



ORIGINAL ARTICLE

A comparison between iron and mild steel electrodes for the treatment of highly loaded grey water using an electrocoagulation technique



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Abstract In the last years, the electrocoagulation (EC) process has been widely used as a potential technique for grey water treatment. However, only a few studies have focused on treating highly loaded GW (HLGW) by EC. In this study, the EC technique was used to compare iron and mild steel electrodes for the treatment of HLGW under different current densities (CDs) (5, 10, 15, and 20 mA/cm²) during 10 min of EC time. The performance criteria included chemical oxygen demand (COD) and turbidity removal efficiencies, current efficiency, energy consumption, and operational costs. It was found that EC using iron or mild steel can be effective electrodes for removing high levels of COD and turbidity from HLGW. At optimum conditions, the study demonstrated that at a CD of 5 mA/cm², mild steel-based electrodes reduced COD by 86.5% while iron-based electrodes achieved 85.3% reduction at 10 mA/cm². In conjunction with these removals, the turbidity removals were 92% and 94% achieved by steel and iron electrodes, respectively. The current efficiency of all the conducted experiments exceeded 90% but was generally higher for iron electrodes. At optimum conditions, analysis of operating costs in terms of energy consumptions and electrode materials requirements were 0.054 \$/m³ and 0.097 \$/m³ achieved by steel and iron electrodes, respectively. Thus, mild steel-based electrodes are considered superior to iron electrodes. Based on the obtained results, the study recommends that further investigations should give attention to the effect of metal alloy type or physical properties of electrodes as performance criteria and designing aspects when studying EC technology for HLGW treatment due to its notable effect

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on removal efficiency and operational costs.

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1. Introduction

Grey water (GW) reclamation and recycling became an acceptable alternative as a valuable non-conventional water resource, with a proven impact on reducing demand for freshwater resources (Giresunlu and Baykal, 2016; Mourad et al., 2011; Smith and Bani-Melhem, 2012). GW may originate from various sources, excluding toilets, and thus contains a wide range of chemical and microbial contaminants (Al-Jayyousi, 2003; Eriksson et al., 2002). Depending on sources and level of contamination loads, GW can be categorized as either highly or weakly loaded GW (HLGW or WLGW) (Boyjoo et al., 2013). The highly loaded sources obviously relate to higher levels of health and environmental risks, due to containing higher levels of contaminants (Dixon et al., 1999). With regards to WLGW, simple treatment methods were reported to demonstrate good removal efficiencies and enhancement in quality (Mohamed et al., 2016; Zipf et al., 2016). However, the treatment of HLGW requires more robust systems, such as biological contractors (Eriksson et al., 2009), Photo-catalytic oxidation (Sanchez et al., 2010; Alrousan et al., 2020), and membrane bioreactor systems (Bani-Melhem et al., 2015; Al-Ghazawi et al., 2018). Sometimes, a combination of treatment systems may be required (Bani-Melhem and Smith, 2012; Daghrir et al., 2016; Li et al., 2009). In terms of application fields, grey water treatment can be a good alternative for wastewater reclamation because it can be reused for non-potable applications, such as irrigation, washing, and flushing of toilets, which allows saving up to 75% of household water consumption (Bani-Melhem et al., 2023a).

In recent years, electrocoagulation (EC) technology has received increasing attention in treating different types of wastewater such as the removal of metal ions from beneficiation plant process water (Das and Nandi, 2021), removal of COD and nutrients from municipal wastewater (Al-Qodah et al., 2020; Al-Shannag et al., 2013; Bani-Melhem and Elektrowicz, 2010), and removal of suspended solids from paper industry effluents (Al-Shannag et al., 2012). Various parameters and variables were investigated to improve the performance of this method, including EC time, initial pH, and combining different degradation processes (Nidheesh et al., 2021; Akansha et al., 2020; Al-Qodah et al., 2018). Many efforts had demonstrated great potential for large scaling and industrial implementation (Al-Shannag et al., 2015; Karichappan et al., 2014; Janpoor et al., 2011).

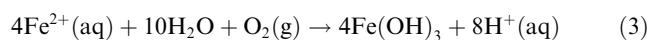
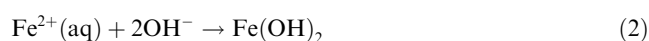
In terms of GW treatment by EC, the process has been conducted by many researchers, and different design and operational parameters were studied to optimize the EC performance. The EC was also integrated with other treatment processes to increase the efficiency of treating highly loaded GW. In a previous study, a sand filtration (SF) unit was integrated with the EC technique as a pre-treatment step to enhance the EC process for treating high-loaded GW (Bani-Melhem et al., 2023a). Three different voltage gradients were investigated (5 V/cm, 10 V/cm, and 15 V/cm) and the results demonstrated that the COD concentration was reduced by 25.5% by the SF step, which allowed a reduction of EC steady state time in the EC unit from 45 min to 30 min at an applied voltage of 15 V/cm. In addition, the energy consumption reduced from 6.21 kWh/m³ without the SF step to 4.11 kWh/m³ after integrating SF with EC. In another study, four different types of metallic wastes (beverage cans, used aluminum (Al) foil, scrap iron, and scrap mild steel) were proposed as sacrificial electrodes for grey water (GW) treatment using the EC technique (Bani-Melhem et al., 2023b). The results demonstrated that using metallic wastes as sacrificial electrodes can achieve a considerable reduction in color, turbidity,

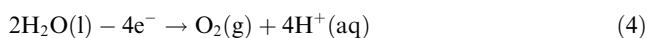
COD, and electric conductivity of about 97.2%, 99%, 88%, and 89%, respectively. In another study, artificial intelligence was used to optimize the EC performance for GW treatment (Nasr et al., 2016).

In order to achieve optimal conditions in EC process, most previous studies had generally modified operational conditions and recorded changes in removal efficiencies and energy consumption. Amongst the frequently investigated parameters in EC technique is the type of electrode material, which is an important parameter as it allows control over the type of coagulating ions released. Theoretically, the type of electrode material (anode) plays an important role in releasing the metallic ions that serve as coagulating species with different adsorption potentials (Yadav et al., 2012). This means that depending on which coagulating particles are presented in wastewater during treatment, the rate of adsorption of pollutants on those particles will differ according to the type of electrodes, resulting in obtaining different removal efficiencies when examining different electrode materials (Barisic & Turkay, 2016). Usually, iron (Fe) and aluminum (Al) are used as electrode materials in the EC process. Some attributes are unique to each material. For example, Al hydroxides may have a higher adsorption capacity and are reported to provide better removal than iron when each was tested independently in various experiments (Daghrir et al., 2016; Yadav et al., 2012; Wang et al., 2003). However, iron electrodes may be considerably cheaper depending on the type of electrode and have shown good removal efficiency for various types of wastewater (Liu et al., 2022; Obi et al., 2022; Ardhan et al., 2014). Some recent experiments had even shown iron electrodes to perform better than aluminum when treating highly loaded wastewater (Potrich et al., 2022), but they do corrode at a faster rate compared to aluminum (Bani-Melhem et al., 2017). While stainless steel electrodes may have less of a risk with regard to potential toxicity (Xu et al., 2002) and have been shown to allow for high removal efficiencies (Karichappan et al., 2014). These types of considerations depend on the type and application desired for the treated effluent, and in some instances, a combination of different types may provide the best result (Barisic & Turkay, 2016). There is a limited ability in identifying the exact limitation and characteristics associated with the exact type and constituents of the electrodes since most of the cited works generally resort to comparing different materials without identifying such details (Boinpally et al., 2023; Potrich et al., 2022; Gasmia et al., 2022).

