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Modified bio-electrocoagulation system to treat the municipal wastewater for irrigation purposes

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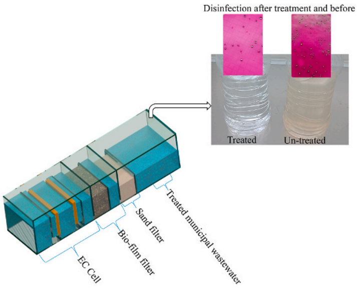
HIGHLIGHTS

- Integration of electrocoagulation system with Biofilm and Sand film filters.
- Parametric optimization of BEC system for the treatment of municipal wastewater.
- Disinfection study: Removal of total coliform and total bacterial flora was studied.
- Water quality testing was performed as per irrigation water quality standards.
- Cost Analysis: Cost analysis in terms of energy consumption was performed.

GRAPHICAL ABSTRACT



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ABSTRACT

A modified biological-integrated electrocoagulation method was explored to treat municipal wastewater (MWW) for irrigation purposes. To use treated wastewater for irrigation purposes a wide range of contaminants removal was focused on in this study (turbidity, hardness, conductivity, TDS, TSS, chloride, Ammonia nitrogen, BOD, COD, and total coliform). Raw municipal wastewater (RMWW) was treated in a modified Bio-Electrocoagulation (BEC) cell. The cell was operated in a continuous flow mode and consisted of an electrocoagulation stage using aluminum (*Al*) electrodes followed by a bioremediation stage using a fixed bio-filter (BF), the design of the cell was further modified by the addition of a sand filter (SF). The effect of several parameters such as applied voltage (22, 26, and 30 V), inlet flow rate (1, 3, and 5 Lh^{-1}), and initial pH (pH 3, 5, 7, 7.4, and 9) was investigated to determine the optimum operating conditions for selected responses. The most effective operating conditions for the BEC were investigated for the different irrigation water quality (WQ) indicators. It was observed that pH 7.4 and 26 V provide maximum removal efficiency of contaminants at the flow rate of 1 Lh^{-1}. A fixed film BF plays a

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positive role to improve the degradation of contaminants after the EC unit up to 4% of NH_3–N, 9.3% of BOD, and 7.8% of COD. In addition, using the SF improved the turbidity removal to 42.6%. The WQ specifications of the treated MWW using the BEC cell were compared with the standard specifications for restricted and unrestricted agricultural irrigation water. The overall operating cost of MWW treatment for irrigation purposes by using a modified bio-integrated electrocoagulation method was 0.76 $\$m^{-3}$.

1. Introduction

Many countries around the world are already under tremendous water stress. With the impacts of climate change to cope with future ever-increasing water demands different countries and authorities are adopting variously augmented and/or sustainable water supply systems. On average, agriculture consumes 70% of freshwater worldwide (Hoekstra et al., 2009). In the Kingdom of Saudi Arabia (KSA), about 80% of its total water use is being used by the agricultural sector (Alhassan et al., 2016), out of which currently only 25% is satisfied from recycled/treated WW (Ouda et al., 2016). Treated WW can be a good source of irrigation water, not only through the augmented water supply but also a source of additional nutrients for the crops. To achieve United Nation's Sustainable Development Goals (SDGs), like many other countries KSA's national water strategy recommends treated wastewater (TWW) to be an integral part of its water resource planning and aims to use it as a major supply source for non-potable water demands (Aziz and Farissi, 2014). The reuse of WW becomes an integral part of water demand management, it promotes the protection of high-quality fresh water and reduces both environmental pollution and overall supply costs (Shakir et al., 2017). However, despite such strong needs and national interest, a wider scale adoption of reusing treated WW is yet to be achieved. Some other countries have progressed a lot in this aspect. Especially, in Australia, many cities are recycling 100% of their WW (HAL, 2006).

One of the major impediments to wider-scale adoption of recycling WW is the remaining contaminants present in the WW even after typical treatments. Moreover, there are concerns about potential alterations of the physical properties of soil after the application of TWW. Urbano et al. (2017) have evaluated the changes in physical, chemical, and microbiological characteristics of a Dusky Red Latosol. They have investigated both the yield and quality of lettuce produced on that soil through irrigation with TWW. It was observed the application of TWW did not damage the physical properties of soil and increased the concentration of some soil nutrients (K, Ca, H, A*l*, and S). The presence of E. Coli bacteria was not observed on lettuce leaves or in the soil. Finally, lettuce production (in terms of fresh weight) was higher in lettuce cultivated with TWW irrigation. Rezapour et al. (2019) investigated the accumulation of different heavy metals in the winter wheat crop irrigated by TWW. They have reported that irrigation with TWW led to a remarkable build-up of metal concentrations in the soil; cadmium (Cd) 178.2%, nickel (Ni) 105.1%, Copper (Cu) 66.4%, Zinc (Zn) 66.0%, and lead (Pb) 40.9%. However, the concentration of heavy metals was significantly greater in wheat roots than in shoots and grains. The maximum concentrations of Zn, Cu, Ni, Cd, and Pb in wheat grains were 3.20, 1.20, 0.52, 0.31, and 0.21 mg kg^{-1}, respectively. However, it was concluded that in terms of the potential health risks calculated as hazard quotients (HQ), except Cd all other heavy metals were within the safe limit regarding non-carcinogenic risks. It is also reported that the quality of irrigation water derived from the effluent of wastewater treatment plant (WWTP) fails to meet the agricultural WQ standards due to the presence of high concentrations of contaminants in the WWTP effluent (Kim et al., 2012). To avoid any potential health hazards different regulatory bodies such as World Health Organization (World Health Organization, 2006), US Environmental Protection Agency (US EPA, 2012) have outlined some criteria, guidelines, and standards for the reuse of TWW for irrigation. For KSA, in 2006 standards for WW reuse in agricultural irrigation were issued by the ministry of water and electricity (Al-Jasser, 2011).

Depending on the quality of the effluent of WWTP, appropriate treatment(s) will be necessary before the effluent can be reused for irrigation. On the other hand, the treatment should be easily achievable within the cost constraint, lest recycling TWW becomes more expensive than drinking water. The conventional WW treatment processes are energy-intensive; typically consuming around 3% of the total electricity produced in many developed countries (Khan et al., 2017). There is a need for more cost-effective methods to purify a wide range of polluted water on-site, and with minimal additives that are the key focus for sustainable water management.

