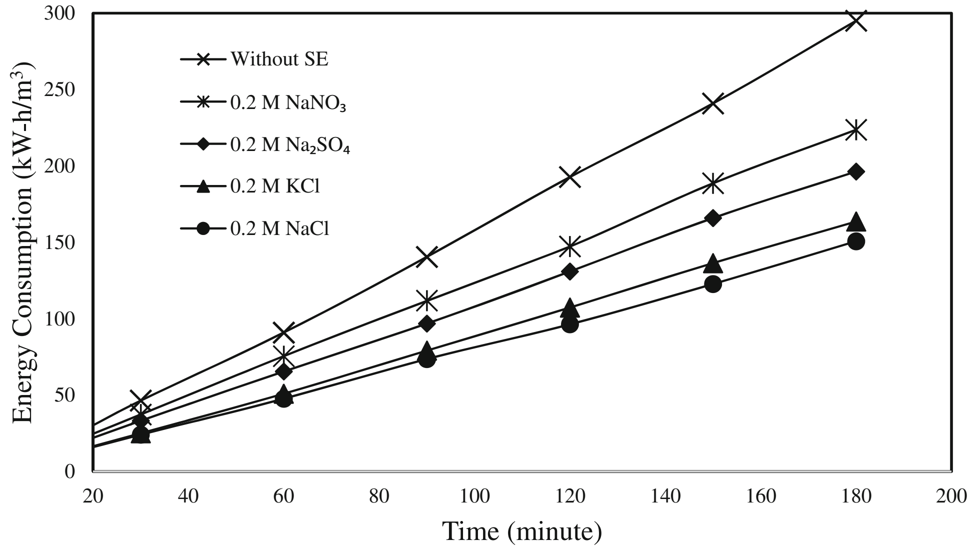


Fig. 7 Effect of SE type on energy consumption (400 rpm, 4.06 mA/cm^2, natural pH ($\approx$ 7), 0.2 M)

shown in Fig. 7. It was observed that the electricity consumption was lower in all trials using SE. The reason for this is that the electrolytes are used to increase the conductivity of the water and thus reduce the energy required for the current passing through the same point. The decrease in the applied potential difference makes the solution electrical with an increase in conductivity. This ensures less energy consumption. According to the energy consumption values, the salts are NaCl < KCl < Na_2SO_4 < $NaNO_3$ < without SE, respectively.

3.5.2 Effect of Supporting Electrolyte Concentration on Energy Consumption

As the SE concentration increases, it is seen that energy consumption decrease. High concentrations of energy consumption are reduced because the conductivity of wastewater increases and the potential difference in the system to generate the same amount of current is reduced. This results in a reduction in energy consumption. The results of the effect of SE concentration on energy consumption are as follows: 295.0 kWh/m^3 245.0 kWh/m^3, 183.6 kWh/m^3, 163.6 kWh/m^3, and 150.2 kWh/m^3 were found at 0 M, 0.1, 0.2, 0.3, 0.4 M concentrations, respectively (Fig. 8). As shown in the figures, the increased concentration of SE reduces energy consumption significantly.

3.5.3 Effect of Initial pH on Energy Consumption

The influence of the pH value of the wastewater in the electrochemical process is reflected both in the utilization of electricity and in the effect of dissolving metal hydroxides [31]. In order to study the effect of wastewater's initial pH value on energy consumption, experiments were carried out at pH 3.0, 7.0, 9.0, 11.0 at the current density of 4.06 mA/cm^2 and NaCl concentration of 0.2 M. The results are shown in Fig. 9. In the results obtained, the results of energy consumption were found as pH 7 > pH 9 > pH 11 > pH 3, respectively. The more ions in ambient conditions, the easier energy transmission will be, so ions should be given to the environment to reduce consumption. Since the conductivity is low in the natural pH value of the wastewater, the consumption has been high. The least energy consumption was observed at pH 3. It is favorable for the oxidation of hydroxyl radicals in acidic conditions.

