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Continuous-flow electrocoagulation (EC) process for iron removal from water: Experimental, statistical and economic study

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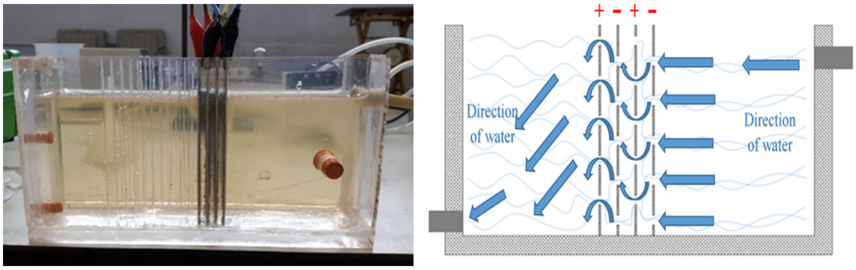
HIGHLIGHTS

- The new EC reactor removed 99.9% of 30 mg/l of iron.
- The performance of the new unit was modelled with R^2 of 0.9788.
- The new EC reactor reduced the need for external mixing devices.
- The operating cost for the iron removal using the proposed EC unit was 0.623 £/m^3.

GRAPHICAL ABSTRACT



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ABSTRACT

The process of Electrocoagulation (EC), the in-situ production of coagulants by passing an electric current through sacrificial electrodes, is free of chemical additives and cost-effective. This makes it the most widely used water and wastewater treatment method. However, the literature highlights some significant drawbacks of this method including EC unit design limitations. This research therefore aimed to develop a new EC unit design using drilled plates (electrodes) to mix the solution being treated without using external mixers, this minimising power consumption. The performance of the new EC unit was validated by applying it to remove iron from water taking into account the effects of applied current density (ACD), the pH of the water (PoW), iron concentration (IC) and treatment time (TT). The effects of these parameters were optimised using the Box-Behnken model. Synthetic water samples containing different concentrations of iron (10-30 mg/l), were treated in a continuous flow, using the new EC reactor at different ACD (1.5–4.5 mA/cm^2), PoW (4–10) and TT (10–50 min). The results revealed that the removal of 99.9% of iron was achieved by keeping PoW, ACD, IC and TT at 7, 3 mA/cm^2, 10 mg/l and 50 min, respectively. The effects of ACD, POW, IC and TT on iron removal could be successfully simulated with $R^2 = 0.9788$. The cost of removing iron using the proposed EC unit was 0.623 £/m^3.

1. Introduction

Environmental pollution is a serious problem that threatens the existence of humankind and other forms of life on planet Earth (Pandey and Singh, 2019; Wen et al., 2017). Unfortunately, recent significant growth in both global population and industry has intensified the problem of environmental pollution via the production of huge amounts of solid wastes and wastewaters (Hashim et al., 2017b). Environmental pollution has already changed our global climate in turn increasing water demand and limiting the availability of freshwater (Hashim et al., 2019; Koop and van Leeuwen, 2017; Zubaidi et al., 2019). At present, water polluted by heavy metals represents a serious challenge for the water industry because of the serious impacts of heavy metals on both human health and the environment (Bosch et al., 2016). Iron is one of the heavy metals commonly occurring in surface water and groundwater (between 0.5 and 50 mg/l) due to its elevated concentrations in the lithosphere and because of the discharge of iron-containing

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wastewater into water resources (Chaturvedi and Dave, 2012). The literature indicates that iron forms 5% of the chemical composition of the lithosphere, and it can be found at elevated concentrations in the effluents of several industries, such as mining and steel industries (Ityel, 2011). Although iron is an essential element for human health, excessive concentrations cause a wide range of health problems, such as cognitive disorder (Hashim et al., 2017b; Yan et al., 2014). Elevated iron concentration is a cause for concerns for both industry and the economy this including the development of unpleasant odours, laundry staining and unwanted colouring in papers and textiles (Dahshan et al., 2013; Tang et al., 2013; Yan et al., 2014). Iron also contributes to pipe clogging as it promotes bacterial growth inside pipes, resulting in costly maintenance (Chaturvedi and Dave, 2012; Ityel, 2011). Based on these considerations, the World Health Organization (WHO) sets 0.3 mg/l as the maximum allowable concentration of iron in drinking water (Hashim et al., 2017b).