Numerous researchers have recently focused on advanced oxidation methods for the treatment of wastewater such as photocatalysis (Lin et al., 2021; Liu et al., 2021), photo-electrocatalysis (Kaur et al., 2021; Gao et al., 2021), sono-photocatalytic (Ding et al., 2021); electrocatalysis (Kaur et al., 2018). These methods provide very good degradation efficiency, but the major drawback of these methods is the generation of harmful secondary pollutants. However, the electro-coagulation (EC) process has not produced any harmful secondary pollutants and avoids the addition of chemicals into the wastewater for treatment. EC process is an electrochemical means of introducing coagulants and removing suspended solids, colloidal material, and metals, as well as other dissolved solids from water and WWs (Garcia-Segura et al., 2017). EC is an emerging technology that combines the functions and advantages of conventional chemical coagulation, chemical flotation, and electrochemistry in water and WW treatment (Vepsäläinen et al., 2011; Kuokkanen, 2016; Sillanpää et al., 2018). EC is a well-established water treatment technology widely applied for the treatment of drinking water supplies, as well as municipal and industrial WWs (Al-Shannag et al., 2013; Demirci et al., 2015) from organic, inorganic, and biological pollutants such as pesticides, dyes, heavy metals, biological pathogens, etc.

EC methods have been in use for WW treatment for more than a decade. EC method is very effective for the treatment of persistent wastewater pollutants as compared to the traditional methods (Sahu et al., 2014; Islam, 2019). EC methods solely are unable to provide treatment up to the required standard by the regulatory authority, an additional component/layer of treatment is applied to achieve desired treatment efficiency, especially for typical pollutants such as heavy metals and microorganisms. Integration of specialized additive layered biofilms with EC system acts as a biofilter and renders additional benefit to EC techniques. The submerged media with promoted biofilm on the surface first removes the pollutants through adsorption, then through degradation of some pollutants by the biofilm (Hasan and Muhammad, 2020). The modified EC cells can provide desired treatment efficiencies (Qian et al., 2018).

Therefore, a detailed investigation is performed with the aim of achieving higher treatment efficiency with a modified BEC cell, which is not yet reported in the literature. The present investigation deals with an experimental study on the effectiveness of a modified BEC for the removal of several contaminants such as turbidity, hardness, conductivity, total dissolved solids (TDS), total suspended solids (TSS), chloride (Cl^-), ammonia nitrogen (NH_3–N), biological oxygen demand (BOD), chemical oxygen demand (COD) and total coliform. The effect of fixed-film BF and SF on the treatment efficiency was also observed along with the operating cost calculations.

2. Materials and methods

In this study, a modified version of the earlier used BEC cell was used for the pollutant's removal investigation. Dimensions of the chambers of the original version were enlarged to a total volume of approximately 3 L. Also, a SF chamber was added. Scoria gravels washed with water and hydrochloric acid were used as BF media. Two Ti-based *Al* pieces with dimensions of 10 cm(W) × 8 cm(L) × 1 cm(T) was used as electrodes with a 5 cm spacing between the electrodes. Experiments were performed under different conditions of applied voltage (ranging from 22 to 30 V), inlet flowrate (ranging from 1 to 5 Lh^{-1}), and pH (ranging from 3 to 9). SI Table 1 shows the list of exact variables used with their identification symbols. All the BEC experiments were carried out in a bench-scale plant, at room temperature with water temperature varying from 25 to 30 °C. A constant flow rate was maintained by the peristaltic pump which connected to a 25 L tank of fresh WW collected directly downstream of the mechanical bar screener chamber from WWTP at King Saud University (KSU). A DC linear power source with constant voltage during treatments was applied. Influent and effluent samples were collected in rinsed bottles with 76% ethanol and distilled water. A schematic experimental setup is shown in SI Fig. 1. SI Table 2 shows the combinations of treatment conditions used in the current study.

The removal efficiencies (R %) have been calculated according to the following Eq. (1)

$$R\,\% = \frac{(C_0 - C_1)}{C_0} \times 100 \tag{1}$$

where, C_0 and C_1 are pollutant concentrations before and after both the EC and BEC treatments, respectively.

Turbidity was measured using a Turbidity meter according to Nephelometric Method (2130 B) (APHA23rd, 2017). Hardness was measured according to Ethylenediaminetetraacetic acid (EDTA) Titrimetric Method (2340 C) (APHA23rd, 2017). EDTA was used as titrant for such a method. The calculation of hardness is as follows (Eq. (2)):

$$\text{Total Hardness } (mgl^{-1} \text{ as CaCO3}) = \frac{A \times B}{V} \times 1000 \tag{2}$$

where, 'A' is the volume of EDTA titrant used (mL), 'B' is the mg CaCO3 Equivalent to 1.00 ml EDTA titrant (= 1 mg $CaCO_3$), 'V' is the volume of the tested sample (mL).

Conductivity was measured using the Instrumental Method (2510 A) (APHA23rd, 2017). TDS and TSS were measured according to Laboratory Method (2510 B) (APHA23rd, 2017). Chloride was measured according to Mercuric Nitrate Method (4500- Cl^{-} C) (APHA23rd, 2017). The pH value was measured according to Electrometric Method (4500-H + B) (APHA23rd, 2017). The ammonia-nitrogen was measured according to the Titrimetric Method (4500-NH3 C) (APHA23rd, 2017). The 5-Day BOD was measured using a modified Iodometric Method, Azide Modification (4500-O C) (APHA23rd, 2017). The COD was measured according to Open Reflux Method (5220 B) (APHA23rd, 2017). The total coliform was measured according to the Standard total coliform membrane filter procedure using Endo media, Method (9222 B) (APHA23rd, 2017).

For most of the studies on water/WW treatment by EC, optimization is performed through varying a single factor while keeping all other factors fixed at a specific set of conditions (Hakizimana et al., 2017). This traditional way of optimization of EC process requires many experimental runs and often causes poor optimization, such as underestimation or overestimation of the effect of the process parameters on EC performances due to non-consideration of the interactions among those variables. To overcome such problem, a factorial design (FD) analysis was applied using SAS 9.2 software to determine the principal effects of independent variables and their interaction according to the General Linear Model (GLM) as shown in the following Eq. (3).

$$Y_{ijk} = \mu + \alpha_i + \beta_j + (\alpha\beta)_{ij} + \varepsilon_{ijk} \tag{3}$$

where, 'Y_{ijk}' is the outcome k from level i (factor A) and level j (factor B), 'μ' is the grand mean, 'α_i' is the contribution of the ith level of factor A, 'β_j' is the contribution of the jth level of factor B, '$(\alpha\beta)_{ij}$' is the combined contribution of the ith level of factor A and the jth level of factor B, and 'ε_{ijk}' is the contribution of the kth individual, which is often called the "error".

3. Result and discussion

3.1. *WW and physicochemical characterization*

The WW used in the present study was collected freshly from the WW treatment plant affiliated with KSU directly after the mechanical bar screen chamber. The collected WW contains a wide and complex mixture of organic and inorganic compounds. The physicochemical characteristics of the raw WW that were used in the experiments have been presented in Table 1.

