

Dye removal and disinfection in one reactor by non-pulsed high voltage current using iron, aluminum and copper electrodes

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ABSTRACT

Electrocoagulation and high voltage disinfection are two effective methods to remove dye and micro-organisms. Existing electrocoagulation procedures use low voltage, less than 100 V, and disinfection procedures utilize pulsed high voltage current. This study investigates the capability of non-pulsed high voltage current to remove methylene blue (MB) and total coliform. A circuit is designed to transform 220 V AC current to 9–15 kV DC current without using capacitor, which may lessen the cost of circuit design. The impacts of electrode material, level of voltage, current density, and distance between electrodes are tested on the MB removal results and disinfection. The results show that dye and total coliform removals are 99% and 99.999998% (7.8 log) by using iron, 88% and 100% (9-log) by using copper, and 93% and 99.999998% (7.4 log) by using aluminum electrodes, respectively. Voltage has a direct impact on dye and coliform removal, and increasing the current density would increase the rate of reactions, but the distance between electrodes has an inverse impact on the rate of removal of MB and total coliform. For each type of electrodes, the effect of voltage amount, current density, distance between electrodes, and the duration of process are modeled by response surface method (RSM) with high precision. All models have R^2 values above 0.92. Both dye and removal processes are optimized with RSM in order to minimize the voltage amount, the current density, the time of process, the electrical energy consumption, and anode consumption.

Keywords: High voltage current; Dye removal; Total coliform; Disinfection; Electrocoagulation; RSM

1. Introduction

One of the major effluent contaminants of various industries such as paper, textiles, lather, plastics, cosmetics, and pharmaceuticals is dyes [1–5]. Wastewater pollutions are normally in the form of colloidal particles which are not removed by flotation, typical filtration, or sedimentation due to their stability in water. Colloids have microscopic size

(in the range of 1 nm to 10 μ m). A balance between the repulsive electrostatic force and the attractive van der Waals force causes the stability of colloids in a solution. At very short distances, the van der Waals forces of attraction of permanent or induced dipoles are significant. Coagulant reduces the repulsive energy between the particles. Therefore, a weak bond, the secondary minimum of potential energy, between particles could result in agglomeration of colloids more easily.

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Flocculation supports and enhances flocks properties such as settling ability and filterability [6–9].

Electrocoagulation is performed based on the dissolution of electrode material used as an anode. This so-called “sacrificial anode” produces metal ions, which act as coagulant agents in the aqueous solution.

During EC process, anode is consumed via dissolution. Faraday’s law dictates that in theory, amount of anode consumption is equal to [7,8,10]:

$$m_{\text{mental}}(\text{g}) = \frac{60 \times I \times t \times M}{z \times F} \quad (1)$$

where I is the applied current (A), t is the treatment time (min), M is the molar mass of the electrode material ($M_{\text{Fe}} = 55.845$ g/mol, $M_{\text{Al}} = 26.982$ g/mol, $M_{\text{Cu}} = 63.546$ g/mol), z is the valency of ions of the electrode material ($z_{\text{Fe}} = 2$, $z_{\text{Al}} = 3$, $z_{\text{Cu}} = 2$), and F is Faraday’s constant (96,485 C/mol).

It can be found that the anode consumption in practice is 105% to 190% more than the theoretical amount. This phenomenon is referred to as super faradaic efficiency [7].

Table 1 shows the basic reactions that occur in an electrocoagulation procedure [2,6–12].

The most common electrodes in experimental EC processes for dye removal comprise Fe and Al electrodes [12–15]. Copper has been also deployed recently as electrode in EC processes [9,16,17].

The most effective parameters in electrical treatment methods are voltage (V), current density (Cd), material of electrodes, surface of electrodes and distance between them (d), pH, time (t), temperature, electrical conductivity of solution and the initial concentration of pollution [7,8,12,13,18–21].

Previous research findings into the field of using electrical current in water/wastewater treatment indicate that most of electrical techniques are in low voltage, that is, under 100 V, for electrocoagulation [7,8,12,14,17,18].

Microorganisms are the other important part of water/wastewater contaminants. Many researchers worked on removing microorganisms (disinfection) by using pulsed high voltage currents, above 500 V, for disinfection purposes. These studies employed pulsed current to produce plasma by spark to remove microorganisms, using properties of plasma such as high temperature and pressure [9,22–26]. These studies used transformer systems, which consist of a transformer and a capacitor to transform urban electricity current into pulsed high voltage DC current.