To meet this limitation, different types of advanced treatment processes have been used to remove iron from water, such as ion exchange, activated carbon, supercritical fluid extraction, bioremediation, oxidation by aeration, chlorination and filtration (Bolisetty et al., 2019; Dahshan et al., 2013; Tang et al., 2013; Yan et al., 2014). Among these methods, electrocoagulation methods (EC) offer attractive merits, such as the in-situ generation of coagulants without the need for chemical additives, ease of operation, low power consumption, requiring less maintenance in comparison with other methods, and the production of only a small amount of sludge (Abdulhadi et al., 2019; Doggaz et al., 2019; Hansen et al., 2019). As such, EC has been used to treat a wide spectrum of pollutants, in particular, heavy metals (Hashim et al., 2020b; Hashim et al., 2018).

In this context, the current research investigates the removal of iron from synthetic water using an aluminium-based electrocoagulation reactor, focusing mainly on the influence of applied current density (CD). The experimental work was carried out under continuous flow conditions at a flow rate of 50 ml/min, initial pH of 6.5, a 5 mm gap between electrodes and a room temperature of 20 ± 1 °C. The initial pH of the water samples was kept constant at 6.5 because aluminium electrodes work efficiently at this value (Chafi et al., 2011; Hashim et al., 2020a).

2. Electrochemical reactions

When aluminium (Al) is used as an electrode material, the anodes produce Al^{3+} ions. These cations instantaneously experience additional reactions forming various kinds of monomeric and polymeric species, which also instantly coagulate forming aggregates (Essadki et al., 2009; Ghosh et al., 2008). These reactions are as follows (Chaturvedi, 2013; Ghosh et al., 2008):

At anodes:

$$Al_{(solid)} \rightarrow Al^{3+}_{(aqueous)} + 3e \tag{1}$$

At cathodes:

$$3H_2O + 3e \Leftrightarrow \frac{3}{2} H_2\uparrow + 3OH^- \tag{2}$$

The produced $Al^{3+}$and OH^- will react forming $Al(OH)_3$ as follows:

$$Al^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H^+ \tag{3}$$

Reactions between Al^{3+} and hydroxide ions form various aluminium monomeric and polymeric species, such as $Al(OH)^{+2}$, $Al_2(OH)_2^{4+}$, $Al_7(OH)_{17}^{4+}$, and $Al_{13}O_4(OH)_{24}^{7+}$, according to the pH of the solution (Ghosh et al., 2008). These species are transformed, according to complex precipitation kinetics, into $Al(OH)_{3(S)}$ (Ghosh et al., 2008).

The removal of iron ions using an aluminium-based EC unit can be summarised as follows (Chaturvedi and Dave, 2012; Ghosh et al., 2008):

$$Fe^{2+} + \left(\frac{1}{4}\right) O_2 + H^+ \Leftrightarrow Fe^{3+} + \left(\frac{1}{2}\right) H_2O \tag{4}$$

$$H_2O \Leftrightarrow H^+ + OH^- \tag{5}$$

$$NaCl \Leftrightarrow Na^+ + Cl^- \tag{6}$$

$$Fe^{2+} + 2Cl^- \Leftrightarrow FeCl_2 \tag{7}$$

$$FeCl_2 + 2OH^- \Leftrightarrow Fe(OH)_2\downarrow + 2Cl^- \tag{8}$$

Cl^- reacts with hydrogen ions near the cathodes, forming hydrochloric acid or evaporating near the anodes, in the form of Cl_2 (Ghosh et al., 2008).