3.2. *Experimental*

Assessment of WW quality is necessary when reusing the treated WW for agricultural irrigation (restricted or unrestricted). The efficiency of contaminants removal using the BEC cell was studied by analyzing effluent samples after applying different levels of the studied factors (Voltage "22, 26, and 30 V", Inlet Flowrate"1, 3 and 5 Lh^{-1}" and pH_i value "3, 5, 7, 7.4, and 9"). The parameters pertinent to this study included turbidity, hardness, conductivity, TDS, and TSS which represented the physical and aggregate properties. Chloride and ammonia nitrogen were represented as the inorganic nonmetallic constituents. BOD and COD have represented the aggregate organic constituents. Total Coliform was represented in the microbiological examination. Hence, a BEC cell was used, where the raw WW is exposed to the EC process, which is a variant of the conventional coagulation process in which coagulant agents are generated in situ through the dissolution of a sacrificial anode by applying current between the anode-cathode electrodes followed by fixed biological filter and filtered with SF.

3.3. *Effect of BEC Parameters on Selected Responses*

3.3.1. *Effect of Cell Voltage and Inlet Flowrate*

Current density applied at electrodes, and inlet flow rate values in a

Table 1

Characteristics of WW from KSU treatment plant.

Parameter	Influent	Typical Concentration		
		Low Strength	Medium Strength	High Strength
Turbidity	98 ± 31	–	–	–
Hardness	338 ± 56	<50	50–300	>300
Conductivity	1212 ± 15	–	–	–
TDS	869 ± 20	270	500	860
TSS	136 ± 16	120	210	400
Chlorides	309 ± 11	30	50	90
pH	7.4 ± 0.2	–	–	–
Ammonia Nitrogen	36.5 ± 08	12	25	50
BOD	41 ± 9.6	110	190	350
COD	150 ± 43	250	430	800
Total Coliform	62933 ± 70647	10^6–10^8	10^7–10^9	10^7–10^{10}

All samples were collected at 07:00 Am, units are in mgL^{-1}, except pH, conductivity (μmho cm^{-1}), coliform MPN100 mL^{-1}, and turbidity (NTU), Due to the Covid-19 pandemic, the organic loads were low due to the suspension of most of the university's activities during that period.

continuous EC system are the vital parameters influencing the performance and economy of the electrocoagulation process. The observed removal efficiencies for turbidity, hardness, conductivity, TDS, TSS, chloride, ammonia nitrogen, BOD, COD, and total coliform were measured at variable operating conditions of current densities and flowrate. The results show the favorable effect of current density and flow rate on the EC process. The results of the effect of operating parameters on the bench-scale EC process are shown in Fig. SI 2 and 3. The applied voltage is expected to exhibit a strong effect on the EC process. The supply of voltage to the EC system determines the amount of Al^{3+} ion released from the respective electrodes and the quantity of resulting coagulant. Thus, more Al^{3+} ions get dissolved into the solution, and the formation rate of $Al(OH)_3$ is increased. Also, it is well known that electrical potential not only determines the coagulant dosage rate but also the bubble production rate and size and the flocs growth which can influence the treatment efficiency of the electrocoagulation process (Bazrafshan et al., 2013).

3.3.1.1. Turbidity removal. Turbidity removal was significantly changed due to the change in applied cell voltage in conjunction with the change of inlet flowrate. SI Fig. 2a shows that applying a 22 V was able to decrease the turbidity concentration by 74.8% and increasing cell voltage up to 26 V increased removal efficiency to 78.8%. However, an increase in cell voltage to 30 (V) negatively affects turbidity removal. The most effective removal efficiency was noted at 26 V with 1 Lh^{-1} with 78.8% removal efficiency varied by Duncan's test (Table 2). Turbidity removal occurs because of the destabilization of colloids by the electric field generated between the electrodes (Verma and Kumar, 2018).

3.3.1.2. Removal Hardness, Conductivity (TSS), and Ammonia Nitrogen. Using different voltage values (22, 26, and 30 V), and 1, 3, and 5 Lh^{-1} as inlet flowrate without any pH_i adjustment, has affected the removal of hardness (SI Fig. 2b), conductivity (SI Fig. 2c), TSS (SI Fig. 2d), and ammonia nitrogen (SI Fig. 2e) ranged (15.1–57.1%), (6.7–28.9%), (63–98.7%), and (11.1–28.2%), respectively. It is noteworthy that the higher the applied voltage leads to higher the removal efficiencies, since practically low removal from the effluent when 22 V was applied for all inlet flowrates and very low removal when inlet flowrate was 5 Lh^{-1} for all applied voltages. The ANOVA analysis of variance (SI Table 3) shows that change in applied voltage has a significant effect on conductivity, TSS, and ammonia nitrogen removal ($Pr < 0.0001$) but was not significant in case of hardness removal ($Pr > 0.05$). The change in inlet flow rate has a significant effect on hardness, conductivity, TSS, and ammonia nitrogen removal ($Pr < 0.0001$). Furthermore, the interaction between applied voltage and inlet flowrate has significant effect on TSS ($Pr < 0.05$), on the contrary the interaction was not significant in hardness, conductivity, and ammonia nitrogen ($Pr > 0.05$). The Duncan's test for hardness (Table 2) shows that the most effective removal efficiency was noted at 1 Lh^{-1} with each 22, 26, and 30 V giving (49.2, 56.8, and 57.1%) removal efficiencies, respectively. In case of conductivity, the most effective removal efficiencies of TSS were noted at 1 Lh^{-1} with both 26 and 30 V giving (28.4% and 28.9%), (98.3% and 98.7%) removal efficiencies, respectively. Moreover, for ammonia nitrogen (Table 3), the most effective removal efficiency was noted at 1 Lh^{-1} and 30 V with 28.2% removal efficiency.

Table 2
Duncan's multiple range Test for physical & Aggregate Properties.