There is a lack of data and experiments about removing dye and disinfection simultaneously. Therefore, in this paper, we intend to design a simple and low capital cost transformer system/circuit that transforms urban electricity current into non-pulsed high voltage current. Following benefits and contributions can be concluded for this study:

- It is not needed to use capacitor that is a delicate and expensive part of the electrical circuit.
- Unlike previous studies that used plasma properties produced by spark, in this study disinfection is accomplished without spark. This reduces the temperature pollutant.
- The system also has the electrocoagulation potential. As a result, disinfection and colloidal contaminant removal could happen in one reactor, simultaneously.

- This study examines the capability of non-pulsed high voltage current for wastewater treatment.

Previous studies have not dealt with the effect of different electrode materials on the disinfection process in detail. The effect of the properties of electrical system such as voltage, current density, electrode type, and time on the removal of Methylene blue dye and total coliform are also investigated in this study.

Finally, the response surface method (RSM) is employed to study and model the interaction between operational parameters and to optimize the efficiency of the processes, for each type of electrodes. These parameters are comprised of V , Cd , d , and t . RSM uses regression method for modeling [27–29].

Table 1
Reactions which occurred in an electrocoagulation procedure with Fe, Al [7,10,12], and Cu [9] electrodes

Iron electrode ^a	
Anode	$\text{Fe}_{(s)} \rightarrow \text{Fe}_{(aq)}^{2+} + 2e^{-}$
Bulk of solution	$\text{Fe}_{(aq)}^{2+} + 2\text{OH}_{(aq)}^{-} \rightarrow \text{Fe}(\text{OH})_{2(s)}$
Cathode	$2\text{H}_2\text{O}_{(i)} + 2e^{-} \rightarrow \text{H}_{2(g)} + 2\text{OH}_{(aq)}^{-}$
Overall	$\text{Fe}_{(s)} + 2\text{H}_2\text{O}_{(i)} \rightarrow 4\text{Fe}(\text{OH})_{3(s)} + 4\text{H}_{2(g)}$
Iron electrode ^b	
Anode	$4\text{Fe}_{(s)} \rightarrow 4\text{Fe}_{(aq)}^{2+} + 8e^{-}$
Bulk of solution	$4\text{Fe}_{(aq)}^{2+} + 10\text{H}_2\text{O}_{(i)} + \text{O}_{2(g)} \rightarrow 4\text{Fe}(\text{OH})_{3(s)} + 8\text{H}_{(aq)}^{+}$
Cathode	$8\text{H}_{(aq)}^{+} + 8e^{-} \rightarrow 4\text{H}_{2(g)}$
Overall	$4\text{Fe}_{(s)} + 10\text{H}_2\text{O}_{(i)} + \text{O}_{2(g)} \rightarrow 4\text{Fe}(\text{OH})_{3(s)} + 4\text{H}_{2(g)}$
Aluminum electrode	
Anode	$2\text{Al}_{(s)} \rightarrow 2\text{Al}_{(aq)}^{3+} + 6e^{-}$
Bulk of solution	$2\text{Al}_{(aq)}^{3+} + 6\text{H}_2\text{O}_{(i)} \rightarrow 2\text{Al}(\text{OH})_{3(s)} + 6\text{H}_{(aq)}^{+}$
Cathode	$6\text{H}_{(aq)}^{+} + 6e^{-} \rightarrow 3\text{H}_{2(g)}$
Overall	$2\text{Al}_{(s)} + 6\text{H}_2\text{O}_{(i)} \rightarrow 2\text{Al}(\text{OH})_{3(s)} + 3\text{H}_{2(g)}$
Cu electrode	
Anode	$\text{Cu}_{(s)} \rightarrow \text{Cu}_{(aq)}^{2+} + 2e^{-}$
Bulk of solution	$\text{Cu}_{(aq)}^{2+} + 2\text{H}_2\text{O}_{(i)} \rightarrow \text{Cu}(\text{OH})_{2(s)} + 2\text{H}_{(aq)}^{+}$
Cathode	$2\text{H}_{(aq)}^{+} + 2e^{-} \rightarrow \text{H}_{2(g)}$
Overall	$2\text{Cu}_{(s)} + 3\text{H}_2\text{O}_{(i)} + \frac{1}{2}\text{O}_{2(g)} \rightarrow 2\text{Cu}(\text{OH})_{2(s)} + \text{H}_{2(g)}$

^aMain reaction takes place at Fe electrodes.