3. Experimental work

3.1. The new EC reactor

Mollah et al. (2004) and Hashim et al. (2017a) have shown that EC reactor designs have not significantly changed over the last few decades. Generally speaking, rectangular reactors with plane rectangular electrodes were, and still are, the predominant design. As such, the first stage of the current study was to develop a new EC reactor which would minimise power consumption by replacing external water mixers with drilled electrodes. This new reactor consists of a 1.0 l rectangular Perspex container, (Fig. 1-A), supplied with four rectangular perforated aluminium electrodes with a total effective area of 280 cm^2

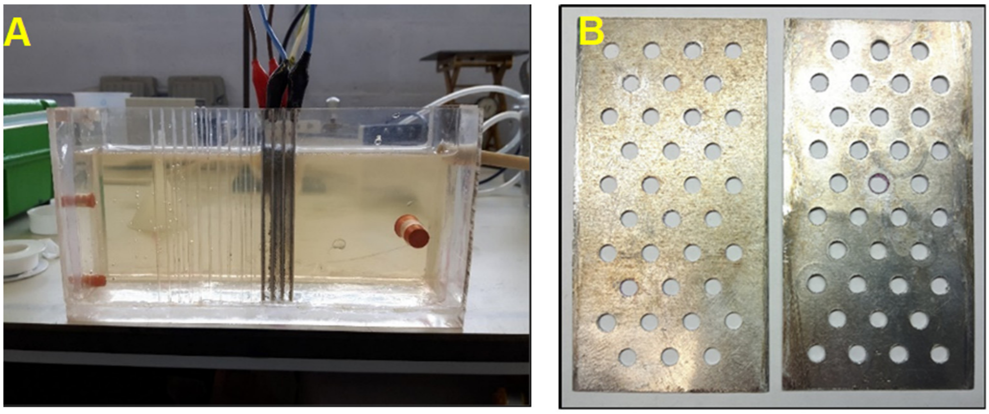


Fig. 1. A) the EC reactor, B) Aluminium electrode.

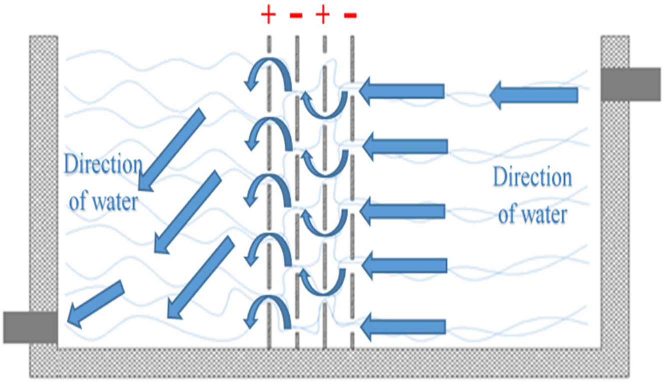


Fig. 2. The design and arrangement of the EC unit's electrodes.

(Fig. 1-B). Each electrode has 35 holes (5 mm in diameter), drilled in 3-holes and 4-holes lines to ensure that holes in the anodes are shifted by 5 mm from those in the cathodes (the very next electrode). This design of electrodes (distribution of holes), forces the solution being treated to flow in a convoluted path, this increasing water mixing efficiency without the need for external stirrers (Fig. 2). This design may be a cost-effective alternative to conventional EC reactors that depend on an external mixing device (magnetic or mechanical stirrers) which needs extra power to work.

Aluminium was used as it is an effective electrode material (Chaturvedi and Dave, 2012), while Perspex was used as it is inert and an affordable material (Ghosh et al., 2008; Heffron, 2015).

The EC reactor was connected to both a benchtop DC power supply (HQ Power; 0–30 V) and a peristaltic pump (Watson Marlow, model: 504 U) to supply the required electrical current and to circulate water through the reactor, as seen in Fig. 3.