	Flow (Lh^{-1})	Voltage (V)		
Turbidity		22	26	30
	1	$74.8 \pm 0.5^{B,a}$	$78.8 \pm 0.6^{A,a}$	$66.9 \pm 0.3^{C,a}$
	3	$40.7 \pm 2.5^{B,b}$	$44.1 \pm 0.5^{A,b}$	$35.6 \pm 1.6^{C,b}$
	5	$20.5 \pm 1.4^{B,c}$	$25.1 \pm 0.2^{A,c}$	$16.8 \pm 1.3^{C,c}$
Hardness				
	1	$49.2 \pm 1.7^{A,a}$	$56.8 \pm 2.5^{A,a}$	$57.1 \pm 1.6^{A,a}$
	3	$29.5 \pm 2.2^{A,b}$	$36.4 \pm 1.8^{A,b}$	$36.8 \pm 0.9^{A,b}$
	5	$15.1 \pm 2.5^{A,c}$	$17.2 \pm 2.3^{A,c}$	$18.1 \pm 1.6^{A,c}$
Conductivity				
	1	$23.3\pm1^{B,a}$	$28.4 \pm 1.3^{A,a}$	$28.9 \pm 0.5^{A,a}$
	3	$15 \pm 1.3^{B,b}$	$19.8 \pm 1.3^{A,b}$	$21.1\pm2^{A,b}$
	5	$6.7 \pm 1.5^{B,c}$	$8.7 \pm 2.4^{A,c}$	$10.1 \pm 2.8^{A,c}$
TDS				
	1	$30.9 \pm 1.8^{B,a}$	$37.4 \pm 1.3^{A,a}$	$34 \pm 1.7^{A,a}$
	3	$22.6 \pm 3.3^{B,b}$	$26.8\pm2^{A,b}$	$25.8 \pm 0.8^{A,b}$
	5	$19.3 \pm 3.6^{B,c}$	$22.3 \pm 1.3^{A,c}$	$20.1 \pm 3.2^{A,c}$
TSS				
	1	$90.6 \pm 3.6^{B,a}$	$98.3 \pm 0.8^{A,a}$	$98.7 \pm 2.2^{A,a}$
	3	$84.8 \pm 3.8^{B,b}$	$87 \pm 2.6^{A,b}$	$84.9 \pm 3.7^{A,b}$
	5	$63 \pm 0.9^{B,c}$	$80.4 \pm 2.8^{A,c}$	$74.9 \pm 2.9^{A,c}$

All Values in the form of (mean ± SD). Same capital letters mean no significant difference between voltage levels, same small letters mean no significant difference between flow levels at $Pr < 0.05$.

3.3.1.3. TDS and Chloride. The relationship between the removal percentages of TDS and Cl^- with the applied voltages at various inlet flowrates are depicts in Fig. SI (2f, 3a), TDS and Cl^- concentration was found to be decreasing with the increase in applied voltage and decrease in inlet flowrate. Although the TDS removal efficiencies was less as compared to turbidity and TSS, since the hydrogen gas liberated at the cathode helped to float the dissolved solids on surface of water (Nawarkar and Salkar, 2019). The ANOVA analysis of variance (SI Table 3) shows that both change in applied voltage and inlet flowrate has a significant effect on TDS and Cl^- removal, while the interaction between applied voltage and inlet flowrate was not significant for TDS ($Pr > 0.05$) but significant for Cl^- ($Pr < 0.0001$). Although the TDS removal efficiency started to decay after raising the applied voltage from 26 to 30 V, the Duncan's test (Table 2) shows that the most effective removal efficiency was noted at 1 Lh^{-1} with both 26 and 30 V giving 37.4% and 34% removal efficiencies, respectively. But for Cl^-, the Duncan's test (Table 3) shows that the most effective removal efficiency was noted at 1 Lh^{-1} with 26 V only giving 27.6% removal efficiency. It is reported that by raising the applied voltage beyond the optimum value led to the production of a high number of hydroxyl radicles, cause a rise in the pH of the solution (A Al-Raad et al., 2019). It is well known that the coagulant breaks down in high pH solutions, ultimately leads to a reduction in the performance of the EC process.

3.3.1.4. BOD and COD removal. The highest decomposition of BOD (SI Fig. 3b), and COD (SI Fig. 3c) was obtained at a current density of about 26 V, over which the destruction of pollutants had the same trends and starts to decay, because the degradation of pollutants at the electrode surface was hindered by the hydroxyl radicals and produce gases. This trend could be rationalized considering that increasing voltage at the *Al* anode may increases its dissolution, according to Faraday's law. The increase in the amount of dissolved *Al*, will lead to the formation of *Al* hydroxides resulting from the release of metal ions from anode and HO^- from cathode (Cañizares et al., 2009). Concurrently, it was observed that the decrease in the inlet flowrate from 5 to 1 Lh^{-1} increase in the BOD and COD removal from 60.8% to 78% and 66.7%–81% respectively at 26 V cell Voltage. It is worth noting that the highest depletion of both BOD and COD concentration was achieved at 1 Lh^{-1} as a function of hydraulic detention time for all applied voltages. The ANOVA analysis of

Table 3
Duncan's multiple range Test for physical & Aggregate Properties.

	Flow (Lh^{-1})	Voltage (V)		
Chloride		22	26	30
	1	$17.6 \pm 0.6^{C,a}$	$27.6 \pm 0.6^{A,a}$	$23.7 \pm 1.3^{B,a}$
	3	$12.2 \pm 1.5^{C,b}$	$18.2 \pm 0.5^{A,b}$	$16.5 \pm 0.6^{B,b}$
	5	$10.4 \pm 0.6^{C,c}$	$13.1 \pm 0.6^{A,c}$	$12.3 \pm 0.6^{B,c}$
Ammonia Nitrogen				
	1	$20.6 \pm 2.1^{C,a}$	$26.7 \pm 0.9^{B,a}$	$28.2 \pm 2^{A,a}$
	3	$17 \pm 2.2^{C,b}$	$22.8 \pm 0.4^{B,b}$	$24.5 \pm 0.7^{A,b}$
	5	$11.1 \pm 0.9^{C,c}$	$14.4 \pm 0.3^{B,c}$	$15.6 \pm 1.2^{A,c}$
BOD				
	1	$66.7 \pm 2.9^{C,a}$	$78 \pm 2^{A,a}$	$73.3 \pm 3.3^{B,a}$
	3	$54.4 \pm 1.9^{C,b}$	$65.8 \pm 3.8^{A,b}$	$61.1 \pm 5.1^{B,b}$
	5	$47.8 \pm 1.9^{C,c}$	$60.8 \pm 1.4^{A,c}$	$65.7 \pm 3.3^{B,c}$
COD				
	1	$71.7 \pm 2.9^{C,a}$	$81 \pm 1.2^{A,a}$	75 ± 3.6 B,a
	3	$60.3 \pm 2.2^{C,b}$	$72.6 \pm 2.1^{A,b}$	$70.4 \pm 4.2^{B,b}$
	5	$53.8 \pm 3.8^{C,c}$	$66.7 \pm 3.3^{A,c}$	$61.3 \pm 2.3^{B,c}$
Total Coliform				
	1	$99.9 \pm 0^{A,a}$	$99.9 \pm 0^{A,a}$	$99.9 \pm 0^{A,a}$
	3	$98.8 \pm 0.1^{A,b}$	$98.9 \pm 0.1^{A,b}$	$98.8 \pm^{A,b}$
	5	$98.5 \pm 0^{A,c}$	$98.6 \pm 0.2^{A,c}$	$98.6 \pm 0.1^{A,c}$

All Values in the form of (mean ± SD). Same capital letters mean no significant difference between voltage levels, same small letters mean no significant difference between flow levels at $Pr < 0.05$.

variance (SI Table 3) shows that both changes in applied voltage and inlet flowrate have a significant effect on BOD and COD removal ($Pr < 0.0001$), despite that the interaction between applied voltage and inlet flowrate was not significant ($Pr > 0.05$). The Duncan's test (Table 3) shows that the most effective removal efficiencies were noted at 26 V and 1 Lh^{-1} with 78% and 81% removal efficiencies of BOD and COD, respectively.