3.5.4 Effect of Current Density on Energy Consumption

The effect of the applied current density on the energy consumption in the electro-oxidation experiments performed at different current densities was investigated and the results in Fig. 10 are obtained. With the help of Eq. (8) in the third chapter, the energy consumption amounts are calculated. Energy consumptions for current densities of 4.06 mA/cm^2, 6.09 mA/cm^2, 8.12 mA/cm^2, 10.15 mA/cm^2, 12.18 mA/cm^2, 183.6 kWh/m^3, 310.9 kWh /m^3, 529.1 kWh/m^3, 763.6 kWh/m^3, 940.9 kWh/m^3, respectively (Fig. 10). The increase in the current density applied to the system increases the energy consumption along with the voltage increase. According to Faraday's Law, an increase in current density causes a decrease in electrode life [32]. For this reason, it is recommended not to choose a very high current density in order not to reduce the electrode life.

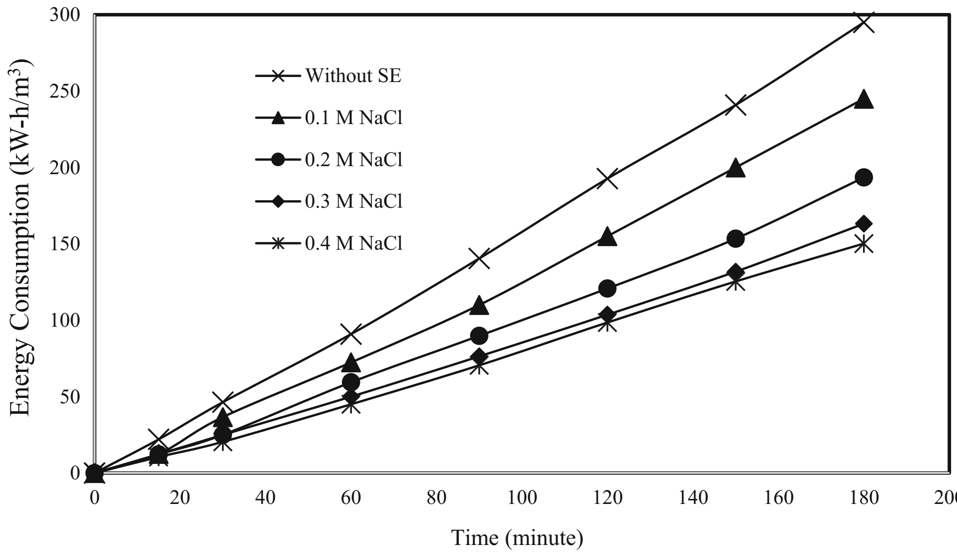


Fig. 8 Effect of SE concentration on energy consumption (400 rpm, 4.06 mA/cm^2, natural pH ($\approx$ 7), NaCl)

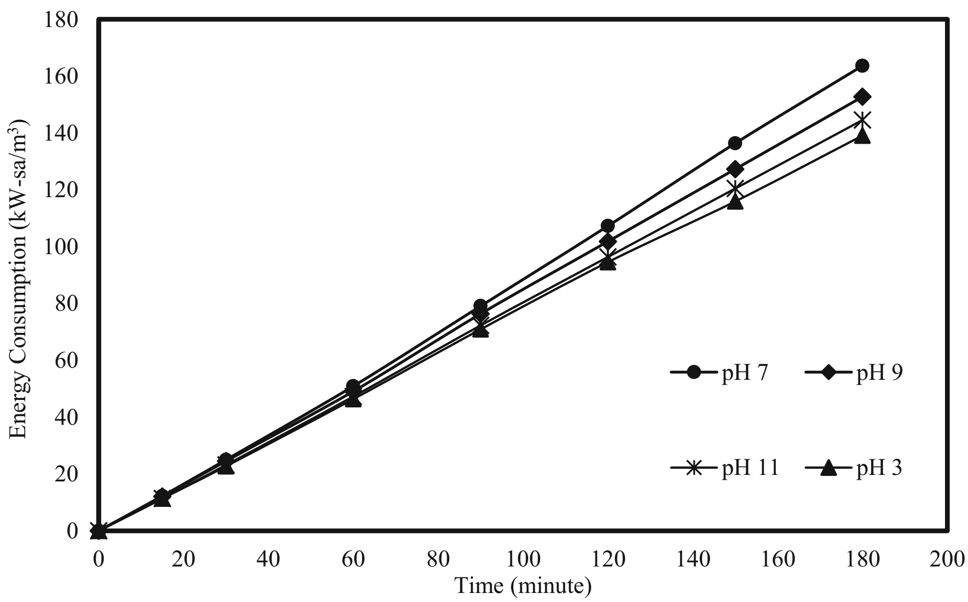


Fig. 9 Effect of initial pH on energy consumption (400 rpm, 4.06 mA/cm^2, 0.2 M NaCl)