3.2. Material and methods

3.2.1. Experimental work

The concentration of iron in natural surface water normally ranges between 0.5 and 20 mg/l but can be as high as 50 mg/l in some cases (Doggaz et al., 2018; Hashim et al., 2017b). Therefore, synthetic water samples with three different iron concentrations (IC) (10, 20 and 30 mg/l) were prepared by dissolving the correct amount of Fe_2 SO_4. $7H_2O$ in 1000 ml of deionised water (Ghosh et al., 2008). To investigate the influence of the other parameters, applied current density (ACD), pH of water (PoW) and treatment time (TT), on the removal of iron, the synthetic water samples were electrolysed at different ACD (1.5, 3 and 4.5 mA/cm^2) and PoW (4, 7 and 10) for different TT (10, 30 and 50 min) under continuous flow conditions. The gap between electrodes, water conductivity, and initial water temperature were kept constant during the course of the experiments at 5 mm, 6.5, 0.4 mS/cm, and 20 ± 1 °C, respectively. The initial pH and initial conductivity were adjusted to the desired level using HCl or NaOH, and NaCl, respectively, all chemicals supplied by Sigma Aldrich.

The progress of iron removal was monitored by collecting 5 ml-samples at specific intervals. These samples were filtered using 0.45 μm filter papers (Whatman filters) to separate outsludge, the residual concentration measured using an atomic absorption spectrophotometer (Thermo Scientific, Model: ICE 3300) (Ghosh et al., 2008). The removal efficiency was calculated as follows:

$$\text{Iron removal }(\%) = \frac{(\text{initial iron concentration} - \text{residual iron concentration})}{\text{initial iron concentration}} \times 100\% \tag{9}$$

3.2.2. Cost-effectiveness estimation

The preliminary operating cost of the new EC reactor was estimated according to the amount of the power consumed, chemicals and electrodes material, all summarised in the following equation (Hashim, 2017):

$$OC = \beta_1\ C_{power} + \beta_2\ C_{electrodes} + \beta_3\ C_{chemicals} \tag{10}$$

where C_{power} (kWh/m^3), $C_{electrode}$ (kg/m^3), and $C_{chemical}$ (kg/m^3) are power consumed, electrodes and chemicals. β_1, β_2 and β_3 are the unit prices of power, electrode materials and chemicals. Unit prices, according to the UK market 2019, were 0.1383 £/kWh of electricity, 0.8 £/kg of aluminium, 0.45 £/l of HCl, and 0.3 £/kg of NaCl.

The amount of electrodes consumed can be measured by weighing the anodes before and after the treatment process. Because deposits on the surfaces of anodes can influence this measurement, Faraday's second Law was used to estimate the consumed weight of electrodes (Hashim, 2017; Vidal et al., 2016):

$$C_{\text{material}}(\text{g}) = \frac{\text{Applied current (amp)} \times \text{treatment time (sec)} \times \text{molecular weight}\left(\frac{\text{g}}{\text{mol}}\right)}{3 \times 96500\left(\frac{\text{C}}{\text{mol}}\right) \times \text{Volume of solution }(\text{m}^3)} \tag{11}$$

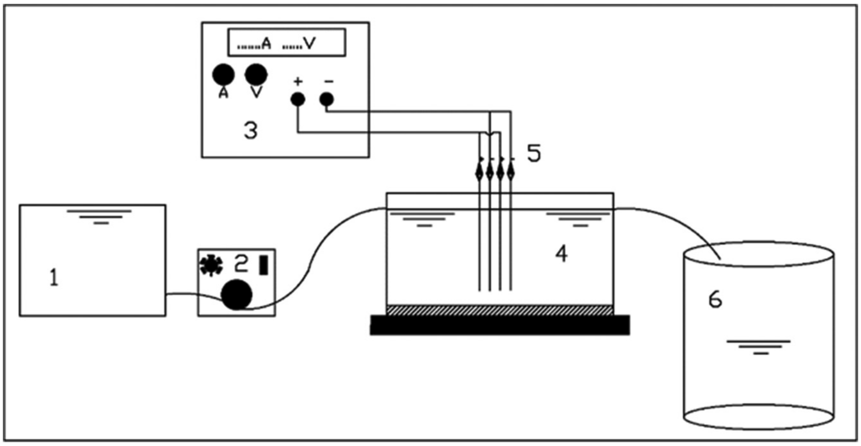


Fig. 3. EC setup: 1. Influent tank (polluted water). 2. Peristaltic pump. 3. Power supply. 4. Plastic container. 5. Wires. 6. Final storage.