^bFerrous iron may be oxidized to Fe^{3+} by atmospheric oxygen or anode oxidation [7,12].

2. Materials and methods

2.1. Contaminants

2.1.1. Methylene Blue

MB is one of the most common dyes in industries that is usually removed by absorption or electrocoagulation methods, as the most effective and low cost methods for dye removal [30–33]. The chemical structure of MB ($C_{16}H_{18}ClN_3S$) is shown in Fig. 1 [30,31].

DR5000 Hatch spectrophotometer is used to evaluate the MB concentration. The optimum wavelength would be 620 nm [30,34].

2.1.2. Total coliform

Various bacteria, parasites, and viruses, known to be pathogenic, have the potential to cause health problems, if they enter the human body. The Environmental Protection Agency (EPA) considers total coliform as an indicator of other pathogens in drinking water. Total coliform is used to determine adequate water treatment and integrity in the distribution system [35]. Total coliforms are group of bacteria commonly found in the environment. However, fecal coliform and *Escherichia coli* are derived from human and animal waste [36,37]. Many researchers consider the total and/or fecal coliform as a main indicator of quality of drinking water [35,37–39].

There exist several methods to determine the total coliform amount in water bodies. These methods require trained operators, sampling and culturing equipment, and the process takes more than a day to accomplish [36,40,41].

In this study, total coliform is collected from positive Brilliant Green solution and cultured on Agar media which

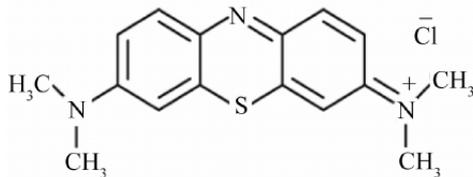


Fig. 1. Methylene blue chemical structure.

contains lactose. After total coliform growth on the media (Fig. 2), total coliform dissolves little by little in Ringer solution to a point that light absorbance reading of the solution reaches the 0.15 in wave length of 620 nm. The number of total coliform in this state will be equal to number of 10^9 (total coliform in 1,000 mL [42]. Properties of Ringer solution are indicated in Table 2.

5 mL of the bacteria containing Ringer solution are added to 495 mL of the bacteria-free solution that will undergo disinfection process. After disinfection process, the number of bacteria is evaluated by the most probable number (MPN) test [41].

2.2. Parameters

The most crucial parameter in electrocoagulation is the material of electrodes, and the most common materials for EC include aluminum, iron, and copper [9,12–17]. Electrodes with the different type of material enjoy the various potential for disinfection and copper seems to have the most significant effect on organism among three electrode materials: Al, Fe, Cu undergo a test here. Cu has a disinfection effect even without electricity current [44,45]. In the present study, two identical electrodes are tested each time in 500 mL of synthetic solution contaminated by MB dye and total coliform.

Voltage, current density and distance between electrodes are the other parameters, the effects of which on the removal of dye and total coliform are investigated in this study. In this study, neon transformers are used to transform the urban electricity into high voltage current. Thus, the voltage and

Table 2
Ringer solution properties [43]

Parameter	Value
NaCl	8,690 mg/L
KCl	300 mg/L
CaCl ₂	480 mg/L
pH	6.4
EC	6,500 μ S/cm

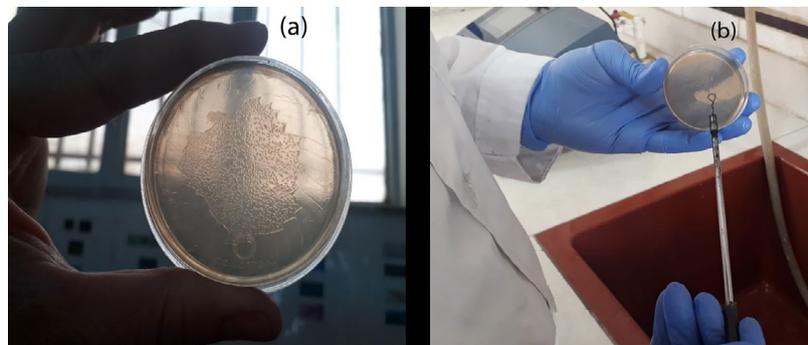


Fig. 2. Cultured colonies of total coliform (a), and amounts of these coliform colonies are detached (b) and solved in Ringer solution little by little to reach the light absorbance of 0.15 in 620 nm. By doing this, we will have the number of 10^9 total coliform in 1,000 mL. coliform-free Ringer solution used as blank solution [42].