3.3.1.5. Total Coliform removal. EC cell was able to decontaminate the studied WW from total coliform by more than 98% for all operating voltages and inlet flowrates (SI Fig. 3d). Ghernaout et al. (2019) stated that, the vital physiological functions of bacteria are programmed into the cell membrane, cytoplasm, and nucleic acids (DNA and RNA). Therefore, harm to any such sub cellular components of bacteria may cause the deactivation of the bacteria. For disinfection, chlorination is the most widely used technique in water treatment, but there are many significant drawbacks in water reuse which cannot be easily overcome i.e., production of hazardous by-products such as organochlorinated (Barrera-Díaz et al., 2015). This significant abatement was attributed to the bacteria adhesion on the surface of the electrogenerated floc. This treatment level has been achieved by Barrera-Díaz et al. (2015) as the researchers used the EC for microbiological disinfection from industrial WW using copper electrodes, the process was effective in disinfection, demonstrating a 99% reduction in total coliform, fecal coliform, *E. coli*, and anaerobic *mesophilus*. EC disinfection seemed to possess a germicidal performance even bigger than ozonation in terms of residence period (Ghernaout, 2017). The ANOVA analysis of variance (SI Table 3) shows that the change in applied voltage has no significant effect on total coliform removal efficiency ($Pr > 0.05$), but at the same time the change in inlet flowrate has a significant effect on total coliform removal ($Pr < 0.0001$). In addition, the analysis shows that the interaction between applied Voltage and inlet flowrate has no significant effect on total coliform removal efficiency with ($Pr > 0.05$). The Duncan's test for total coliform (Table 3) shows that the most effective removal efficiency was noted at 1 Lh^{-1} with each 22, 26, and 30 V giving 99.91, 99.90, and 99.90% removal efficiencies, respectively.

3.3.1.6. WQ Test. There were many factors were chosen to judge the WQ after EC process, and to choose the most effective synthesis of applied voltage and inlet flowrate; a global quality index (GQI) has been applied to specify the proper synthesis. GQI is a scale from zero to one which can be applied on normalized data. The closer the value to one, the better the synthesis. The GQI is calculated according to Eq. (4).

$$GQI = \frac{\sum_{1=1}^{N} \widehat{x_i} \cdot w_i}{N} \tag{4}$$

where $\widehat{x}_i$ is the normalized value of quality parameter x, w_i is the weight factor of the quality parameter, N is the number of parameters and GQI is the global quality index. As mentioned, once the data were normalized, the weight factors for all variables were equal, so the GQI was calculated according to Eq. (5).

$$GQI = \frac{\sum_{1=1}^{N} \widehat{x_i}}{N} \tag{5}$$

As expected, the highest values were located at the synthesis which has the lowest inlet flowrate, as shown in SI Fig. 3e the synthesis (A_2B_1) was the closer synthesis to one with 0.61 GQI value. Besides, the Duncan's test for GQI (SI Table 4) shows that the synthesis (A_2B_1) was significantly higher than other syntheses. Thus the (A_2B_1) synthesis which represents 26 V and 1 Lh^{-1} has been chosen as the most effective synthesis and was used to study the influence of pH_i variation on EC process.

3.3.2. Effect of pH_i

The pH_i of the WW could have positive or negative effect on the treatment performance, as it affects the stability of various hydroxide species formed in the system. Likewise, changes in the pH_i values can change the surface charge of the particles, and this has the effect of removing the colloidal dispersed organics from solution. Laboratory experiments were performed by changing pH_i values to investigate the effects of pH_i of WW on EC process. During the experiments, only the pH_i was set and was no further controlled. The BEC cell has been operated

Table 4
Duncan's multiple range Test for initial pH variations.

Parameters	Initial pH Value				
	3	5	7	7.4	9
Turbidity	14.8 ± 2.3^{E}	46.8 ± 1.6^{D}	81.3 ± 0.5^{A}	78.8 ± 0.6^{B}	52.7 ± 0.2^{C}
Hardness	18.9 ± 1.8^{E}	33.6 ± 2.7^{D}	52.1 ± 3.4^{C}	56.8 ± 2.5^{B}	69.4 ± 1.8^{A}
Conductivity	14.6 ± 3^{C}	19.9 ± 2^{B}	28.3 ± 0.5^{A}	28.4 ± 1.3^{A}	31.2 ± 1.3^{A}
TDS	16.9 ± 1.5^{D}	23.7 ± 1.3^{C}	39.4 ± 0.2^{A}	37.7 ± 1.3^{A}	30.5 ± 0.8^{B}
TSS	71.2 ± 1.4^{D}	83 ± 5^{C}	99.1 ± 0.8^{A}	98.3 ± 0.8^{A}	89.7 ± 0.9^{B}
Chloride	13.2 ± 0.9^{D}	18.5 ± 0.5^{C}	27.3 ± 1^{A}	27.6 ± 0.6^{A}	25.5 ± 0.8^{B}
Ammonia Nitrogen	10.6 ± 0.7^{D}	18.5 ± 1.1^{C}	25.8 ± 1.7^{B}	$26.7 \pm 0.9^{A,B}$	29.1 ± 2^{A}
BOD	38.3 ± 6^{C}	62.8 ± 2.5^{B}	81.8 ± 1.7^{A}	78 ± 2^{A}	65.3 ± 2.1^{B}
COD	46.4 ± 2.4^{D}	60.3 ± 2.1^{C}	82.2 ± 0.7^{A}	81 ± 1.2^{A}	68 ± 1.5^{B}
Total Coliforms	99.9 ± 0.0^{A}	99.9 ± 0.0^{A}	99.9 ± 0.0^{B}	99.9 ± 0.0^{B}	99.9 ± 0.0^{A}

All Values are in the form of (mean ± SD). Same capital letters mean no significant difference between initial pH values.