Table 1
Maximum and minimum limits of the studied parameter.

Parameters	Maximum limit	Minimum limit
ACD (mA/cm^2)	1.5	4.5
PoW	4	10
IC (mg/l)	10	30
TT (min)	10	50

Power consumption was calculated as follows (Hashim, 2017):

$$C_{power}\ (kWh/m^3) = \frac{\text{Appplied current (amp)} \times \text{cell voltage (volt)} \times \text{treatment time}}{1000} \tag{12}$$

The amount of chemicals consumed was measured manually during the experimental work.

3.2.3. Optimisation of the effects of the studied parameters

The Box-Behnken model was used to optimise the effects of applied current density (ACD), pH of water (PoW), iron concentration (IC) and treatment time (TT) on the removal of iron using the electrocoagulation method. This model minimises the number of experiments required and it produces a regression model that can be used to predict the behaviour of the process under different operation conditions (Acharya et al., 2018; Chauhan et al., 2013). To perform the Box-Behnken model, the maximum and minimum limits of the parameters selected were specified as seen in Table 1 (Hashim et al., 2017b; Isa et al., 2014), the model applied using Minitab software (version 19.2).

Table 2 details the resultant experiment designs.

4. Results and discussion

4.1. Cost-effectiveness estimation

The experiments listed in Table 2 were carried out under controlled conditions, the results listed in Table 3. The relationships between the removal of iron and the studied parameters are shown in Fig. 4.

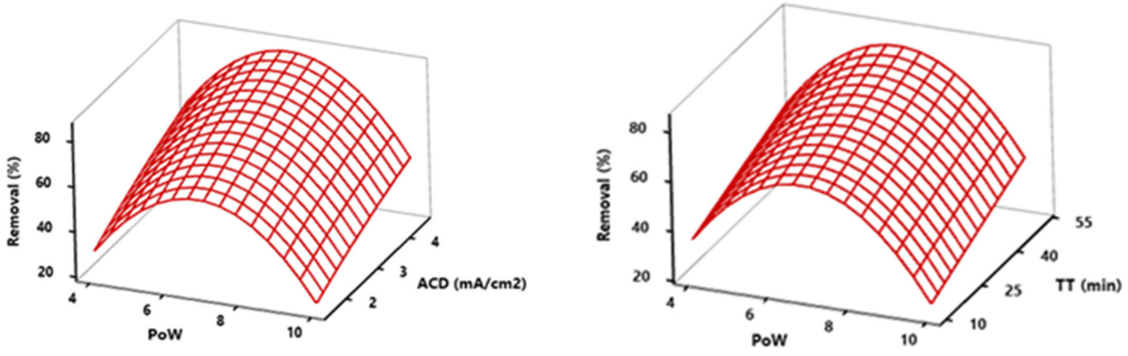
Almost all the iron (99.9%) was removed at an initial neutral PoW (7.0), the longest TT (50 min), the lowest IC (10 mg/l) and an ACD of 3 mA/cm^2, while the least amount of iron removal occurred when the PoW was alkaline (10.0), ACD at its lowest (1.5 mA/m^2) and with moderate values of TT and IC. There are two known significant effects of PoW on iron removal. Firstly, PoW changes the chemical formula (speciation) of the freshly produced aluminium hydroxides this changing their capacity for pollutants; the predominant type of aluminium hydroxide at neutral PoW is $Al_{13}O_4(OH)_{24}^{7+}$, this having a good capacity to remove pollutants (Hashim et al., 2017a; Kim et al., 2016; Matis and Peleka, 2010). Secondly, Alam et al. (2017) found H_2 bubbles which are produced at neutral pH, are small in size that increases the surface area of these bubbles, consequently enhancing the separation of pollutants through a floatation path. A wide body of literature proved the direct relationship between ACD and the production rate of aluminium hydroxides and H_2 bubbles, an increase in ACD resulting in a significant increase in the amount of aluminium hydroxides and H_2 bubbles in the solution being treated (Betancor-Abreu et al., 2019; da Mota et al., 2015). This impacts positively on the removal of iron. In the same vein, increasing the TT, leads to an increase in the amount of aluminium hydroxides and H_2 bubbles, this also positively reflected in the removal of iron (Kolesnikov et al., 2017; Perfil'eva et al., 2016). Finally, an increase in iron concentration (IC) requires more aluminium hydroxides to remove it (Hashim et al., 2017b), this negatively influencing removal efficiency.