current density are determined by the current and voltage ratings of available neon transformers. In addition, preliminary tests showed that if distance between electrodes is under 2 cm or over 10 cm, the efficiency of the process will be significantly reduced. Specifications of tests that were conducted in the present study come in Table 3.

2.3. Transformer

The first step of this study was to design and build a transformer that transforms AC power into DC power with high voltage. To this end, first, 220 V AC to 15,000 V AC power is transferred. Then, the high voltage DC is produced [46,47]. The neon light transformer seems to be proper for the objectives of the present study [23]. Three different sizes of the neon transformer with output voltages of 9, 12, 15 kV are used here to transform the public electricity into high voltage AC. To transform AC into DC power, two circuits consisting of a proper number of N007 diodes are set up in series to produce positive pole, and the other circuit is set up in reverse direction to produce the negative pole [23,46,47]. One of the innovations of the present study is that in all of previous studies the transformer circuit used the earth pole as the negative pole (Fig. 3) while here, a real negative pole is made to provide more stable power for the system. In the previous studies, the high voltage discharge was made by

Table 3 Specifications of tests of the present study

Initial concentration of MB	100 mg/L
Initial number of total coliform	10 ⁹ NTC/L
Type of water	Distilled water
Electrical conductivity of water	~0 μS/cm
Time	0–120 min
Temperature	25°C
pH	~7
Material of electrodes	Fe, Al, Cu
Surface of electrodes	20 cm ²
Distance between electrodes	2, 5, 10 cm
Reactor	1,000 mL Beaker
Voltage	9, 12, 15 kV
Nominal current density	1.5, 3, 4.5 mA/cm ²
Added NaCl	1,000 mg/L
Overall electrical conductivity of solution	2,100 μS/cm

NTC: Number of total coliforms.

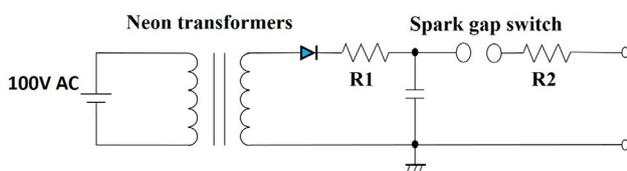


Fig. 3. Schematic of the circuit used by Shimizu et al. [23], the earth pole is used as a negative pole. Most of the other similar studies use same circuit with different type of transformer.

using capacitor. However, in this study, non-pulsed current is produced without using any capacitor.

Fig. 4 shows the schematic of the designed circuit. To avoid the heating problem, two similar transformers and four diod-based circuits are working in turn that is managed by a timer switch. Fig. 5 shows the real system built for this study.

In order to change the amount of current, one to three identical transformers with output current of 30 mA are set up in parallel to produce 30, 60 and 90 mA. To change the voltage, three different sizes of neon transformers with output voltages of 9, 12 and 15 kV are deployed in the circuit.

Due to the rated current of the neon transformers, the current densities and voltages are as presented in Table 3.

For both electrocoagulation and disinfection processes, all tests are conducted for each electrodes material, considering different experiment times, 30, 60, 90, 120 min. A magnetic stirrer with 180 rpm is used to increase the probability of impacts of colloids. For settlement of flocks, after each experiment samples remain (move less) in beaker for 10 min for MPN test and 1 h for MB concentration. Then, 100 mL of sample is taken from the above of beaker for measurement. The duration of settlement for MPN test is shorter to avoid the settlement of bacteria by their own weight.

2.4. RSM analysis

RSM design and analysis are performed by Design-Expert 10 software. In this study, R² value is selected to demonstrate the accuracy of these models.

