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Energy efficient electrocoagulation using baffle-plates electrodes for efficient Escherichia Coli removal from Wastewater

Energy efficient electrocoagulation using baffle-plates electrodes for efficient *Escherichia Coli* removal from wastewater

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Abstract

A new electrocoagulation reactor (EC), which utilises the concepts of baffle-plates, has been applied to remove *Escherichia coli* (*E. coli*) from wastewater. This new aluminium-based EC reactor utilises perforated baffle-plates electrodes to mix water, which reduces the need for mechanical or magnetic mixers that require extra power to work. This new reactor has been used to treat *E. coli* containing wastewater samples, considering the effects of different parameters such as treatment time (TT), inter-electrode distance (IED), and current density (CD). A statistical analysis has also been commenced to evaluate the influence of each parameter on the removal of *E. coli*. Additionally, an economic study has been conducted to assess the operating cost of the new reactor. The outcomes of the experimental work confirmed that the new reactor removes as high as 96% of the *E. coli* within 20 minutes of electrolysis at IED of 0.5 cm, and CD of 1.5 mA/cm^2. Additionally, it has been found that the operating cost of the new reactor is 0.11 US $\$/m^3$ (for *E. coli* removal), which is less than operating cost of traditional reactors. Finally, it has been found that the effect of the studied parameters on *E. coli* removal followed the order: $TT > CD > IED$.

Keywords: Electrocoagulation; *E. coli*; wastewater; operating cost; statistical analysis.

1. Introduction

Though water treatment industry has recently witnessed a remarkable progress, water pollution has also significantly increased because of the rapid growth of both global population and industrial activities [1-3]. It is undeniable fact that the available water resource are polluted with a wide spectrum of pollutants that could cause different health problems [4, 5]. Among these pollutants, the pathogenic and non-pathogenic microorganisms are classified as a higher risk than other pollutants due the high number of illness and death that they could cause [6-8]. For instance, it has been reported that the pathogenic microorganisms cause different waterborne diseases, such as diarrhoea and gastrointestinal, which in turn cause about 2,000,000 deaths/ year [6, 9]. Therefore, different disinfection methods, such as chlorination, Ozonation, and irradiation with ultraviolet, have been used to remove pathogenic and non-pathogenic microorganisms from water and wastewater. For instance, chlorination method has been extensively used during the 1970s as an efficient and affordable disinfection method, where the powerful oxidizing ability of chlorine destroys the essential enzymes of microorganisms, which results in the death of these biological pollutants [10, 11]. The major disadvantage of chlorination method is the generation of extremely dangerous by-products, for instance the reaction between the chlorine and some humic substances could produce trihalomethanes, which categorised as carcinogenic pollutants [10, 11]. Microfiltration and ultrafiltration techniques are also effective methods for the removal of microorganisms, however the application of these techniques are highly limited by the fouling problems and the high operational cost [12]. Ozonation is another technique that has been applied as a disinfection method, where it has been reported that the ozone is a powerful oxidant that can kill the microorganisms by destroying their cell wall [10, 13]. Although the Ozonation process is very efficient and it does not generate trihalomethanes by-products, it is still expensive in comparison with other traditional methods [14], and it has been proved it could induce the formation N-nitrosodimethylamine (NDMA) [13].

Recently, the disinfection technology has witnessed noticeable advancement by combining more than one technology or by using new composite materials. For example, Zhou, et al. [15] developed a new disinfection method that utilises a tubular coaxial-electrode copper ionization cell to disinfect drinking water. The obtained results indicated that this method removed 6-log of *E. coli* within 2 min of treatment at operating voltage of 1.5 V. Moreno-Andrés, et al. [16] used advanced electrochemical cell that supplied with boron doped diamond electrodes to disinfect seawater. The outcomes of this study indicated that this advanced electrochemical cell removes 4.8-Log of natural marine heterotrophic bacteria at energy consumption of $0.264\ \mathrm{kWh.m^{-3}}$. Moreno-Andrés, et al. [16] utilises the nano-technology to develop a disinfection method that consists of an anodic multiwall carbon nanotube filter to remove viruses and *E. coli* from water. The authors demonstrated that this method reduced the number of viruses and *E. coli*, in 30 sec at voltage of 3 V, to below detection limit.