4 Discussion

This study aims to investigate the purification of SWW with $Ti/IrO_2/RuO_2$ anode by the electro-oxidation method. Electro-oxidation, which is one of the electrochemical treatment methods, is a very effective method for removing undesirable parameters such as color, turbidity, SS, COD from wastewater. It also has advantages over other methods. For example, it causes less sludge formation after treatment; requires less space; it is easy to operate and use. However, in larger-scale enterprises, the design should be redesigned, and its cost should be considered. In addition to these, the material to be used in the treatment should be cheap and long-lasting, and the conductivity of the water to be treated should be high. In the studies, it was concluded that the most suitable SE was NaCl, the most suitable pH value was the natural pH of the wastewater, and the optimum concentration should be 0.2 M. To reduce energy consumption, the current density was chosen as 4.06 mA/cm^2. In this case, the choice can be made according to electricity cost or efficiency. The electro-oxidation process is one of the most appropriate methods for wastewater with high conductivity. In addition, we can support it with additional treatment processes according to the characteristics of the wastewater. SE contributes to the oxidation process. In the results, it was found that 66.46% of COD was removed and 183.6 kWh/m^3 energy was consumed after a 3-h reaction with 0.2 M NaCl supporting electrolyte from SWW. Electrochemical treatment is recommended for SWW.

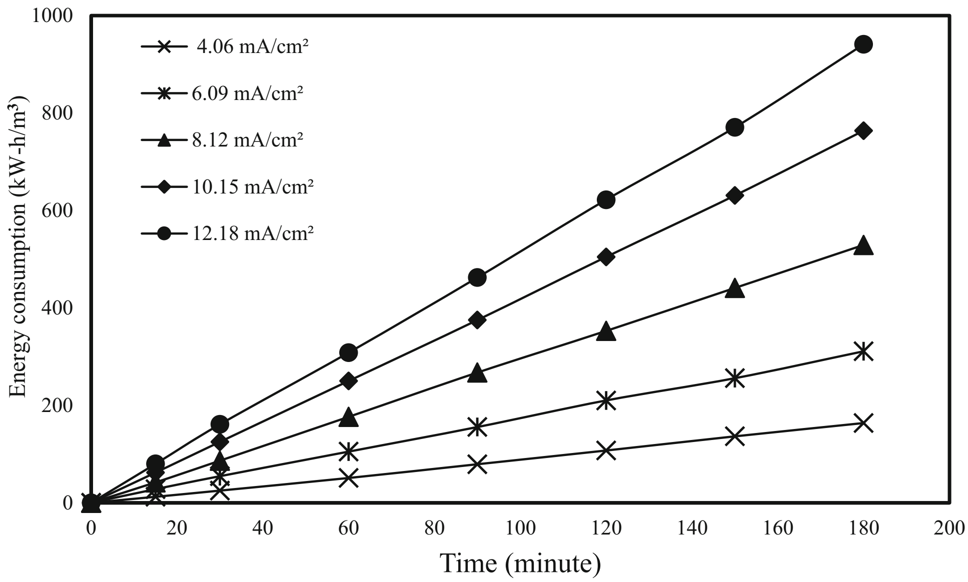


Fig. 10 Effect of current density on energy consumption (400 rpm, natural pH (≈ 7), 0.2 M NaCl)

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Author Contributions Sermin Günaslan (MSc student) conducted all the experiments.
Visualization and editing done by Baybars Ali Fil.

Declarations

Conflict of Interest The authors have no conflicts of interest to declare that are relevant to the content of this article.

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