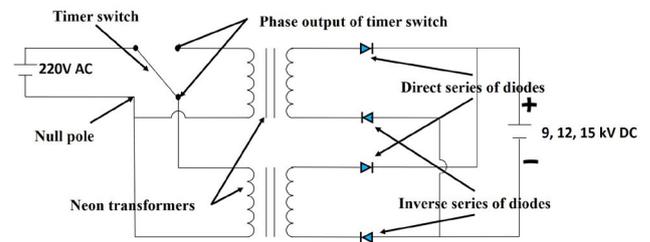


Fig. 4. Schematic of the circuit designed for the present study.

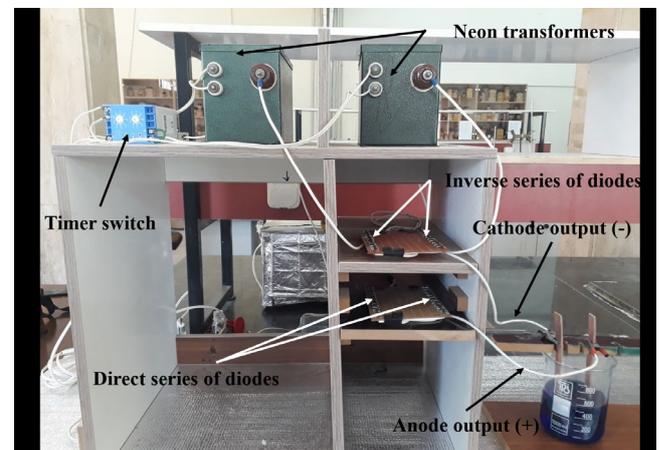


Fig. 5. Real system built for the present study.

$$R^2 = 1 - \frac{SS_{res}}{SS_{tot}} \quad (2)$$

where

$$SS_{tot} = \sum_i (y_i - \bar{y})^2 \quad (3)$$

$$SS_{res} = \sum_i (y_i - f_i)^2 = \sum_i error_i^2 \quad (4)$$

where, y_i is the real data of i th result, f_i is the i th estimation result, and \bar{y} is the average of real data.

RSM is also used to optimize the electrical energy consumption (EEC, kWh/L) by minimizing voltage, current density, and the time of process. EEC is calculated by Eq. (1) [7,13]:

$$EEC \left(\frac{kWh}{m^3} \right) = \frac{V(V) \times I(A) \times t(min)}{60 \times Vol(L)} \quad (5)$$

Or

$$EEC(kWh/kg \text{ of removed pollution}) = \frac{V(V) \times I(A) \times t(min)}{60 \times Vol(L) \times C(g/L)} \quad (6)$$

3. Results

3.1. MB removal

Figs. 6–8 show the results of MB removal with the different electrode material, voltage, electrodes distance, and current density.

Fig. 6 shows that the voltage has a direct impact on dye removal and higher voltage increases the speed and final removal of dye. These tests also reveal that Fe electrodes are the best for MB removal.

Fig. 7 demonstrates that distance between electrodes has an inverse impact on dye removal and increasing the distance will increase the duration time of removal and the final concentration of MB.

Fig. 8 shows that increasing the current density will increase the speed of removal reaction, and the final concentration will be decreased.

For each electrode type, RSM is employed in order to study and model the interaction between the operational parameters of: V (9–15 kV), Cd (1.5–4.5 mA/cm²), d (2–10 cm), t (30–120 min) as the numerical parameters and the MB removal (mg/L) as the target of analyses. The specifications of these tests are the same as those presented in Table 3.

Table 4 shows the polynomial models for calculating MB removal with respect to the operational parameters.

RSM is also employed for optimization of the MB removal process. Given Eq. (6), EEC depends on V , I , t , and value of removed MB. Therefore, for minimizing the EEC, we try to minimize V , Cd , t , while maximizing MB removal, and d value is in the range. Results of the optimization of MB removal process are concluded in Table 5. Desirability