under the optimal applied voltage of 26 V and inlet flowrate of 1 Lh^{-1} from the previous experimental stage with the constant factors (*Al* electrodes, 5 cm electrode spacing). Five different pH_i values were applied (3, 5, 7, and 9). The contaminants removal trends were similar, for turbidity (SI Fig. 4a), TDS (SI Fig. 4f), TSS (SI Fig. 4d), Cl^- (SI Fig. 5a), BOD (SI Fig. 5b), and COD (SI Fig. 5c). The removal of these contaminants was low at the acidic condition $pH_i = 3$. This phenomenon can be explained by the distribution of *Al* ions. Different hydroxides formed during EC were affected by WW pH_i, the surface charge of particles can also be modified by the change in pH, which influence the contaminants removal ability. In addition, useful *Al* ions for contaminants removal need to be stable and insoluble. According to predominance zone diagram for *Al*(III), Al^{3+} and $Al(OH)^{2+}$ are predominant under acidic conditions while $Al(OH)^{4-}$ ions are predominant under alkaline conditions and these ions are soluble. As a result, the solubility of these species is not effective for contaminants removal. On the other hand, at neutral pH floc of $Al(OH)_3(s)$ have large surface areas, which is useful for rapid adsorption of soluble organic compounds and trapping of colloidal particles. In addition, the pollutants removal efficiencies were found to be the best near neutral pH using Al electrode (Chen, 2004; Le et al., 2021).

The raising the pH_i, the removal efficiency has increased from (14.8–46.8) %, (16.9–23.7) %, (71.2–83) %, (13.2–18.5) %, (38.3–62.8) %, (46.4–60.3) % for turbidity, TDS, TSS, Cl^-, BOD, and COD, respectively. The highest decomposition has been achieved at neutral phase where it reached to (81.3, 39.4, 99.1, 27.3, 81.8, 82.2) % for turbidity, TDS, TSS, Cl^-, BOD, and COD, respectively. But raising the pH_i beyond the neutral has caused a decay in the removal efficiency. These results were in accordance with (Al-Raad et al., 2020) as both acidic and alkaline conditions considerably decreased the contaminants removal performance of the system because of the increase in solubility of generated $Al(OH)_3$. For hardness (SI Fig. 4b), conductivity (SI Fig. 4c), and ammonia nitrogen (SI Fig. 4e), the removal of these contaminants was low at the acidic condition $pH_i = 3$, and by raising the pH_i from 3 to 7, the removal efficiency has increased from (18.9–52.1) %, (14.6–28.3) %, (10.6–25.8) %, respectively. On the contrary of the other contaminants, the removal efficiency of these contaminants did not decay but kept going up as it reached to (69.4, 31.2, 29.1) %, respectively at $pH_i = 9$. The increase in pH_i has led to an increase in contaminants removal efficiency (Zhao et al., 2014; Helmy et al., 2017) but the increasing pH_i from 9 to 11 has no significant decrease of contaminants removal efficiency (Le et al., 2021).

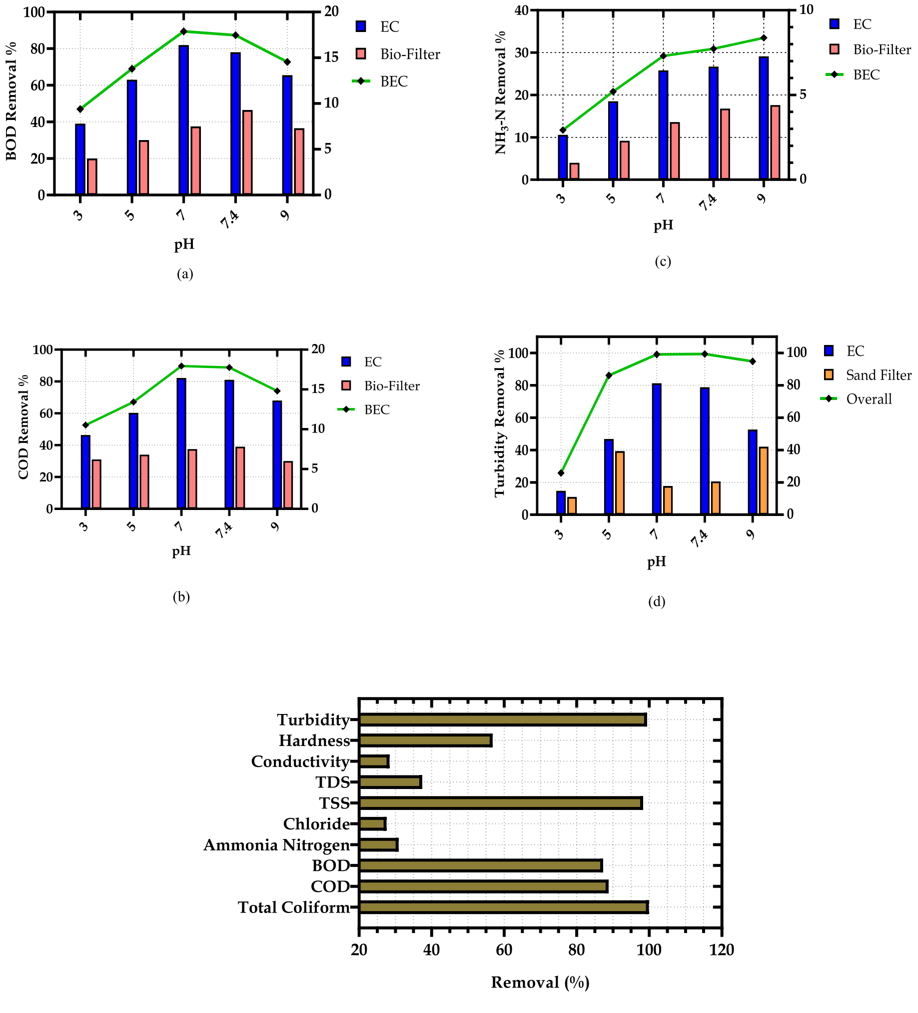
Furthermore, total coliform removal by applying different pH_i values was noticed to be very close in all treatments, although at acidic and alkaline cases the removal efficiency was relatively higher than the $pH_i = 7$ and the original pH_i of WW as shown in SI Fig. 5d. The EC inactivation of microbes contributes in several stages as follows (a) cell death due to electrochemically produced antimicrobial agents (oxidants) which means that, cell destruction occurs by either the stable pores formation resulting destabilization of cell membranes, lose their essential cellular constituents and destruction of chemical gradients by oxidants transport through transient pores; (b) irreversible permeabilization of cell membrane by the applied electric field and (c) direct electrochemical oxidation of important cellular components by external electric field (Govindan et al., 2018). So, the synergistic action of both microbial agents and electric field attribute to microbe inactivation process. The direct and indirect effect of electrolysis on microbes during EC cannot be separated (Drogui et al., 2001). pH_i values from 5 to 8.5 provides an increase in removal of total coliform and total bacteria from 97.9 to 99.8 and 97.2–99.7% respectively (Chopra and Sharma, 2014). They indicated that the removal efficiency of total coliform with an increase in the pH_i from 5 to 7. In present study, the increase in pH_i of municipal WW beyond 7 decreased the removal efficiency of total coliform and total bacteria. The variation in pH impact the solubility of metal hydroxides, ultimately affect the inactivation of microbes (Chen and Hung, 2007).