Despite the impressive amount of scientific research on investigating different electrodes materials, one area that had received no attention in the optimization of EC for GW treatment is the specific type of electrode material such as comparing alloys of the same metal, or alloys with different properties, which is a significant consideration since physical properties such as surface smoothness can influence variables such as bubble generation rate and size (Lakshmi and Sivashanmugam, 2013). This study aims to address such considerations by investigating the treatment of highly loaded grey water (HLGW) using iron and mild steel electrodes. Both of which are iron-based electrodes with highly similar compositions but different physical properties. Both electrodes would generate the same reactions at the anode and cathode sides as follows:

At the anode side (Chen et al., 2004; Kabdashi et al., 2009):

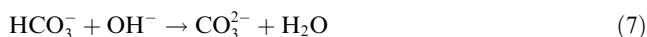




At the cathode side (Mollah et al. 2004; Mollah et al., 2001):



Furthermore, sedimentation of calcium and magnesium carbonates on the cathode of iron and steel electrodes would also take place which forms an inhibiting layer via the following reactions (Ge et al., 2004; Janpoor et al., 2011):



Layer formation may differ between iron and steel electrodes, due to the presence of Mn in mild steel, which once released to the solution, it may not necessarily be fully removed by Fe(II) and Fe(III) species but could precipitate on cathodes (Shafaei et al., 2010).

In this study, iron and mild steel electrodes will be examined under different current densities (CD). These electrode materials will be evaluated based on their ability to reduce COD and turbidity. Variation between the electrodes will be determined and attributed by recording chemical and physical changes in the treated solution to observe if there was a difference in dominant reactions between electrode types. Current efficiency will also be compared for both electrodes to determine if there is a variation in how electrode materials are affected by pitting corrosion and side reactions which will be based on proper corrosion calculation methods. Finally, economic aspects will be addressed based on electrical and material consumption at optimum conditions for both materials.

2. Materials and methods

2.1. Grey water collection and sampling criteria

For the purpose of this study, GW samples with relatively high loads of COD were obtained by conducting source characterization prior to collecting samples for treatment. Samples were collected from all GW sources to exclude those that contributed to low levels of COD from treatment (Table 1). The threshold for COD was set at 250 mg/l, meaning that sources which demonstrated COD concentrations below this value would not be included in the treatment experiments. This limit (250 mg/l) was chosen as it was the Jordanian standard COD limit for safe reuse, suggesting that sources with COD levels below this value may need simple treatment and do not serve the purpose of this study.

Sample collection was carried out within faculty and administrative buildings at the Hashemite University, Jordan. Sources that were examined include 1) sinks in students' bath-

rooms, 2) sinks in buffet and staff facilities, 3) laboratory sinks, 4) ablution basins, and 5) floor mopping carts. The sampling campaign included both academic and administrative departments, facilities, and centers from 14 different buildings. Grey water samples were collected directly from their respective sources. This was due to the lack of a separation system for grey and black water in the study area. Sampling was done around noon time to ensure collection of HLGW generated in the same day of sampling. Analysis and treatment experiments were done within 48 h of the sample's arrival at the lab.

2.2. Experimental setup and procedure

The experimental procedure reported in this study was conducted using a bench scale batch electrocoagulation (EC) unit (Fig. 1). A cylindrical glass beaker (300 ml) was used as an EC reactor and was replaced in each run to eliminate the interference of residual coagulants in consecutive experiments. This was based on a preliminary test showing that metallic residues from electrochemical experiments were not easily removed from glassware by washing. A 250 ml of HLGW solution was treated during each run. Metal electrodes were placed vertically in parallel within the EC reactor.

Two types of metal electrodes were compared in this study: iron and mild steel electrodes. Each material was used separately as both anode and cathode for treatment (Iron - Iron or Steel - Steel). The total surface area of electrodes was about 22.72 cm² (height: 71 mm, width: 32 mm), while the effective surface area in contact with GW solution was 16 cm². The electrodes were soaked in diluted sulfuric acid for 24 h prior to usage to remove oxidation films. The distance between the electrodes was fixed at 1 cm in the EC reactor. A direct current (DC) power supply (Maisen™, model MS303D, China) was used to provide a voltage range of 0–30 V and a direct current range of 0–3 A. The EC reactor was placed over a magnetic stirrer (VELP Scientifica, Italy) to allow homogeneous agglomeration in the HLGW solution during electrolysis. The stirring speed was set at 180 rpm. The electrolysis time was 10 min for all experiments, and samples were given

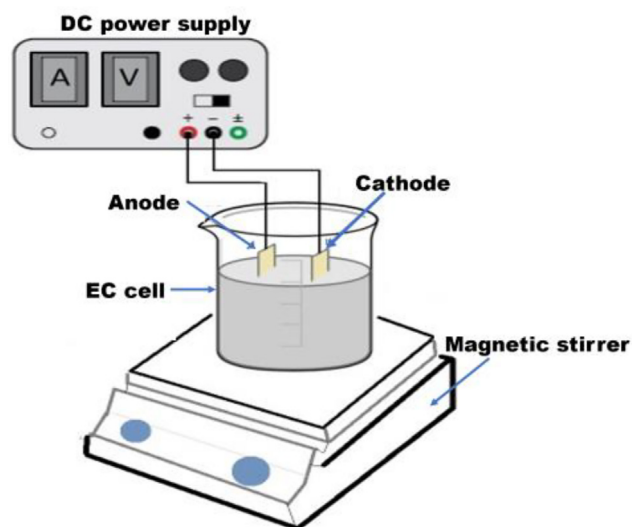


Fig. 1 Schematic of the experimental setup used in this study.

Table 1 Stages of characterization and sampling of highly loaded grey water.

1 st stage	2 nd stage	3 rd stage
General characterization of grey water sources in the study area	Identifying highly and weakly loaded sources of greywater	Collecting fresh samples from highly loaded sources for treatment

2 min for settling after turning off the power supply. The experiments were conducted under different CDs ($5 - 20 \text{ mA/cm}^2$) by fixing the applied current to the following values: 0.08, 0.16, 0.24 and 0.32 A, while voltage was adjusted during each experiment to maintain the desirable current. The operational parameters of the EC reactor are presented in Table 2. The actual dissolution rate (mg/l) was determined as the difference between the weights of electrodes before and after treatment divided by sample volume. The electrodes were rinsed with distilled water and dried by wiping gently with paper tissues and left to dry for approximately 1 h before taking weight readings. Several readings (3–7) were taken for each experiment to verify the reading precision of the laboratory scale.

2.3. Analytical procedures

Several key parameters were measured for HLGW samples before and after EC treatment to estimate removal performance and chemical changes. COD was measured using Spectrophotometer photoLab® 7600 UV-VIS (WTW, Germany), a bench-scale laboratory equipment commonly used for water analysis, which was calibrated at least twice a day. Turbidity was measured using an MD600 photometer (Lovibond, Germany). Electrical conductivity and pH were measured using a pH electrical conductivity meter (Hanna HI 5521, USA), which was calibrated before usage. Turbidity and COD were selected as they are amongst the most widely used parameters in treated wastewater reuse standards. Nutrient concentrations were not analyzed in this study because no specific reuse criteria were considered.

2.4. Description of electrode materials

Electrodes used in this study were provided by the workshop at the Hashemite University. Iron electrodes were pure Fe plates, while steel electrodes were made from low carbon mild steel plates which contained 98.81–99.26% Fe, 0.18% C, 0.6–0.9% Mn, and may have contained traces of P and S (0.04–0.05%). The presence of these constituents, particularly carbon, at these percentages, gives mild steel its distinct mechanical properties. Composition and physical properties of the electrodes used are presented in Table 3.