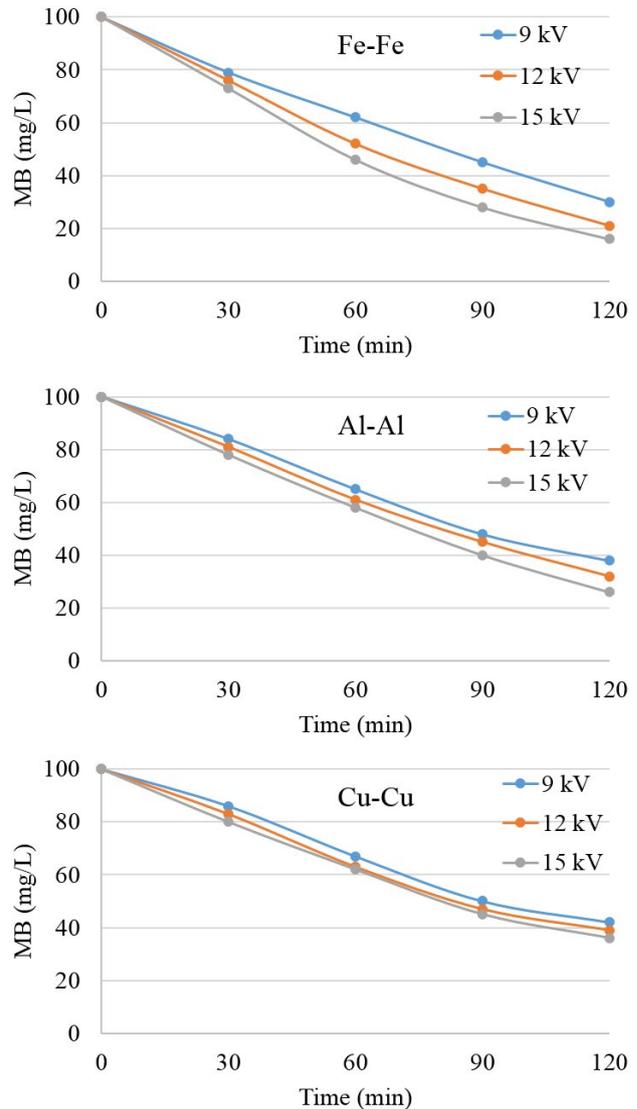


Fig. 6. Effect of voltage on the MB removal. $Cd = 1.5 \text{ mA/cm}^2$, $d = 2 \text{ cm}$.

of the results is evaluated with a number between 0 and 1. 0 and 1 express the most undesirable and desirable conditions, respectively. Theoretical anode consumption is also indicated in Table 5 as m_{anode} in milligrams.

3.2. Total coliform removal

Figs. 9–11 show the results of total coliform removal with the different electrode material, voltage, electrodes distance, and current density.

Results reveal that the Cu electrodes have a strong inhibition and removal effect on total coliform and decrease the total coliform to absolute zero. Voltage has a direct impact on total coliform removal.

Fig. 10 shows that increasing the distance between electrodes will increase the disinfection process time and increase the final NTC of the solution. However, in the case of Cu

electrodes, even with 10 cm distance, the final NTC would be equal to zero finally.

As can be seen in Fig. 11, the current density has a direct impact on total coliform removal and increasing it will

increase the speed of reaction and decrease the final NTC of the solution.

RSM method is employed so as to investigate the effect of operational parameters on NTC removal, and optimization of this process. The details of RSM design and analysis are the same as abovementioned in MB removal section. However,