Electrocoagulation method (EC) has recently brought a big deal of attention as an effective method to remove microorganisms from wastewater and water because of its simplicity, selectivity, and relatively low operating cost [17-19]. Additionally, the EC technology does not need chemical additives to achieve the treatment process, and it could be easily automated and integrated with other treatment units [20-22]. Furthermore, EC method significantly minimises the volume of the generated solid waste (sludge) that requires high treatment cost [23, 24], which in turn significantly minimises the operational cost of the EC method. These advantages of the EC method make it very promising alternative to the traditional treatment methods. Table 1 summarises some of the recent applications of the EC method in water and wastewater treatment.

Table 1: some of the recent applications of the EC method in water and wastewater treatment.

No.	Authors	Targeted pollutants	Materials of electrodes	Results
1.	Hussin, et al. [25]	Lead Pb(II)	Zinc electrodes	The authors demonstrated that about 99.9% of Pb(II) was removed using zinc electrodes after 10 min of electrolysing at 1.13 $\mathrm{mA.cm^{-2}}$.
2.	Gilhotra, et al. [26]	Arsenic As(V)	Stainless steel electrodes	The authors found that 99.6 % of 10 ppm of As(V) could be removed using stainless steel electrodes. The optimum operational conditions were 5.2 for pH and 20 min for treatment time. Additionally, the authors stated that the removal efficiency dropped to 86 % as the concentration of As(V) increased to100 ppm.
3.	Hashim, et al. [27]	Fluoride	Aluminium electrodes	The authors demonstrated that about 98% of fluoride was removed using aluminium electrodes after 25 min of electrolysing at 2 $\mathrm{mA.cm^{-2}}$.
4.	Mohtashami and Shang [28]	Total suspended solids	Stainless-steel electrodes	The authors found that about 95% of total suspended solids was removed 8 min of electrolysing, with stainless-steel electrodes, at 100 $\mathrm{mA.cm^{-2}}$.
5.	Hansen, et al. [29]	Selenium	Iron electrodes	The authors demonstrated that 0.3 mg/L of selenium could be minimised to 0.03 mg/l after 360 min of electrolysing at 76.7 $\mathrm{A.m^{-2}}$, and pH of 5.0.

However, lack of reactor design (simple horizontal or vertical arrangement of square or rectangular plate electrodes inside a container) and the sensitivity of the electrocoagulation to the chemical composition of the liquid being treated represent the main drawbacks of the electrocoagulation technology [30].

The current investigation therefore, is devoted to develop a new energy efficient design of EC reactors. The performance of this baffle-plates aluminium-based EC reactor (BPECU) has been explored in terms of *E. coli* removal from synthetic wastewater samples. The main objectives of this study could be summarised as follows:

- Present a new energy efficient design for the electrocoagulation units that utilises the concept of baffle-plates to mix water without the need for external mixers. This could significantly enhance the cost-effectiveness of this method.
- Investigate the performance of the new energy efficient EC reactor in terms of *E. coli* removal from synthetic wastewater considering the effects of different parameters, such as treatment time (TT) (5 to 30 min), current density CD (0.5 to 2.5 mA/cm^2), and inter-electrode distance IED (0.5 to 1.5 cm).
- Estimate the preliminary operational cost of *E. coli* removal using the new electrocoagulation unit (BPECU).
- Conduct a statistical study to assess the relative influence of each parameter on the removal of *E. coli*.

2. Brief theoretical background

2.1. Disinfection mechanism

The literature shows different interpretations for the mechanisms of microorganisms' deactivation, which could be summarised in the following categories [31-34]:

- Cell lysis due to the cell wall damaging.
- Permeabilization of the cytoplasmic membrane that allow the essential nutrients to escape.
- Changing the nature of protoplasm, the latter could be severely damaged when the cell subjected for photons, heat, or pH shock.
- Altering the activity of enzymes due to the oxidisation, some oxidising agents destruct the chemical structure of enzymes that produces lethal effects.