3. Results and discussion

3.1. Characteristics of raw grey water

GW samples used for treatment in this study were collected from different sources that highly contributed to COD and

Table 3 Physical properties and composition of electrodes tested in this study (based on manufacturer specs).

Electrode type	Iron	Steel
Material type	Pure	Alloy (mild steel)
Elemental composition	Fe	Fe, Mn, C
Electric conductivity (IACS)	18 %	10.84 %
Thickness of material	0.8 mm	0.74 mm
Shape	Plate	Plate
Surface smoothness	Moderately smooth	Very smooth
Roughness coefficient	$\sim 0.25 (10^{-3} \text{ m})$	$\sim 0.045 (10^{-3} \text{ m})$

suspended solids, resulting in obtaining HLGW with high contamination loads. A comparison between the characteristics of raw grey water used in this study and the corresponding values reported for GW in other studies is shown in Table 4. In addition to the proposed studied parameters, the table contains key parameters of GW such as electrical conductivity, biological oxygen demand (BOD), and total dissolved solids (TDS). Sources of GW in this study were found to contribute to the highest loads of COD and thus used for treatment experiments, including bathroom sinks and floor-mopping carts. Table 4 Also contains the concentrations of soluble COD (CODs) as it plays a significant role in GW treatment by EC (Bani-Melhem et al., 2017). Based on analytical results, it can be reasonably assumed that samples collected in this study can be categorized as HLGW (Boyjoo et al., 2013).

3.2. Changes in physiochemical proprieties

3.2.1. Changes in pH, conductivity, and current stability

The initial pH level of the collected samples in this study was found around optimal values reported by previous investigators who had studied the effect of initial pH on EC treatment of HLGW (Janpoor et al., 2011; Barisci and Turkay, 2016). Accordingly, the initial pH was not adjusted in this study. However, the pH was monitored because it is an important parameter in indicating dominant reactions, and thus can be used for comparing two electrodes with very similar compositions. It is worth mentioning that the range of applying CD in EC reactor may have a direct impact on the pH of the treated solution by releasing the dominant ions in terms of hydrogen ions (H^+) or hydroxyl ions (OH^-) according to the domination of the anodic and cathodic reactions occurred on the electrodes in EC reactor. Therefore, final pH readings of the treated solution were compared against initial pH levels (Fig. 2), by measuring pH after shutting down the power supply.

Table 2 Operational parameters of electrocoagulation reactor used in this study.

Parameter	Values chosen or tested			
Applied current (A)	0.08	0.16	0.24	0.32
Current density (mA/cm^2)	5	10	15	20
Voltage range (V)	4.5 – 14.5 (adjusted during electrolysis)			
Electrode materials	Iron, Mild steel			
Electrode distance (cm)	1			
Sample volume (ml)	250			

Table 4 Characteristics of raw grey water used in this study and corresponding values reported for grey water in other studies.

Quality Index	Unit of measure	This study	Range reported for grey water by other studies *	
			Min	Max
pH	–	7.8	5.2	10.2
Electrical conductivity	$\mu\text{S}/\text{cm}$	1335	194	650
COD	mg/l	1290	22.9	1307
CODs	mg/l	516	27	873.12
BOD	mg/l	730	10	1056
Turbidity	FAU	506	12.6	370
TDS	ppm	900	180	650

*Sources: Kim et al., 2007; Lin et al., 2005; Bani-Melhem & Smith, 2012.

Based on whether the final pH reading was over or below initial levels; it was determined which reactions were dominant for each electrode type at each CD (Kurt et al., 2008).

Two observations can be drawn from Fig. 2. First, it was observed that the change in pH levels was not significant, which is consistent with previous investigators (Janpoor et al., 2011; Barisci and Turkay, 2016), and this might be due to the relatively short EC time applied in this study. Second, the general trend for both examined electrodes was that at the lower CD range ($5 - 10 \text{ mA}/\text{cm}^2$), pH in the treated solution was below initial values. The pH reduction suggests that the production of H^+ ions (Eqs. (3) and (4)) exceeded the production of OH^- ions (Eq. (6)). On the other hand, at a higher CD range ($15 - 20 \text{ mA}/\text{cm}^2$), it was observed that final pH

readings were higher than initial pH levels. This suggests that at this CD range, OH^- production was higher.

Based on the results presented, it can be concluded that when either type of electrode was used, the reactions that took place at the anode side (Eqs. 1 – 4) were likely more dominant at the lower CD range ($\leq 10 \text{ mA}/\text{cm}^2$), while at higher CD range ($\geq 10 \text{ mA}/\text{cm}^2$) the reactions at the cathode side (Eqs. (5) and (6)) were likely dominant. Also, it was observed that pH change was more severe when using mild steel electrodes; as the treated solution produced after using mild steel electrodes was more acidic at a lower CD range and more basic at a higher CD range than those of iron electrodes, suggesting that current as a driving force for these reactions has more effect when using mild steel electrodes than when using iron

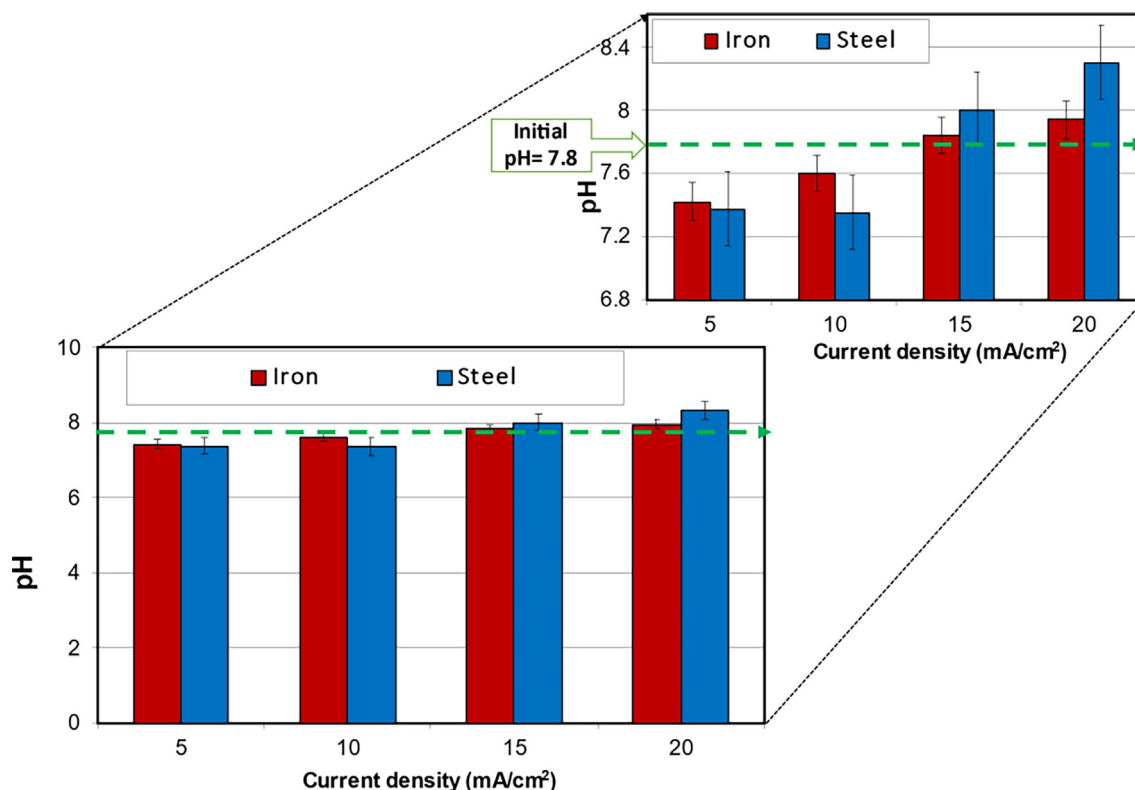


Fig. 2 Evolution of pH levels in treated GW solution using iron and mild steel electrodes at different current densities.

electrodes. The change in dominant reactions when CD shifts from a lower to a higher range is caused by an increase in voltage to over 10 V to elevate CD beyond 10 mA/cm² (Fig. 3). This likely caused a breakdown of inorganic deposition on the cathode (Eq. (5), Eqs. (7) – (9)) and thus layer formed by precipitating salts could no longer obstruct cathodic reactions, which had led to higher generation rate of OH⁻ ions (Janpoor et al., 2011).