The ANOVA analysis of variance (SI Table 5) shows that changing in pH_i was significant ($Pr < 0.0001$), for turbidity, hardness, conductivity, TDS, TSS, chloride, ammonia nitrogen, BOD, COD, and total coliform. According to the Duncan's test for the studied parameters (Table 4), the highest removal efficiency for turbidity was achieved at $pH_i = 7$ with 81.3%, and for hardness at $pH_i = 9$ with 69.4%. But for TDS, TSS, Chloride, BOD, and COD the highest removal efficiencies was noticed at both $pH_i = 7$ and 7.4 with (39.4 and 37.7) %, (99.1 and 98.3) %, (27.3 and 27.6) %, (81.8 and 78) %, (82.2 and 81) %, respectively. In case of conductivity, the highest removal values were at $pH_i = 7$, 7.4, and 9 with (28.3, 31.2, and 28.4) %, respectively. On the other hand, the Duncan's test for ammonia nitrogen was more complex, the highest removal efficiency was achieved at both $pH_i = 7.4$ and 9 with (26.7 and 29.1) %, as there was no significant difference between them. However, there were no significant difference between the $pH_i = 7.4$ and $pH_i = 7$. Eventually, for total coliform the analysis showed that all studied pH_i values resulted in more than 99.9% removal of colonies. The most effective synthesis on contaminants removal which was (26 V and 1 Lh^{-1}) using the GQI technique. The influence of initial pH during EC process has been tested by applying several initial pH values with the chosen synthesis. Likewise, another GQI was made and plotted in SI Fig. 5e to choose the best applied pH_i. In addition, the electrical energy consumption was plotted to judge its application to practical reality.

Statistically $pH_i = 7$ provide maximum removal at optimized voltage and flow rate. The original pH_i of the MWW was 7.4. In present study, it was clear that the original pH_i (7.4) of MWW was not significantly different from $pH_i = 7$ for most of tested parameters such as COD, BOD, TDS, TSS, chloride, and conductivity. All tested parameters result with original pH_i did not exceed the maximum allowable limitations of the standards for restricted and unrestricted irrigation except NH_3–H which exceeded the limitations in both original pH_i and $pH_i = 7$. Moreover, the energy consumption (9.4 WhL^{-1}) while using the original pH_i was lower than the energy consumption (11.4 WhL^{-1}) at the $pH_i = 7$. According to the justifications it is recommended using the original pH_i of WW instead of the optimized pH_i at optimized applied voltage and inlet flowrate.

3.3.3. *Effect of BF*

The attached microbial growth process (fixed film biological process) was selected to be use in this study, consisting in a bioreactor where the microorganisms responsible for the conversion of organic material by forming a biofilm, to an inert packing material. The packing was submerged completely in liquid or with air or space above the biofilm. The main advantages of this type of systems are the high resistance of the microorganisms to toxic compounds and the low biomass production, as compare to free culture processes (Pérez et al., 2016). Scoria rocks were used as a BF packing. The results showed that using EC has conducted in (38.3, 62.8, 81.8, 78, 65.3) % removal efficiency at 26 V and 1 Lh^{-1} with the studied pH_i values. By using the BF, the BOD removal efficiencies have increased to (47, 69, 89.4, 87.3, 72.7) %, respectively as shown in Fig. 1a. The *t*-test (Table 5) shows that using BF after EC treatment was significantly effective on BOD removal efficiency with P value = 0.0001. In case of COD (Fig. 1b), using EC followed by BF resulted in an increase of removal efficiencies from (46.4–52.6) %, (60.3–67.1) %, (82.2–89.7) %, (81.1–88.8) %, (68.1–74) %, respectively for investigated pH_i values. This increase was proven to have a significant effect on the enhancement of COD removal efficiency as noted from *t*-test, where P value was equal to 0.0001. The contribution of BF on the removal efficiency of ammonia nitrogen is relatively less than its contribution in BOD and COD removal. Despite this, the *t*-test (Table 5) shows that using a sequential EC and BF treatment was significantly effective on ammonia nitrogen removal efficiency with P value = 0.0076. Fig. 1c shows the effect of BEC cell on ammonia nitrogen where the removal increased from (10.6–11.7) %, (18.5–20.8) %, (25.8–29.2) %, (26.7–30.9) %, (29.1–33.5) %. Researchers concluded that hybrid EC is even capable of removing petroleum hydrocarbons (Mousa, 2016). The apparent low removal rates of



(a)

(c)

(b)

(d)

(e)

Fig. 1. a)Combined effect of BEC process on BOD removal at 26 V, Voltage & 1 Lh^{-1}, flow rate.b)Combined effect of BEC process on COD removal at 26 V, Voltage & 1 Lh^{-1}, flow rate.c)Combined effect of BEC process on ammonia nitrogen removal at 26 V, Voltage & 1 Lh^{-1}, flow rate.d)Combined effect of BEC process on turbidity removal at 26 V, Voltage & 1 Lh^{-1}, flow rate.e)Global effect of BEC cell at 26 V, Voltage & 1 Lh^{-1}, flow rate.

Table 5
Paired sample T-test between (EC-BEC) and (EC-SF).

parameter	EC	BEC	SF	R^2	P-Value
TD	54.9 ± 27.1	–	81.04 ± 31.3*	0.82	13.1 × 10^{-3}
NH_3–H	22.1 ± 7.5	25.2 ± 8.9**	–	0.86	7.6 × 10^{-3}
BOD	65.5 ± 16.8	73.1 ± 17.1***	–	0.98	1 × 10^{-3}
COD	67.6 ± 14.9	74.4 ± 15.5***	–	0.99	1 × 10^{-3}

ammonia nitrogen at the BF comparing to EC are due to the intermittent operation of the BEC cell that leads to eliminate the growth and activity of biota.