Table 2
Experiment designs.

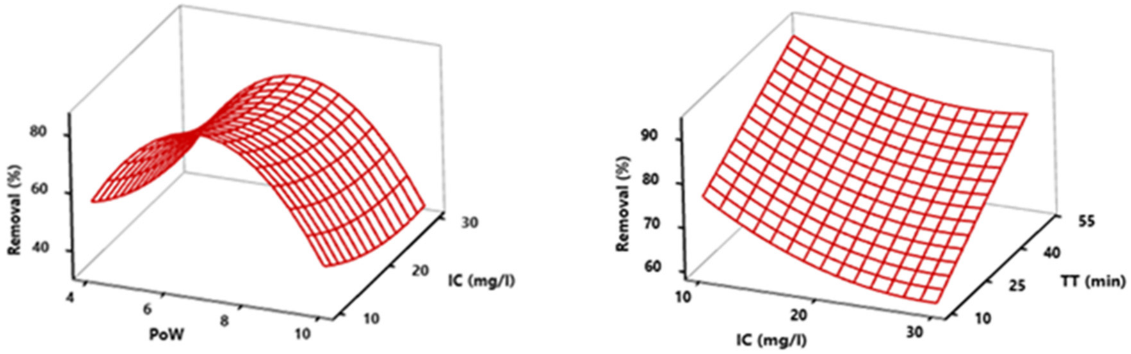
Run	PoW	ACD	IC	TT	Run	PoW	ACD	IC	TT
1	4	1.5	20	30	15	7	1.5	30	30
2	10	1.5	20	30	16	7	4.5	30	30
3	4	4.5	20	30	17	4	3	10	30
4	10	4.5	20	30	18	10	3	10	30
5	7	3	10	10	19	4	3	30	30
6	7	3	30	10	20	10	3	30	30
7	7	3	10	50	21	7	1.5	20	10
8	7	3	30	50	22	7	4.5	20	10
9	4	3	20	10	23	7	1.5	20	50
10	10	3	20	10	24	7	4.5	20	50
11	4	3	20	50	25	7	3	20	30
12	10	3	20	50	26	7	3	20	30
13	7	1.5	10	30	27	7	3	20	30
14	7	4.5	10	30					

Table 3
Experimental removal of iron using the electrocoagulation method.