2.2. Electrocoagulation disinfection mechanism

In the EC method, in addition to the mentioned mechanisms, the microorganisms could be deactivated due to the direct adsorption to the surface of the anode followed by electron transfer, and physical removal by floating of the microorganisms with produced hydrogen gas and/ or precipitating with the generated flocs [31, 32, 35]. It is noteworthy to highlight that very strong oxidising agents, such as $HOCl, OCl^-, ClO_2$ and Cl_2, are produced during the EC treatment according to the following reactions [31, 34]:

$$2Cl^- \rightarrow Cl_2 + 2e^- \tag{1}$$

$$Cl_2 + 2HO^- \rightarrow H_2O + OCl^- + Cl^- \tag{2}$$

$$Cl_2 + 4H_2O \rightarrow 2ClO_2 + 8e^- \tag{3}$$

These agents could damage the wall of the cell that results in the death of the microorganisms. Additionally, in case of using aluminium electrodes, the following reactions will take place [36]:

$$Al \rightarrow Al^{3+} + 3e^- \quad \text{(Anode)} \tag{4}$$

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \quad \text{(Cathode)} \tag{5}$$

The redox reactions that occur with aluminium anodes are [37]:

$$2H_2O + 2e^- \rightarrow H_{2(g)} + 2HO^-_{(aq)} \quad \text{(Cathode)} \tag{6}$$

$$2H_2O \rightarrow O_{2(g)} + 4H^+_{(aq)} + 4e^- \quad \text{(Anode)} \tag{7}$$

3. Materials and methods

3.1. Experimental set up

The experimental work has been commenced using the new EC reactor (BPECU). This reactor is consists of a rectangular Perspex container having a net volume of 12,600 mm^3. This container was supplied with four perforated baffle-plates (as electrodes) that made from aluminium (purity of 99.5%); each electrode has dimensions of 4 cm in width and 9 cm in height, and contains 35 holes. The latter were 4 mm in diameter and distributed in 10 rows, figure 1 (A). It can be noticed from figure 1(A) that these holes were distributed in four-hole and three-hole lines. This distribution enforces the water to flow in convoluted paths, which helps to effectively mix water without the need for external mixing devices. Fourteen slots, 1 mm × 1 mm, were cut in both sides of the new reactor at distances of 5 mm, which were used to hold the electrodes at the required position, Figure 1(B). The total effective area of electrodes, immersed in water, was 207 cm^2. It is noteworthy to mention that aluminium has been used, in the current study, as electrodes material Aluminium has been used as electrodes material because of its cost effectiveness, ready availability, and it requires comparatively less oxidation potential [38].

A peristaltic pump (Watson Marlow type, model: 504U), and a DC power source (Type: HQ, Model: PS 3010) was connected to the BPECU to pump water and deliver the required current density, respectively. Both temperature and pH of water were monitored using a pH/temperature meter (Type: Hann a; Model: HI 98130).