With respect to the chemical changes, electrical conductivity was also monitored. As demonstrated in Fig. 4, Mild steel electrodes were significantly more efficient in reducing electric conductivity than iron electrodes at all values of the applied CD. It was also observed that under a low CD range, iron-based electrodes had almost no effect on reducing electrical conductivity. The higher reduction in electric conductivity obtained when using mild steel electrodes could be related to the release of alloying elements and more precipitation on the cathode surface area, and could possibly relate to differences in surface smoothness, which would cause different bubble formation patterns (Barıscı & Turkay, 2016; Lakshmi and Sivashanmugam, 2013). Theoretically, the release of ions of alloying elements from mild steel could have also further contributed to TDS in HLGW solution which would increase electrical conductivity. It is also possible that all these effects have taken place at different capacities. These observations strongly suggest that future works examining physical characteristics of electrode materials should be investigated thorough chemical analysis beyond those employed in this study.

The poor reduction of electrical conductivity by iron electrodes had likely played a role in showing lower voltage requirement for iron electrodes than that of mild steel electrodes (Fig. 3). This might be explained as the higher electrical conductivity of HLGW solution when using iron electrodes caused a lower resistivity-induced over potential (IR) drop which allowed for a lower ohmic loss in electrical current (Karichappan et al., 2014). This effect can be better explained by the following formula (Mollah et al., 2004):

$$\eta_{AP} = \eta_{IR} + \eta_k + \eta_{Ml} \quad (10)$$

Where η_{AP} is the voltage applied by the power supply, η_{IR} is the resistivity-induced over potential (known as the IR drop), η_k is the activation (kinetic) potential and η_{Ml} is the

mass transfer over potential. It seems that the ability of electrode material to reduce electric conductivity of the solution is the primary effect on voltage; because there is strong evidence from plotting of current–voltage behavior in Fig. 5 which shows that both electrodes initiate reaction at identical voltage values at lower CD and shows very similar voltage requirement at high CD. This suggests that variation in electric conductivity of the electrodes' materials had likely not reflected difference in voltage requirement, and that attention should be given to other electrode properties which may had likely affected conductivity of the solution.

The aesthetic appearance is another indication of EC performance (Bani-Melhem and Elektorowicz, 2011). Flotation and sedimentation effects were rapidly observed in this study; sludge was separated from the treated solution within less than two minutes of shutting down the power supply (Fig. 6).

The sludge produced in this study was observed to settle down to the bottom of the EC reactor, indicating high density of sludge obtained when using iron or mild steel electrodes. There was a notable difference in color of the precipitating species in samples collected from EC reactor after treatment (Fig. 7). When iron-based electrodes were used, a yellow-reddish precipitate appeared in settled samples, while a dark brownish precipitate was produced after using mild steel.

The yellow-reddish color produced by using iron-based electrodes is an indicator of residual Fe(III) species, which has oxides characterized by a reddish rusty color (Bani-Melhem, 2008; Adhoum and Monser, 2004). While dark brownish color produced by using mild steel electrodes can associate with oxides of Fe(II) and/or oxides of Mn(III) and Mn(IV) species, which is accounted by the content of Fe and Mn in mild steel electrodes and their electrochemical release in the solution (Flenner, 2007). However, it is suggested that Fe(II) species were mostly responsible for the dark color appearing after using mild steel electrodes due to the very low fraction of Mn in mild steel, and the high tendency of Mn ions to be trapped and removed by sweep coagulation, or precipitate on cathodes (Shafaei et al., 2010). Other trace elements in mild steel (P, S, and C) were likely removed by coagulation and had likely not affected the process (Damaraju et al., 2017; Behbahani et al., 2011; Murugananthan et al., 2004), sulfur could have impacted pH

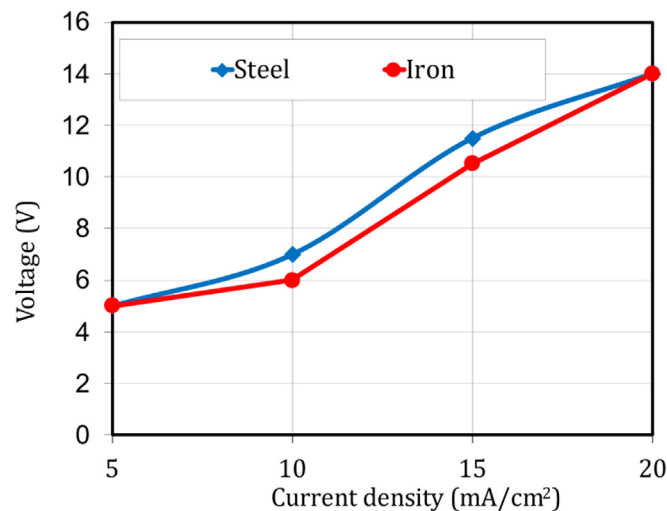


Fig. 3 Voltage required to maintain desired current density when using iron and mild steel electrodes.

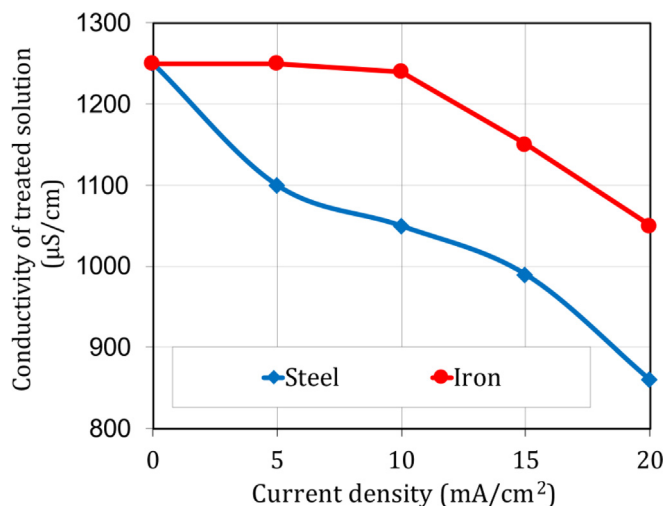


Fig. 4 Electrical conductivity of treated solution as a function of current density when using iron and mild steel electrodes.