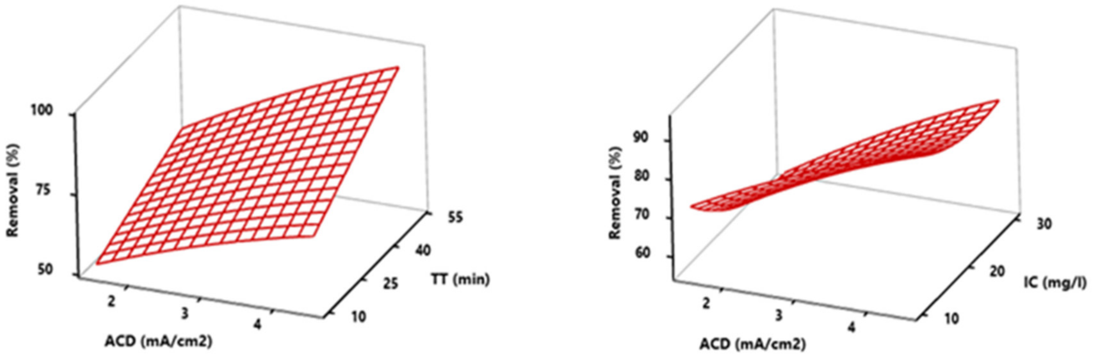
Run	PoW	ACD	IC	TT	Removal (%)	Run	PoW	ACD	IC	TT	Removal (%)
1	4	1.5	20	30	34.6	15	7	1.5	30	30	53.3
2	10	1.5	20	30	26.4	16	7	4.5	30	30	88.8
3	4	4.5	20	30	54.1	17	4	3	10	30	51.2
4	10	4.5	20	30	44.2	18	10	3	10	30	40.8
5	7	3	10	10	79	19	4	3	30	30	43.5
6	7	3	30	10	56.2	20	10	3	30	30	34.7
7	7	3	10	50	99.9	21	7	1.5	20	10	51
8	7	3	30	50	83.5	22	7	4.5	20	10	69.9
9	4	3	20	10	38.1	23	7	1.5	20	50	66
10	10	3	20	10	27.2	24	7	4.5	20	50	95.3
11	4	3	20	50	51.5	25	7	3	20	30	74.8
12	10	3	20	50	45.2	26	7	3	20	30	73.6
13	7	1.5	10	30	68.8	27	7	3	20	30	73.6
14	7	4.5	10	30	98.5						



A) Iron removal vs. PoW and ACD. B) Iron removal vs. PoW and TT.



C) Iron removal vs. PoW and IC. D) Iron removal vs. IC and TT.



E) Iron removal vs. ACD and TT. F) Iron removal vs. ACD and IC.

Fig. 4. Relationships between the removal of iron and the studied parameters.

The results were used in the Box-Behnken method to develop a regression model that simulates the effects of ACD, PoW, IC and TT on the removal of iron using the electrocoagulation, the regression model shown below.

$$\begin{aligned}\text{Removal }(\%) = {} & -93.9 + 50.76 \times \text{PoW} + 8.61 \times \text{ACD} - 3.02 \\ & \times \text{IC} - 0.046 \times \text{TT} - 3.774 \times \text{PoW}^2 - 0.685 \times \text{ACD}^2 \\ & + 0.0437 \times \text{IC}^2 - 1.4 \times 10^{-4} \times \text{TT}^2 - 0.094 \times \text{PoW} \\ & \times \text{ACD} + 0.0133 \times \text{PoW} \times \text{IC} + 0.0192 \times \text{PoW} \\ & \times \text{TT} + 0.097 \times \text{ACD} \times \text{IC} + 0.0867 \times \text{ACD} \times \text{TT} \\ & + 8 \times 10^{-4}\ \text{IC} \times \text{TT} \end{aligned} \tag{13}$$

This model was used to predict the removal of iron using different values of ACD, PoW, IC and TT, as shown in Table 2, the results of this shown in Table 4. To validate the performance of the model, predicted levels of iron removal were compared to observed removal (Table 4). These results show a good agreement between measured and predicted removal percentages, the maximum difference between measured and predicted removal of iron 4.48%. Fig. 5 shows that the coefficient of determination (R^2) of the relationship between predicted and measured removal of iron was 0.9788. The regression model can therefore predict 97.88% of the effects of the studied parameters on iron removal using the electrocoagulation method.

Table 4
Experimental and predicted removal of iron.