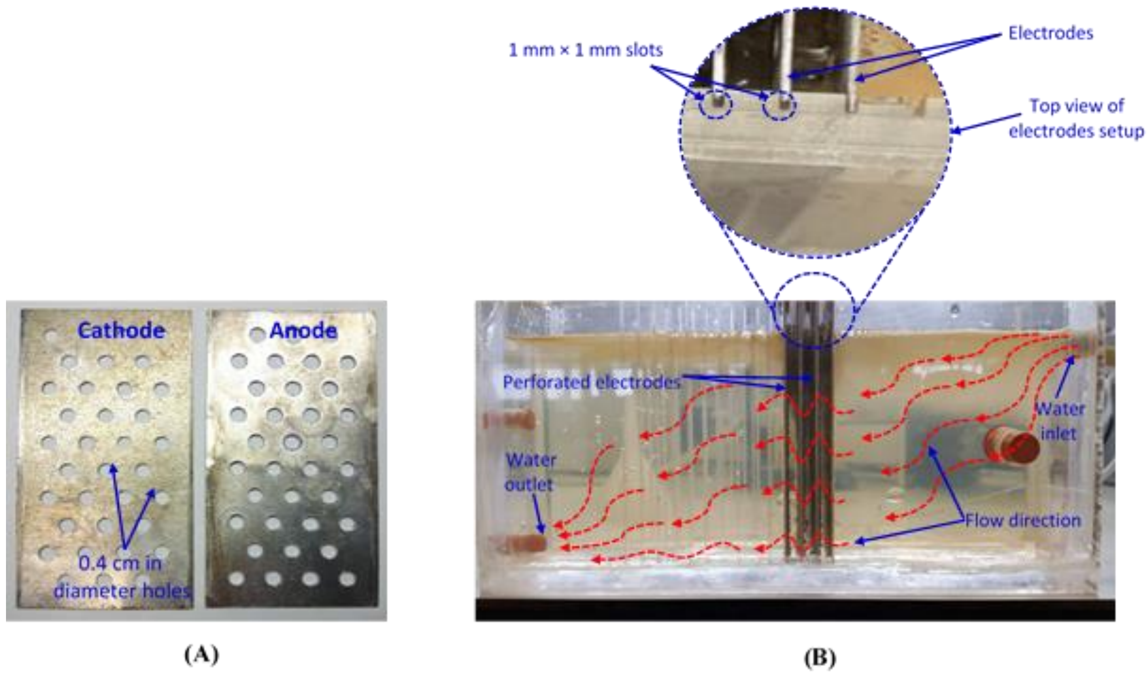

Figure 1: (A) Electrodes, (B) BPECU.

3.2. Mixing efficiency of the new EC reactor (BPECU)

Water mixing is a key factor in the electrocoagulation process as it helps to create a good homogenisation of coagulants and pollutants inside the EC reactor, which in turn helps to create big enough flocs to settle. In the current study, perforated baffle-plates have been used in the design of BPECU to achieve the water mixing process without the need for external mixing devices. The mixing efficiency of the BPECU, along with a traditional EC reactor, was investigated in continuous flow mode. The traditional EC reactor has similar design to BPECU but with solid electrodes.

Measurement of water mixing efficiency was initiated by filling each reactor with clear deionised water and then water with an initial concentration of red drain dye of 400 mg/L, which was pumped continuously into the reactors by a peristaltic pump. Water pumping was conducted at a constant flow rate of 30 mL/min for 30 min. A visible tracing for water mixing process across the reactor was conducting using an accurate HUE HD camera. This camera was installed at 30 cm to the studied reactors to give a clear view of the mixing process during the 30 minutes of treatment. Video records help to check if there is an unmixed area inside the reactor (stagnation zones). The camera records then were separated into frames using VirtualDub software. The unmixed areas, on these frames, were calculated using AutoCAD-2014 software.

3.3. E. coli removal

E. coli containing synthetic wastewater sample was prepared according to [39]. Initially, the *E. coli* (ATCC 35218 supplied by Fisher Scientific) was cultured in a flask containing a 0.1 l Luria Broth Base. The incubation process was carried out by shaking the flask at 150 rpm for 24 hrs at temperature of 37 °C using a temperature controlled shaker. Then, the growth media was separated from the cells by centrifuge the culture. The separated cells were washed buffer solution, and then suspending in buffer solution. The latter was prepared by 0.01 and 1.0 mole of KCl and $NaHCO_3$ in 1.00 L of distilled water, respectively. The initial concentrated concentration of *E. coli* was 10^8 UFC/ 100 mL. Lower concentrations (10^5 UFC/L) of initial concentrated concentration of *E. coli* were diluted from this stock solution. After each dilution process, the caps and top edge of the bottles were sterilised by flame to avoid the pollution with external types of bacteria.

These diluted solutions were treated using the new electrocoagulation reactor at different TTs, CDs, and IEDs. Then, the remaining *E. coli* were calculated after each run of treatment. The required chemicals were provided by Sigma-Aldrich, UK.

3.4. Experimental work

The electrocoagulation process was initiated by placing 1.0 L of the diluted sample in the new reactor. This sample was treated at different TTs (from 5 to 30 min), CDs (from 0.5 to 2.5 mA/cm^2), and IEDs (from 0.5 to 1.5 cm). All the experiments were performed at room temperature (20 ± 1^0C), initial pH of 7, flow rate of 30 mL/min, and water conductivity of 0.32 mS/cm (using NaCl salt).

The *E. coli* removal (RE %) was calculated as follows:

$$\mathrm{RE\%} = \frac{C_i - C_f}{C_i} \times 100\% \tag{8}$$

Where C_i is the numbers of the *E. coli* cells before treatment (in UFC/L), and C_f is the numbers of the *E. coli* cells after treatment (in UFC/L).