3.3.4. *Effect of SF*

The original BEC cell manufactured with cooperation technicians of Civil and Environmental Engineering Department, Northeastern University, Boston, MA, USA was not designed with SF stage. But the treatment results were not satisfying specially for turbidity which directly was affected due to the transference of scum and floc produced in EC chamber through the BF chamber to the effluent chamber. Leaving the effluent to settle down will violate the principle of continuity as a chosen operating mode. To overcome this issue, a SF chamber was added to the BEC cell. SF has a significant effect on turbidity removal (Fig. 1d) where the removal efficiency raised from (14.8–25.8) %, (46.8–86.1) %, (81.3–99.1) %, (78.8–99.4) %, and from (52.7–94.8) % for pH_i (3, 5, 7, 7.4, and 9) respectively. SF has participated in the global removal of turbidity by (11, 39.3, 17.8, 20.6, and 42.1) % in respect of the previous order of pH_i. It was observed, at $pH_i = 3$ the efficiency of SF was very low. This might be due to the high turbid concentration water produced from EC which caused a partial SF clogging and decreased its efficiency on blocking the scum and other impurities. When the efficiency of removing turbidity of the produced effluent from the EC chamber at a pH_i=(5 and 8) increased, the efficiency of SF increased too. At the optimum pH_i (7 and 7.4) the effluent produced from EC chamber has low turbid level - comparing to other pH_i values – thus, SF was able to block most of scum and impurities. The *t*-test shows that using SF after EC treatment was significantly effective on turbidity removal efficiency with P value = 0.01.

3.3.5. *Global effect of BEC cell*

The removal efficiencies for all studied parameters using the BEC cell under the optimum operating conditions of 26 V, 1 Lh^{-1}, and $pH_i = 7.4$ of studied real WW. Fig. 1e shows RWW sample from KSU WW treatment plant before and after treatment using the BEC cell.

3.4. Reusability of treated WW

The optimum values of aforementioned operating parameters, the treated WW quality was compared with WW reuse quality standards (SI Table 6). The comparison allows to judge the effectiveness of BEC process for the treatment of MWW to be utilized for landscape irrigation and plantation. The analysis shows that turbidity, hardness, conductivity, TDS, and TSS are within the allowable limits for standards. However, chloride slightly exceeds the limits of standards. The final pH value of treated WW is basic (8.1 ± 0.2) which is within the allowable limits. Similarly, BOD and COD are below allowable limits. Microbiologically, the total coliform analysis shows that after EC treatment their count became 663 MPN100 mL^{-1}. This ensures that the treated WW is microbiologically safe. Electrical conductivity and TDS have great importance in irrigation works. Higher electrical conductivity and TDS may damage the soil and become a potential irrigation problem. The effluent produced from BEC cell shows that the conductivity and TDS values of studied WW are coming under good class (Class 2) of irrigation water (SI Table 7). Investigation proves that the quality of treated WW is safe for landscape irrigation and plantation.

3.5. Operating cost

Operating cost (OC) of the EC process includes material, mainly electrodes and electrical energy costs, as well as labor, maintenance, sludge dewatering and disposal, and fixed costs. The latter cost items are largely independent of the type of the electrode material. In this study, energy and electrode material costs for calculation of the OC ($SARm^{-3}$) (Eq. (6)) were taken into account as major cost items (Kobya et al., 2016):

$$OC = aENC + bELC \tag{6}$$

where: ENC is energy consumption ($kWhm^{-3}$), ELC is electrode consumption (kgm^{-3}), while a and b were for prices in the Riyadh market (March 2021G, 0.16 $SARkWh^{-1}$)for agricultural electrical energy, 10 $SARkg^{-1}$ of commercial Al.

On the other hand, ENC and ELC were calculated from Eq. (7) and Eq. (8), respectively.

$$\text{ENC} \quad (\text{kwh/m3}) = \frac{U \times i \times t_{EC}}{Vol} \tag{7}$$

Where: U is cell voltage (V), i is current (A), t_{EC} is operating time (hour), Vol is volume (m^3) of the WW.

$$\text{ELC} \quad (\text{kg/m3}) = \frac{i \times t_{EC} \times Mw}{Z \times F \times Vol} \tag{8}$$

where: M_w is molecular mass of electrode (M_w, Al = 0.02698 $kgmol^{-1}$), t_{EC} is operating time (s), Z is number of electrons transferred (for Al = 3), and F is Faraday's constant (96.487 $Cmol^{-1}$). Then, the required energy amount under the optimum operating conditions in the EC process will be 9.9 $kWhm^{-3}$ of WW. Further, the consumption of Al electrodes was 0.13 Kgm^{-3}, this will cost 1.28 $SARm^{-3}$ of raw WW. Therefore, the cost of EC process per unit cubic meter shall be 2.9 $SARm^{-3}$ raw WW for agricultural purposes. For domestic and commercial use, the price for energy consumption raises from 0.16 to 0.18 and 0.2 $SARkwh^{-1}$ respectively, this will raise the OC to 3.06 and 3.26 $SARm^{-3}$ RWW, respectively.

4. Conclusions

Treatment of MWW using the modified BEC cell is promising to overcome the disadvantages of electrochemical and biodegradation treatment separately. However, raw wastewater is not suitable for direct reuse since its various quality parameters were above the standards. Therefore, running the BEC cell at a bench scale was utilized to determine the optimum operating conditions, and to bring the quality up to the required level. This study provided a very useful reference for the treatment of RWW by utilizing inert material electrodes followed by BF and SF. The influence of the BEC zone, bacterial efficiency, energy consumption and removal efficiency of turbidity, hardness, TDS, TSS, chloride, NH_3–N, BOD, COD, and total coliform were investigated and found that the BEC process achieved the removal of 78.8% of turbidity,56.8% of hardness, 28.4% of conductivity, 37.4% of TDS, 98.3% of TSS, 27.6% of chloride, 26.7% of NH_3–N, 78% of BOD, 81% of COD, and 99.9% of total coliform. The contribution of BF increased the pollutant removal to be 87.3% of BOD, 88.8% of COD, and 30.9% of NH_3–N, the use of SF increased the turbidity removal to 99.4%. The maximum removal efficiency was achieved when the applied voltage was 26 V with 1 Lh^{-1} as inlet flowrate. The pHi was found to be an important parameter for the EC process, the optimum pHi ranged from 7 to 7.4 with using Al electrodes. The energy consumption was 9.9 whL^{-1}, and the operating costs under the chosen synthesis were 0.76 $\$m^{-3}$ of MWW. The WQ specifications of the treated MWW using the BEC cell were compared with the standard specifications for restricted and unrestricted agricultural irrigation, and it was found that all the indicators of the studied WQ confirm the specifications except for chloride and NH_3–N. Therefore, for chloride and NH_3–N there is a need of further treatments to reach the water standards for agricultural irrigation.

Author statement

A.A-O: Conceptualization, Supervision, Investigation, Experimental, Data curation, Funding acquisition, Validation, Resources; **P.K**: Visualization, Manuscript—Writing – review & editing; **M.A.I**: Manuscript—Writing – review & editing; **M.E.H.I**, Experimental; **M.S**: Validation, Resources, Writing – review & editing, Funding acquisition; **M.A.M.K**: Supervision, Formal analysis.

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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.

Data availability

Data will be made available on request.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2022.135746.

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