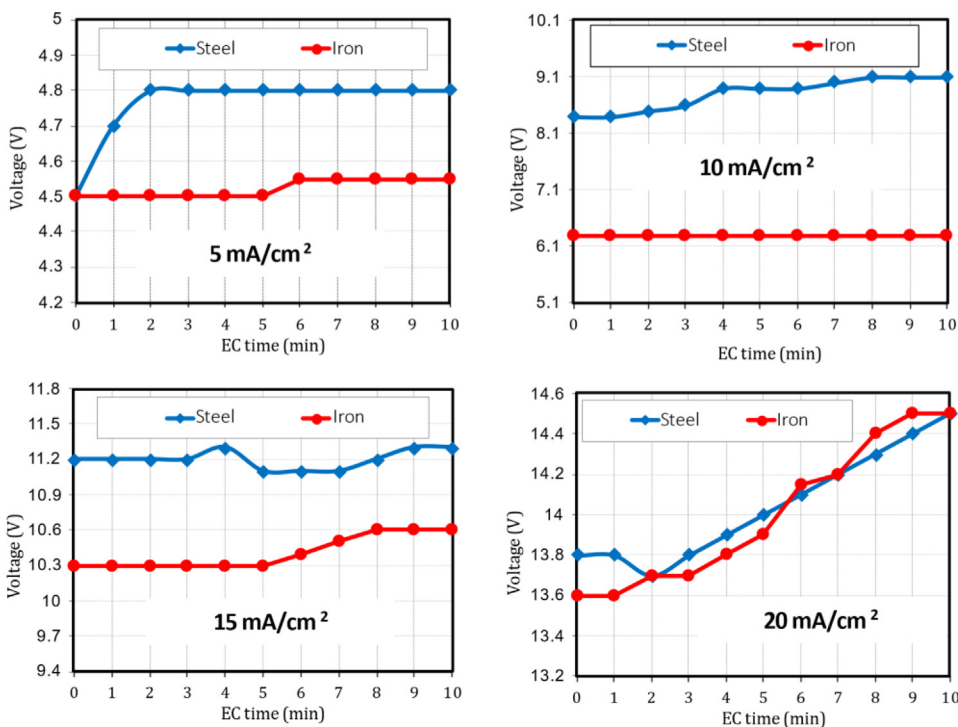


Fig. 5 Current-voltage behavior during electrolysis at different current densities.

if it was at higher concentrations. These observations indicate that when using iron electrodes, Fe(II) is oxidized to form Fe (III), which would also contribute to removal effects (Kabdash et al., 2009), while such oxidation effect may not necessarily have taken place when mild steel electrodes were used. A variation in the type of Fe ions produced at each electrode type would also account for variation in the severity of pH change (Fig. 2) as Fe(II) and Fe(III) ions bond with different numbers of hydroxyl ions (Kurt et al. 2008). The explanations presented are only speculative, an examination of the oxidation states of metallic ions would be required to obtain more certainty. These observations, however, clearly show that different pro-

cesses can take place when using electrodes of almost identical composition, suggesting that the mechanical properties of electrodes can impact the chemistry of the EC process.

3.2.2. Removal efficiency

In this study, COD removal was used to indicate the removal efficiency of organic and oxidizable species, while turbidity removal was used to indicate the removal of colloidal and suspended particles. The following formula was used to calculate removal efficiency (R%):

$$R\% = 100 \times \left(1 - \frac{C}{C_0}\right) \quad (11)$$

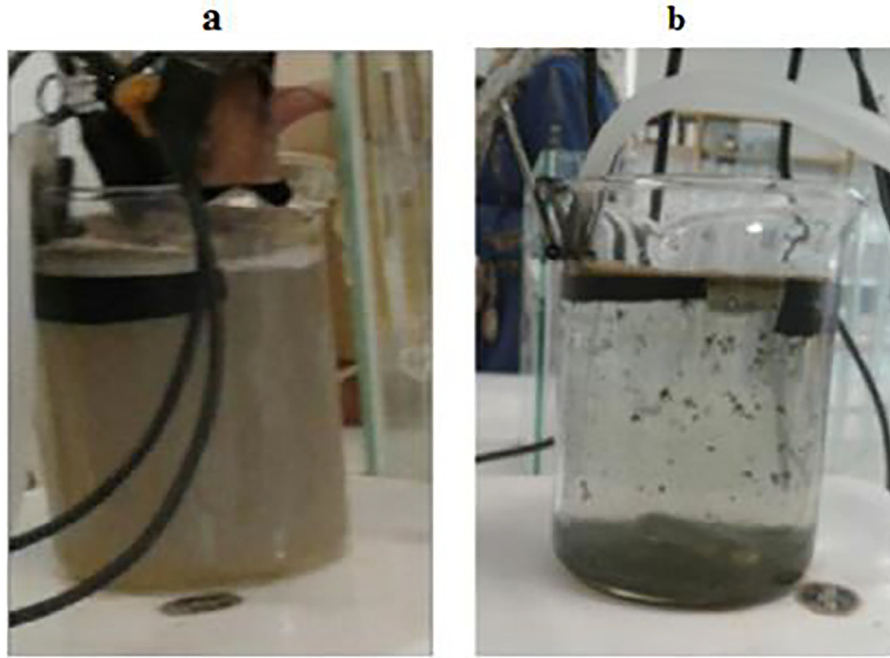


Fig. 6 Grey water solution in EC reactor using steel electrodes at 20 mA/cm²: (a) after 10 min of EC treatment, and (b) after two minutes of settling after EC shut down.

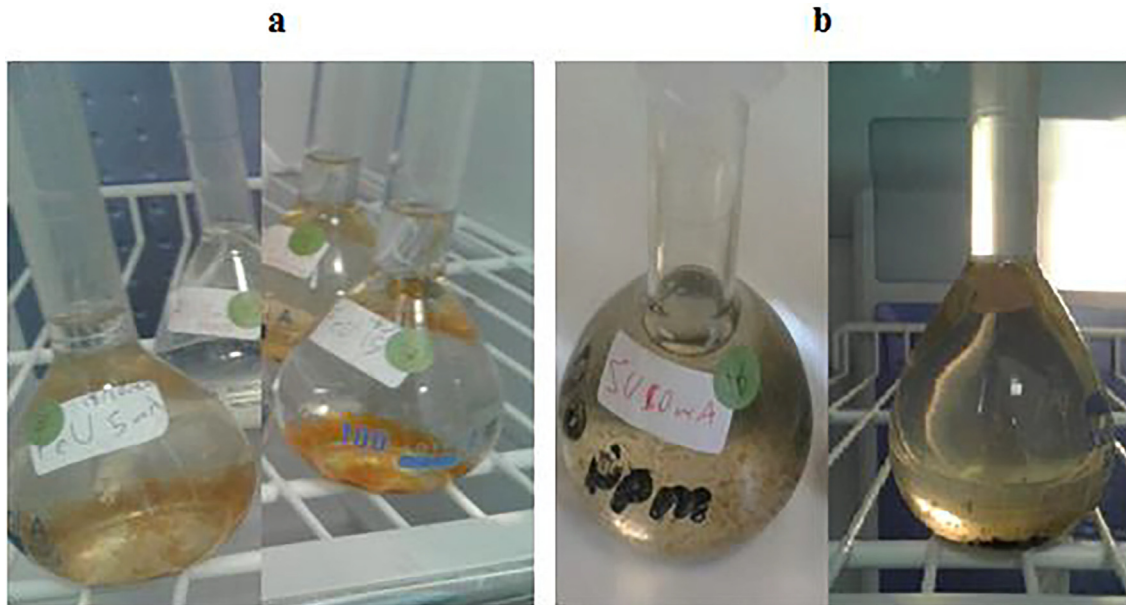


Fig. 7 Aesthetic appearance of color in samples collected from EC cells: (a) using iron-based electrodes and (b) steel-based electrodes.