It is noteworthy to highlight that initial pH of the diluted samples was kept constant at 7 (within the normal pH range of wastewater (≤ 7) [40, 41]).

At the end of each treatment run, the surfaces of the electrodes were cleaned with HCl acid, and then rinsed with deionised water for three times.

Finally, the residual aluminium concentration in the treated water has been measured, using Thermo atomic absorption spectrophotometer (Model: ICE 3300), and compared to the World Health Organisation (WHO) guideline that recommends 0.2 mg/l as the maximum allowable concentration for aluminium in drinking water [42].

3.5. Operating cost

In field works, the operating cost of any treatment unit must cover the costs of construction, maintenance, labour, sludge handling, consumed power, materials, and chemicals [43]. While for lab-scale units, the estimation of operating cost should include the costs of the consumed power, materials, and chemicals [43, 44], which could be calculated as follows [43]:

$$\text{Operating cost} = \gamma_{\text{electrodes}} \times Q_{\text{electrode}} + \gamma_{\text{power}} \times Q_{\text{power}} \tag{9}$$

Where, $\gamma_{\text{electrodes}}$ represents the price of electrode material, and $Q_{\text{electrode}}$ (kg of Al /m^3) represents the consumed weight of electrodes material. The latter could be estimated as follows [20]:

$$Q_{\text{electrode}}\,(\text{grams}) = \frac{I \times t \times m}{Z \times F} \tag{10}$$

Where I, t, M, Z, and F represent the electrical current in amperes, treatment time in seconds, the molecular weight of electrodes materials (26.98 g/mol for aluminium), Z the number of electrons in the electrodes materials (3 for aluminium), and Faraday's constant (96487 C/mol), respectively.

While γ_{power} represents the price of power unit, and Q_{power} (kWh/m^3) is the consumed power. The latter has been calculated as follows [45]:

$$\text{Power Consumption} = \frac{I * V * t_e}{\text{Vol.}} \tag{11}$$

Where V, t_e, and Vol represent the potential (V), electrolysing time (hours), and volume of water (m^3), respectively.

3.6. Analysis of the relative importance of operational parameters

In this step, the relative influence of each parameter (TT, CD, and IED), on the removal of *E. coli* using BPECU, was be measured by determining its Beta coefficient (β) value; where the higher the *β* value, the higher the effect [20, 46]. This analysis indicates whether the studied parameter significantly influences the removal of *E. coli* or not. SPSS-26 package has been used to perform the statistical analyse and to determine the values of β coefficient for each parameter.

4. Results and discussion

4.1. Mixing efficiency of the new EC reactor (BPECU)

As mentioned before, water mixing is a key factor in the electrocoagulation process for creating a good homogenisation of coagulants and pollutants inside the EC reactor, which in turn helps to create big enough flocs to settle.

In the current study, the mixing efficiency was examined by pumping coloured water, at flow rate of 30 mL/min for 30 min. The mixing process was monitored using an accurate camera. The obtained results confirmed that the new reactor, BPECU, needed 15 min to create a homogenous colour distribution across the whole reactor, while the traditional EC reactor did not achieve homogenous colour distribution even after 30 min of continuous pumping process, figure 2.

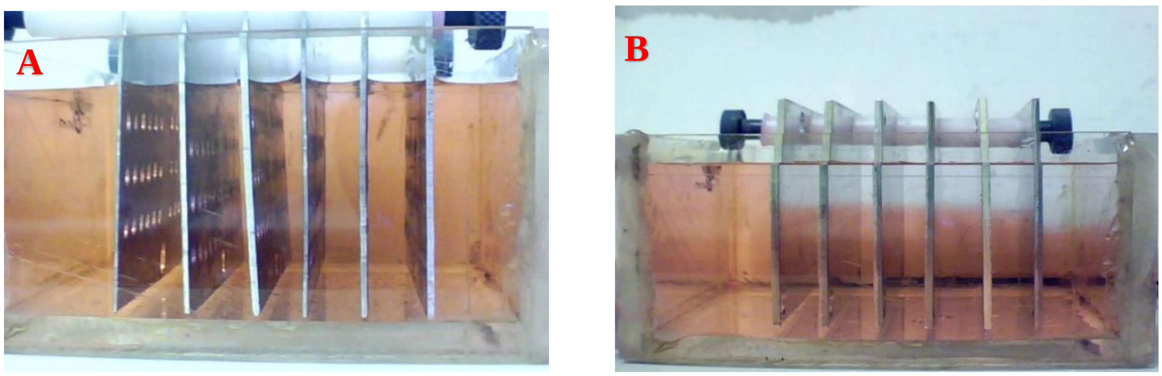

Figure 2: Mixing efficiency of (A) BPECU, (B) A traditional EC reactor.