Run	PoW	ACD	IC	TT	Measured removal (%)	Predicted removal (%)	Difference (%)
1	4	1.5	20	30	34.6	30.12	4.48
2	10	1.5	20	30	26.4	21.87	4.53
3	4	4.5	20	30	54.1	56.11	−2.01
4	10	4.5	20	30	44.2	46.17	−1.97
5	7	3	10	10	79	76.47	2.53
6	7	3	30	10	56.2	60.31	−4.11
7	7	3	10	50	99.9	93.27	6.63
8	7	3	30	50	83.5	83.51	−0.01
9	4	3	20	10	38.1	35.75	2.35
10	10	3	20	10	27.2	24.35	2.85
11	4	3	20	50	51.5	53.45	−1.95
12	10	3	20	50	45.2	46.66	−1.46
13	7	1.5	10	30	68.8	72.26	−3.46
14	7	4.5	10	30	98.5	94.50	4.00
15	7	1.5	30	30	53.3	56.40	−3.10
16	7	4.5	30	30	88.8	84.46	4.34
17	4	3	10	30	51.2	55.91	−4.71
18	10	3	10	30	40.8	46.01	−5.21
19	4	3	30	30	43.5	42.15	1.35
20	10	3	30	30	34.7	33.85	0.85
21	7	1.5	20	10	51	52.50	−1.50
22	7	4.5	20	10	69.9	72.45	−2.55
23	7	1.5	20	50	66	67.31	−1.31
24	7	4.5	20	50	95.3	97.66	−2.36
25	7	3	20	30	74.8	74.08	0.72
26	7	3	20	30	73.6	74.08	−0.48
27	7	3	20	30	73.6	74.08	−0.48

4.2. Cost-effectiveness estimation

To calculate the preliminary operating costs of the new EC unit, the amount of electrodes material and power consumed were calculated using Eqs. (11) and (12), respectively. The operating cost was calculated using Eq. (2), standing at0.623 £/m^3 (approximately 0.78 \$/m^3). Other comparative treatment methods, i.e. membrane filtration method, cost approximately 0.94 \$/m^3 (Alzahrani and Mohammad, 2014; Sagle and Freeman, 2004).

5. Conclusion

The results obtained from the current study demonstrate the successful application of the new EC unit for iron removal from water based solutions. The results of the present study indicate that the performance of the new EC unit depends on the applied current density (ACD), pH of water (PoW), iron concentration (IC) and treatment time (TT). Generally, the removal of iron using the new EC unit increases with an increase of ACD and TT, but it decreases with an increase of IC. In terms of PoW, the results indicate that both acidic and alkaline environments are not beneficial for iron removal.

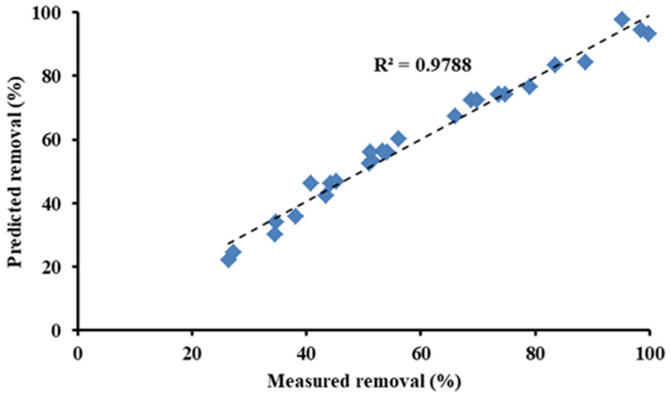


Fig. 5. Relationship between predicted and measured removal of iron using the electrocoagulation method.

The statistical analysis revealed that the effects of ACD, PoW, IC and TT on iron removal using the new EC unit could be simulated using the Box-Behnken model. The simulated results had a strong linear agreement with the experimental measurements at $R^2 = 0.9788$. The operating cost of the new EC reactor is 0.623 £/m^3 (approximately 0.78 \$/m^3), whereas the cost of the traditional treatment method i.e. membrane filtration, is 0.94 \$/m^3.

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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Author contributions

B.A. and P.K. conceptualized the content and methodology of this paper. B.A., K.H. and M.M. worked on the software, data validation and formal analysis. B.A. and K.H. prepared the samples used while B.A. and K.H. undertook data acquisition. The original writing and draft preparation was carried out by B.A., K.H., P.K. and M.M., reviewing and editing by R.A. and A.S. The project was supervised by P.K., A.S. and K.H.

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