Where C_0 is the initial concentration of COD (mg/l) or Turbidity (FAU), and C is the value for the corresponding parameter found in the treated solution. While specific energy consumption (SEC) was also taken into consideration for choosing the optimal CD. The following formula was used to calculate SEC:

$$\text{SEC} \left(\frac{\text{kWh}}{\text{m}^3} \right) = \frac{U_{\text{Avr}} \times I \times t_h \times 1000}{v} \quad (12)$$

$$U_{\text{Avr}}(\text{V}) = \frac{\sum_{i=0}^{10} U_i}{10} \quad (13)$$

Where U_{Avr} is the average voltage (V), I is the applied current (A), t_h is EC time (h), v is the volume of the treated sample (m³) and U_i is voltage (V) recorded for the i^{th} minute during EC process.

As demonstrated in Fig. 8, the EC process using mild steel-based electrodes was almost not affected by applied CD with

regards to COD removal, demonstrating a percentage removal between 86.5% and 87.5% regardless of the applied CD. This was not the case with iron electrodes, which showed an increase in COD removal efficiency from 81.5% to 85.3% when the applied CD increased from 5 to 10 mA/cm², respectively. However, when the applied CD was increased beyond this level, COD removal efficiency decreased to 84.5% and 77% for the applied CD of 15 and 20 mA/cm², respectively. It is assumed that soluble COD fraction was responsible for not obtaining complete removal (Bani-Melhem et al., 2017), because soluble and colloidal organics are not readily removed by the coagulation process. However, the evolution of hydroxyl radicals during electrolysis can account for some removal of the soluble organic fraction (Aswathy et al. 2016). Larger organic particles comprised the larger fraction of COD in the samples treated in this study (Table 4), which can explain the relatively high removal efficiency.

The significant difference in removal behavior between the two types of electrodes clearly suggests that different mechanisms had taken place for each electrode material. When using mild steel-based electrodes, it seems that the lower level of applied CD was sufficient to remove almost all organic and oxidizable species that would react with Fe(II) and Fe(III) species, and so increasing CD had no enhancing effect on COD removal. On the other hand, when using iron-based electrodes, it seems that elevating CD to a higher level decreased COD removal. Due to both materials having a similar chemical composition, this may only be explained by differences in physical characteristics. One possible explanation for reduced removal efficiency with increased current is unfavorable bubble production (Eq.6) at a high CD range that may have destroyed flocs formed and thus reduced removal efficiency (Barisci & Turkay, 2016). Also, unfavorable bubble formations due to increased current would reduce the likeliness of contact between coagulating ions and colloidal species present in the HLGW solution (Lakshmi and Sivashanmugam, 2013). Favorable bubble formation during electrolysis is highly related to surface smoothness; finer surfaces produce finer bubbles at better rates. Mild steel plates used in this study are generally designed to have a smoother surface than iron. A smoother surface would

account for better bubble production and thus would not have negatively affected removal efficiency when CD was increased. Since no experimental tests or analyses have been done on bubble formation in this study, other factors could possibly be the reason behind the differences in the obtained removals. This would highlight the need to examine this parameter within this scope (i.e., comparing alloys of the same material in the EC process) in future research works.

With regards to the removal of turbidity (Fig. 9), it was noted that increasing CD had generally enhanced turbidity removal for both electrode types. This would be expected since elevating CD increases the production of Fe(II) and Fe(III) species which effectively aggregate colloidal particles, causing them to be readily separated from the solution (Kurt et al., 2008; Lin et al., 2005). Also, the higher generation of H₂ gas at the cathodes (Eq. (6) when CD is increased favors the sweeping effect of large particles and their eventual separation from the bulk of the HLGW solution.

The results demonstrated that complete removal of turbidity (100%) can be achieved by EC treatment using either iron or mild steel electrodes if the CD is elevated sufficiently. This would confirm that soluble particles were in fact the limiting factor for COD removal. This is based on the fact that turbidity, which is a function of colloidal and suspended particles, was completely removed at high CD while total removal of COD could not be achieved due to the soluble fraction of COD in the grey water solution (Bani-Melhem et al., 2017). In general, mild steel electrodes demonstrated better removal of turbidity than iron electrodes. This may also indicate better bubble production or higher effectiveness of ferrous hydroxides produced. The higher removal efficiencies obtained when using mild steel electrodes may not be attributed to alloying elements, as there is no source in the literature that indicates or suggests that carbon or manganese have coagulating properties or any effects that would enhance removal efficiency during electrochemical treatment of wastewater, or any other type of water for that matter. In fact, manganese could precipitate on cathodes and contribute to layer formation which would negatively affect EC performance. However, it is more likely to be removed by sweep coagulation due to its physical nature

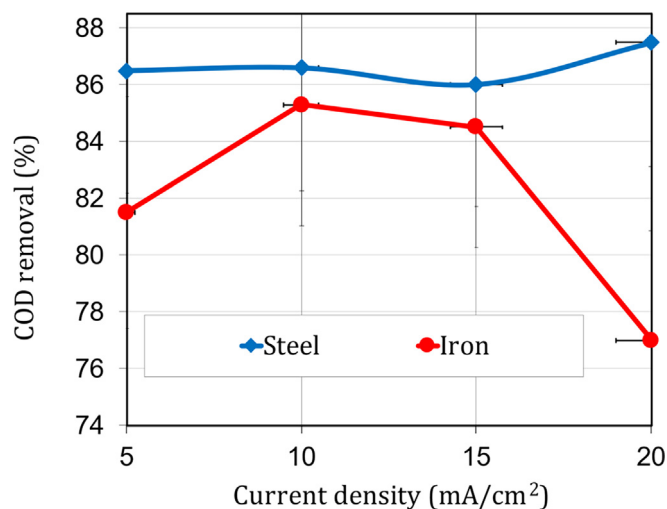


Fig. 8 Effect of current density on COD removal using iron and steel electrodes.

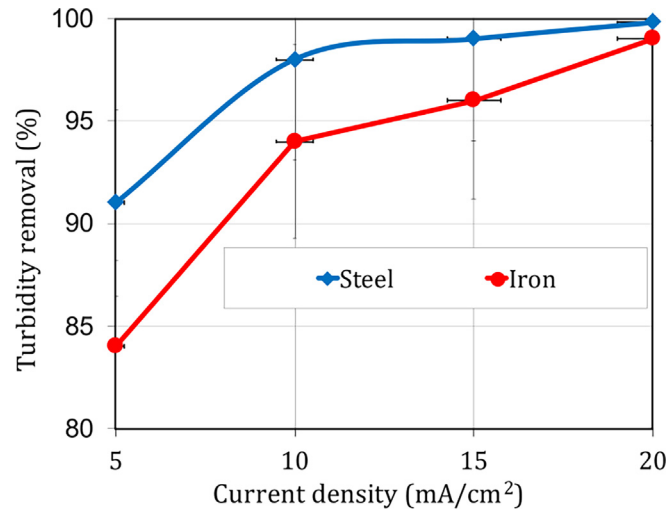


Fig. 9 Effect of current density on turbidity removal using iron and steel electrodes.

(Shafaei et al., 2010). This is further evidence that mechanical or physical properties of mild steel were the dominant factor behind obtaining better removal efficiencies than that of iron electrodes.

Based on the above results, a CD value of 5 mA/cm² was considered optimal for mild steel electrodes as 86.5% COD removal and 92% turbidity removal can be achieved at this level. Otherwise, elevating CD beyond this value had no considerable effect on COD removal while accounting for substantially higher energy consumption (Fig. 10). On the other hand, for iron-based electrodes, a CD value of 10 mA/cm² was considered optimal as 85.3% COD removal efficiency can be achieved in conjunction with 94% turbidity removal.