4.2. Influence treatment time

Electrolysing time exerts a significant effect on the efficiency of the EC unit as it determines the amount of coagulants produced electrochemically in the process [47]. Therefore, the effect of this important parameter on the performance of BPECU has been explored by treating *E. coli* containing water samples at different treatment times (5 to 30 min). During the electrolysing process, the initial pH, IED, and CD were kept constant at 7, 0.5 cm, and 0.5 mA/cm^2, respectively. A rapid removal of *E. coli* was noticed during the first 20 min of electrolysing, and then a slight increase was noticed over the rest of electrolysing time, Figure 3(A). The increase in the removal of *E. coli* during the first 20 min of electrolysing process is resulting from the increase in the quantity of freshly produced coagulants [20]. However, the slight increase in the removal efficiency over the last 10 min of treatment could be attributed the development of a passive layer on the surfaces of nodes that reduces the production coagulants, which decreases the removal of *E. coli* [20]. It is noteworthy to mention that there are several techniques could be used to inhibit or minimise the development of a passive layer on the surfaces of nodes, for example adding 60 mg/L of chloride will inhibit the development of this passive layer [48]. In addition, figure 3(B) shows a significant increase in the power consumption with the progress of treatment time. Hence, TT of 20 min will be used to complete the experimental work.

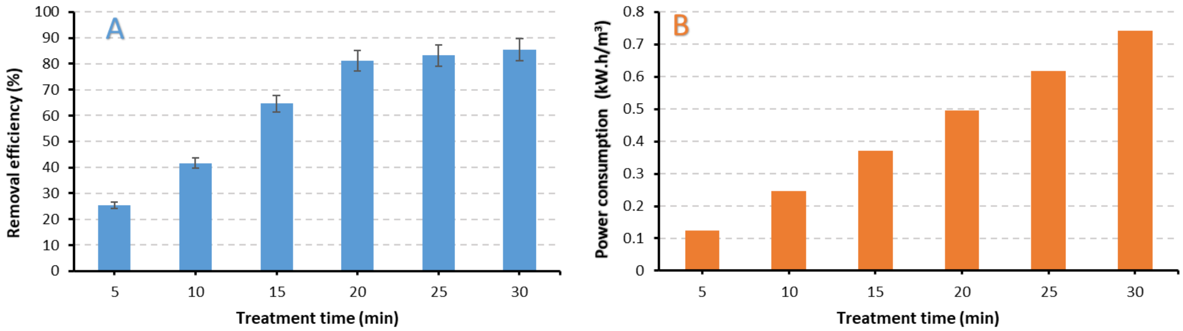

Figure 3: Effect of TT on: (A) *E. coli* removal, (B) Power consumption, error bars represent the values of the standard deviation.

4.3. Influence current density (CD)

The applied CD is also exert a significant effect on the efficiency of the EC unit as it controls the production of both coagulants and the hydrogen bubbles, which means it directly determines the removal efficiency [47]. Thus, the effect of this key parameter on the removal of *E. coli* has been investigated in the current study at three different values, which are 0.5, 1.5, and 2.5 mA/cm^2.

The results indicated that the CD significantly influence the removal efficiency, figure 4(A). It has been found that increasing the CD from 0.5 to 2.5 mA/cm^2 increased the *E. coli* removal efficiency from about 81% to 100%, respectively. This increase in the removal of *E. coli* might be attributed to fact that the number of the generated aluminium ions increases with the increase of the CD [20, 30], which enhances the removal efficiency. However, figure 4(B) indicates that the power consumption considerably increased with the increase of the applied CD. Therefore, in the current study, CD of 1.5 mA/cm^2 will be used to complete the experimental work.

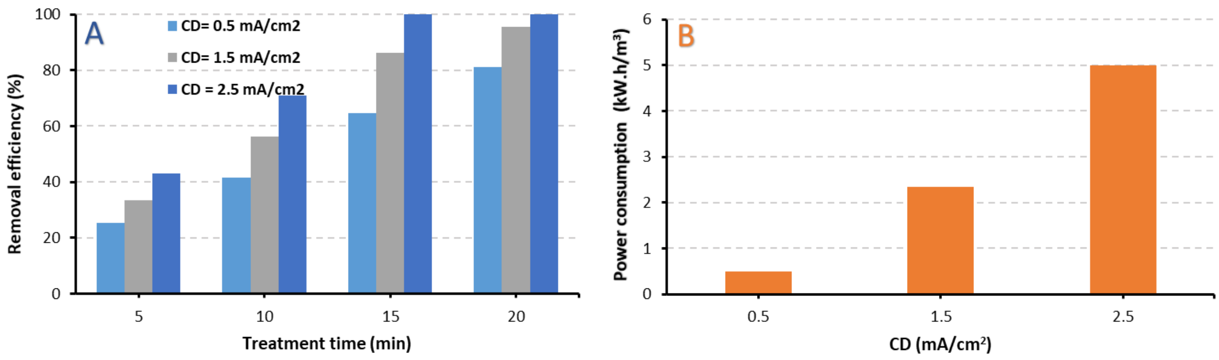

Figure 4: Effect of CD on: (A) *E. coli* removal, (B) Power consumption.

4.4. Influence of inter-electrodes distance (IED)

The relevant literature confirms that the gap between electrodes effects the efficiency of electrocoagulation process as it determines the electrical resistance inside the EC units, and the growth of the passive layer on the surfaces of the anodes [47]. Therefore, some experiments have been commenced to explore the influence of the IED on the removal of E. coli using BPECU. These experiments were commenced at IED ranging from 5 to 15 mm, keeping the initial pH, CD, and TT constant at 7, 1.5 mA/cm^2, and 20 min, respectively.

The results obtained, figure 5(A), showed that removal of *E. coli* decreases with the increase of the IED. In addition, figure 5(B) confirms that the power consumption increases with the increase of the IED. As mentioned before, these negative impacts of long IEDs on the performance of the new electrocoagulation reactor is due to the growth of a passive layer on the anode and the increase in electrical resistance, which limits the production of coagulants, and consequently minimises the efficiency of the electrocoagulation reactor. Therefore, taking into account the results of power consumption and *E. coli* removal, an IED of 5 mm was considered as the optimum value to calculate the operating cost.

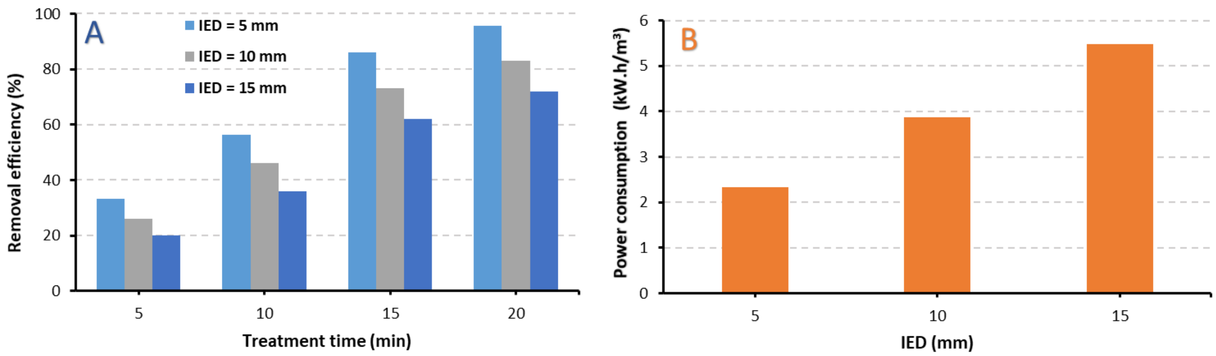

Figure 5: Effect of IED on: (A) *E. coli* removal, (B) Power consumption.