It is worth mentioning that both CD values chosen for electrodes demonstrated considerable removal efficiency for turbidity, which could also be further improved by primary or subsequent treatment stages, which is not necessarily the case for COD removal efficiency, that is why COD removal efficiency was considered as the main factor for choosing optimum CD.

3.2.3. Metal dissolution and current efficiency

During EC treatment, the separation of contaminants is affected by the coagulating species such as Fe(OH)₂ and Fe(OH)₃ which are produced by the passivation of electrodes (Eqs. (2) and (3)) (Chen et al., 2000; Kurt et al., 2008; Vasudevan et al., 2009). Therefore, it is important to measure the dissolution rate for both electrodes examined to conclusively arrive at which factors affected removal, and also to estimate operational costs. The dissolution rate was represented as the mass loss due to electrolysis with accordance to the following formula:

$$C_{\text{measured dissolution}} (\text{mg/l}) = \frac{(W_0 - W_f) \times 1000}{v} \quad (14)$$

Where W_0 is the initial weight of the electrode prior to electrolysis (g), W_f is the electrode's weight after electrolysis (g) and v is the volume of treated HLGW (l). Furthermore, to establish a benchmark for both electrodes, theoretical dissolution was also calculated. The classic form of Faraday's formula was used to determine the theoretical dissolution for iron electrodes as follows:

$$C_{\text{theoretical dissolution}} (\text{mg/l}) = \frac{I \times t \times M_w \times 1000}{z \times F \times v} \quad (15)$$

Where I is applied current value (A), t is EC time (s) M_w is the molecular weight of Fe (55.85 g/mol), z is the valence of Fe (2), F is Faraday's constant (96485C/mole) and v is the volume of HLGW solution (l). While for mild steel electrodes, standard practice G 102 (ASTM, 1999) was adopted to calculate theoretical dissolution. This procedure is based on Faraday's law; only in that it allows for the calculation of theoretical corrosion rate for anode materials in electrochemical cells while accounting for the existence of multiple metals in alloy electrodes. It should be noted that this procedure does not account for non-metallic compositions such as carbon. However, this was not problematic due to the insignificant amount of carbon in mild steel. The practice also does not account for oxidation selectivity, meaning that it does not reflect if a certain component of the alloy is being oxidized at a higher rate, nor if certain components are not oxidized at all. Also, due to Mn exhibiting multiple valence values, as it is a transitional metal, the authors of the practice recommend adopting some rationale in assigning valence values.

Standard practice G 102 suggests the following formula for calculating the equivalent weight (EW) for metal alloys:

$$EW = \frac{1}{\sum \frac{n_i \cdot f_i}{W_i}} \quad (16)$$

Where f_i is the mass fraction of the i^{th} element in the alloy, n_i is the valence of the i^{th} element of the alloy and W_i is the atomic weight of the i^{th} element in the alloy. In standard practice G 102, a formula is used to calculate corrosion in terms of mass loss rate (g/m².d) or in terms of penetration rate (mm/yr) by taking into account the CD and material density. However, because the actual dissolution measured in this study is given in terms of mg/l (mg of material loss per liter of solution treated), the equivalent weight formula (Eq.16) was directly incorporated in Faraday's law formula (Eq.15) to present theoretical dissolution rate values in terms of (mg/l) to allow easier calculation of current efficiency for mild steel electrodes and to better compare it value with that of iron. The final formula adopted for calculating theoretical dissolution for mild steel anode is as follows:

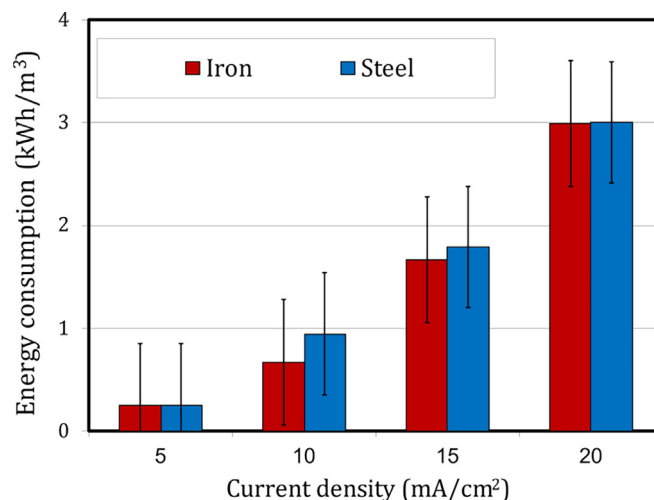


Fig. 10 Effect of current density on energy consumption when using iron and steel electrodes.

Table 5 Theoretical and actual dissolution for iron and steel electrodes at different current densities.

CD	Dissolution amount for Fe based electrodes (mg/l)			
	TD-Iron *	AD-Iron *	TD-Steel *	AD-Steel *
5 mA/cm ²	58.55	60	58.063	60.4
10 mA/cm ²	117.10	111.6	116.127	108
15 mA/cm ²	175.65	165.6	174.19	161.6
20 mA/cm ²	234.20	220.8	232.253	215.2

*TD: theoretical dissolution calculated, AD: actual dissolution measured.

$$C_{\text{theoretical dissolution}}(\text{mg/l}) = \frac{I \times t \times 1000}{F \times V} \times \sum \frac{Wi}{ni \times fi} \quad (17)$$

The current efficiency (C_E) was used as a performance indicator for both electrodes, as it is helpful to identify if there was any variation in the utilization efficiency of current with regard to metallic dosing between the two electrodes. The current efficiency was calculated using the following formula:

$$C_E(\%) = \frac{C_{\text{measured dissolution}}(\text{mg/l})}{C_{\text{theoretical dissolution}}(\text{mg/l})} \times 100\% \quad (18)$$

The results of theoretical calculations and measures of mass change for both electrodes examined are presented in Table 5.

As demonstrated, the dissolution rate for iron electrodes was slightly higher than that of mild steel electrodes. This was somewhat consistent with theoretical calculations that show mild steel to theoretically produce a lower metal dosage during electrolysis, mainly due to the lower molecular weight of Mn and the presence of carbon in mild steel. Such differences could also be a result of experimental error.

Assigning different valence values for Mn had no considerable effect on the theoretical dissolution rates calculated (Fig. 11), and so a valence value of 7 was adopted for Mn when presenting a comparison (Table 5). The lower dissolution rate of mild steel is further evidence that it was the physical and mechanical properties that allowed for better removal efficiencies, since the coagulating dosage released was higher for iron.

The results indicate that super faradaic efficiencies can be obtained using either iron or mild steel electrodes at low CD (5 mA/cm²), while at higher CD, the current efficiency is reduced to below 100% (Fig. 12).

This suggests that pitting corrosion, which increases dissolution beyond theoretical values, was more dominant at minimal CD, while side reactions that limit dissolution rate were more dominant at higher CD for either type of electrodes (Chen et al. 2000; Mouedhen et al., 2008; Shen et al., 2003). The results presented in this section highlight the importance of optimization efforts that aim to reduce CD required when using Fe-based electrodes such as iron and mild steel for the treatment of HLGW using EC, as it seems that current is not only a driving factor of energy and cost, but also to side reactions that limit current efficiency. Finally, the results obtained lead to conclude that dissolution rate was not behind the superiority of mild steel electrodes with regards to removal efficiency, and that alloying composition or physical properties do not account for significant variation in current efficiency, unlike CD which significantly reduced current efficiency.