4.5. Residual aluminium concentration in the treated wastewater

The residual aluminium concentration in the treated wastewater has been measured and compared to the World Health Organisation (WHO) guideline (0.2 mg/l). It has been found the average residual concentration of aluminium was 0.24 mg/l, which is slightly higher than the recommended limit by the WHO. This residual concentration of aluminium has been accepted in this study because the treatment process is proposed for wastewater not for drinking water, which means the effluent wastewater from the EC unit will be discharged into a surface water body. As a solution for this problem, physical treatment unit, such as filtration unit, could be used to remove this extra amount of aluminium.

In conclusion, the experimental results confirm that the new electrocoagulation reactor (BPECU) could remove 96% of the *E. coli* within 20 min of electrolysing at initial pH of 7, CD of $1.5 mA/cm^2$, and IED of 5 mm. It is noteworthy to mention that, under the mentioned operational conditions; the pH of water being treated increased during the first ten minutes to vicinity of 9 and kept almost the same level for the rest of treatment time.

4.6. Operating cost

The operational cost of the new reactor, in terms of *E. coli* removal, has been estimated depending on the unit prices of the Iraqi market in May 2019 (power cost is 2.5 cent/kWh, and cost of 1 kg of aluminium is 1.53$). The consumed quantities of both power and electrode material have been calculated using equations 10 and 11, respectively. According to equation 5, the preliminary operating cost of *E. coli* using BPECU is about 0.11 US $\$/m^3$. Relevant literature showed different operational costs of traditional EC units (different designs of the EC cell) , such as 0.186 €/m^3 (0.2 $\$/m^3$) [49], 0.22 $\$/m^3$ [47], and 0.29 $\$/m^3$ [50]. In terms of other types of treatment methods, Downing, et al. [51] demonstrated that combined methods (consists of membrane, filtration, and ultraviolet units) could remove *E. coli* efficiently from wastewater, but the operational cost of this method was 0.698 $/m^3$.

It can be seen that the operational cost of the new reactor is less than that obtained, for other pollutants, by other researchers. This reduction in operating cost is because the new reactor reduced the need for external mixers and aerators that required extra power to work. Thus, the BPECU could be a cost-effective alternative to the traditional lab-scale electrocoagulation units.

4.7. Analysis of the relative importance of operational parameters

As it has been mentioned above, β coefficient has been calculated for each operating parameter to assess its relative effect on the removal of *E. coli* using BPECU. According to figure 6, the TT has the highest β value that means it exerts the highest influence on the removal of *E. coli*. While, IED has the lowest influence on the removal of the *E. coli*.

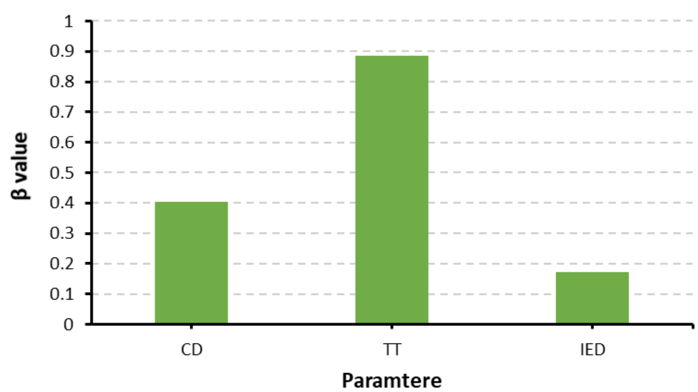

Figure 6: β values for the studied parameters.

5. Conclusion

The results obtained from the current investigation confirmed that BPECU is able to remove 96% of *E. coli* from synthetic wastewater sample, which is very similar to the reported efficiencies in the literature. However, the obtained results indicated that BPECU consumes less power than traditional units do, as it does not require external water mixing devices.

Generally, the obtained results indicated that *E. coli* removal is more efficient at high current densities and long treatment time. Contrarily, the removal of *E. coli* is reversely proportional to the gap between electrodes. Statistically, it has been found that the treatment time is the most significant factor in the removal *E. coli*, while the distance between electrodes has the lowest influence.

For future work, the mechanism of *E. coli* removal, using the BPECU, should be investigated.

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