3.2.4. Operational costs

According to Adhoum and Monser (2004), the primary cost factors of the EC process include consumptions of energy and electrode material. And so, operational costs associated with the use of EC technique for the treatment of HLGW in

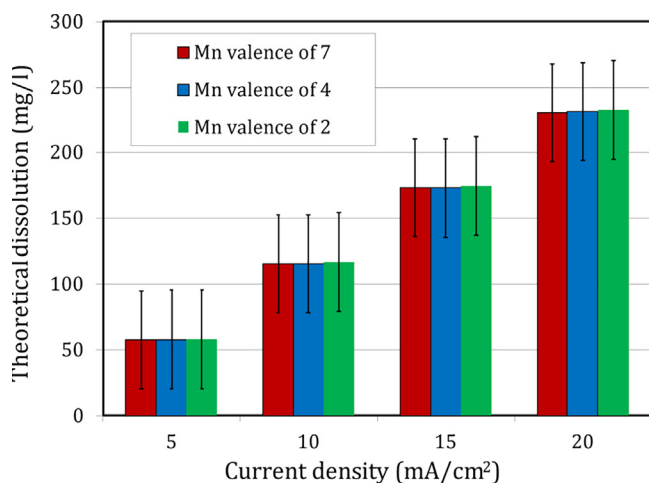


Fig. 11 Theoretical dissolution rate calculated for mild steel when assigning different valence values for Mn.

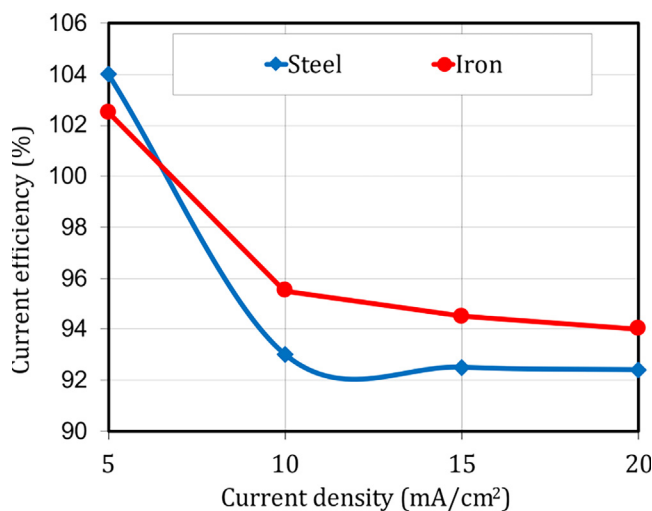


Fig. 12 Current efficiency for iron and steel electrodes at different current densities.

this study were determined for each electrode type examined based on dissolution rates and energy consumption.

The process of cost calculation incorporated results obtained under optimum CDs assigned for each electrode (Table 6). Based on assigning Jordanian market prices for cost factors, it was found that using mild steel electrodes demonstrate lower operational costs (Table 7). This conclusion would be obtained regardless of pricing systems used; this is because mild steel electrodes allowed for higher removal efficiency at a lower CD density when compared with iron electrodes. This

Table 7 Operational costs of electrocoagulation process when using iron and steel electrodes.

Electrode type	Operational cost factors		
	Electrical cost (\$/m ³)	Material cost (\$/m ³)	Total cost (\$/m ³)
Iron	0.037	0.06	0.097
Steel	0.014	0.04	0.054

resulted in lower material mass requirement (dissolution) and lower energy consumption (Table 6).

Results obtained on comparison between iron and mild steel electrodes for EC treatment of HLGW in this study are summarized in Table 8. As demonstrated, operational cost is another criterion in which mild steel is superior to iron as an electrode material for treatment of HLGW. Better COD removal and current efficiencies are also obtained when using mild steel electrodes.

The experimental results show how EC can substantially improve grey water quality, and due to the compactness and high optimization amenity of EC, it has good potential for applications such as decentralized wastewater treatment and reuse in agriculture and municipal uses like toilet flushing. Optimization and exploring different electrode characteristics reflected a substantial reduction in operating costs, this highlights the importance of bench scale and pilot testing prior to application of EC technology. This also shows that materials of almost identical composition can yield significantly different results. This study suggests the need for further testing of some aspects that were only speculative in this research. Some specific aspects are suggested including exploring the impact of alloy and mechanical properties of electrodes on removal of specific chemical ions, the oxidation states of coagulating metallic ions and hydroxides, and bubble formation and generation. Exploring these aspects would provide valuable insights into the effect of metal properties on the treatment process of GW and can substantially reduce the cost of the treatment process.

4. Conclusions

In this study, iron and mild steel electrodes were compared for the treatment of highly loaded grey water (HLGW) using an electrocoagulation technique. The comparison was achieved at different current densities (5, 10, 15, and 20 mA/cm²) which were applied in the EC reactor for 10 min of reaction time. The results demonstrated that, for both electrodes, cathodic reactions were dominated at higher applied current densities (> 10 mA/cm²), while anodic reactions were dominated at lower applied current densities (< 10 mA/cm²). Both electrodes demonstrated good removal efficiencies of COD and turbidity. The increasing CD did not substantially improve removal efficiency for either iron or mild steel but was shown as a driving factor for side reactions that limit current efficiency for both electrodes. However, mild

Table 6 Optimum operational conditions assigned in this study for iron and steel electrodes.

Electrode type	Optimal operational variables assigned			
	Current density (mA/cm ²)	Direct current (A)	Energy consumption (kWh/m ³)	Dissolution rate (g/m ³)
Iron	10	0.16	0.672	111.6
Steel	5	0.08	0.254	60.4

Table 8 Summary of results obtained for using iron and steel electrodes under optimal conditions.

Electrode type	Performance indicators				
	COD removal %	Turbidity removal %	Current efficiency (%)	Energy consumption kWh/m ³	Operational cost (\$/m ³)
Iron	85.3	94	95.30	0.672	0.097
Steel	86.5	92	104.02	0.254	0.054

steel electrodes demonstrated lower operational costs as they required lower CD (5 mA/cm²) and lower dissolution rate to remove 86.5% of COD in comparison with iron electrodes which required relatively higher CD (10 mA/cm²) to remove 85.3% of COD, and thus mild steel-based electrodes are considered superior to iron electrodes. Both electrodes showed higher current efficiencies at minimal CD applied, suggesting that current effects pitting corrosion similarly for both electrodes. The CD affected dissolution rates similarly for both electrodes, but iron anodes showed higher passivation, which supports that the physical properties of electrodes were the influencing factor, rather than the dosage of coagulating species. At optimum conditions, analysis of operating costs in terms of energy consumptions and electrode materials requirements were 0.054 \$/m³ and 0.097 \$/m³ achieved by steel and iron electrodes, respectively. The results of this study show that two electrode materials with almost identical chemical compositions can produce different results. It is speculated that the physical characteristics of electrode materials, namely surface smoothness, may have impacted bubble formation and deposition. The study recommends that the effect of mechanical and physical properties of electrode materials and testing different alloys of similar metals should be further explored as it can reflect a substantial reduction in operating costs of EC treatment of HLGW, rather than only exploring and investigating different materials.

CRediT authorship contribution statement

Khalid Bani-Melhem: Conceptualization, Project administration, Supervision, Investigation, Methodology, Data curation, Visualization. **Muhammad Rasool Al-Kilani:** Investigation, Data curation, Writing – original draft, Formal analysis.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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