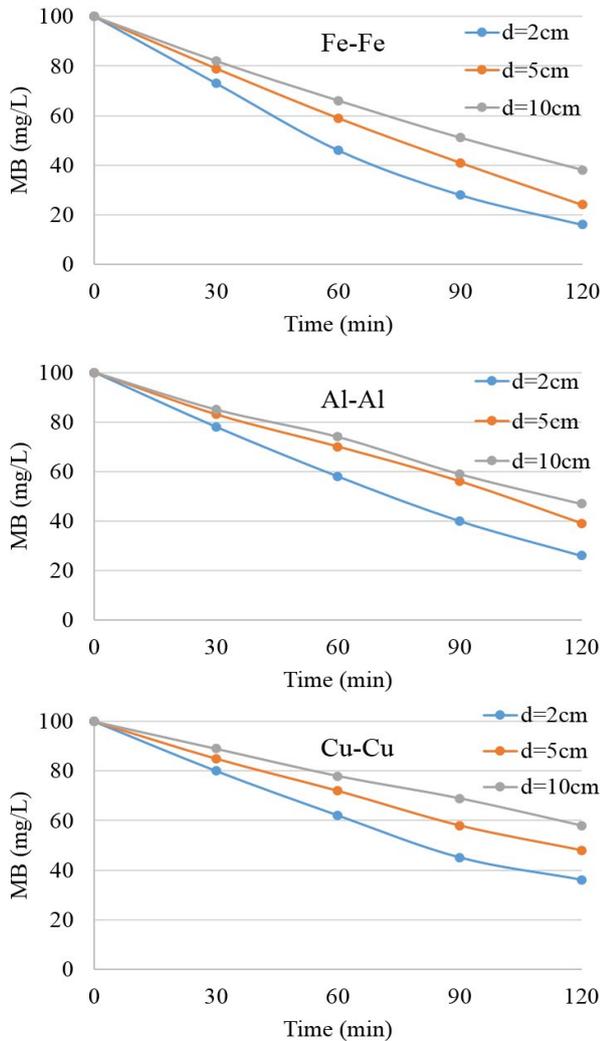


Fig. 7. Impact of distance between electrodes on the MB removal. V = 15 kV, Cd = 1.5 mA/cm².

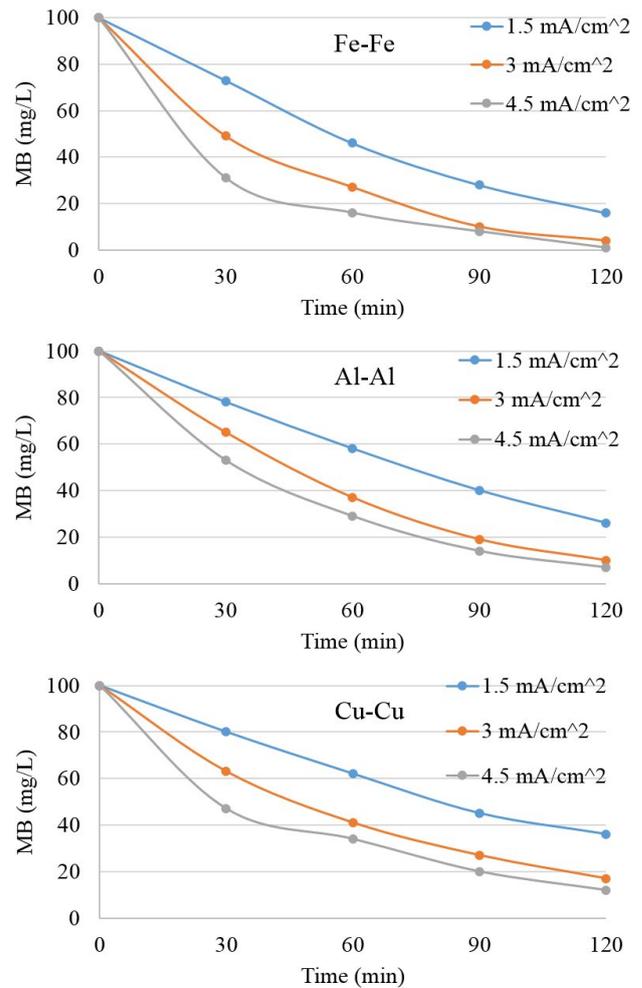


Fig. 8. Effect of current density on the MB removal. V = 15 kV, d = 2 cm.

Table 4
MB removal models

MB removal (mg/L) with Fe electrodes:	R^2
$(-0.189 + 0.109 \times V + 1.308 \times Cd - 0.166 \times d + 0.093 \times t + 0.0196 \times Cd \times d - 0.009 \times Cd \times t - 0.0002 \times t^2)^2$	0.982
MB removal (mg/L) with Al electrodes:	
$(-0.159 + 0.131 \times V + 0.809 \times Cd - 0.186 \times d + 0.087 \times t + 0.0269 \times Cd \times d - 0.0055 \times Cd \times t - 0.00025 \times t^2)^2$	0.973
MB removal (mg/L) with Cu electrodes:	
$(-0.700 + 0.153 \times V + 0.988 \times Cd - 0.173 \times d + 0.074 \times t - 0.0057 \times Cd \times t - 0.00017 \times t^2)^2$	0.945

Table 5
Optimum conditions for MB removal

Electrode type	V (kV)	Cd ^a (mA/cm ²)	d (cm)	t (min)	MB removal (mg/L)	Desirability	EEC (kWh/kg MB)	m _{anode} (mg)
Fe	9	2.9	2	82	66	0.52	21,618	83
Al	10.2	2.7	2	95	59	0.43	29,563	29
Cu	11.2	3.2	2	95	59	0.40	38,472	120

^aCd could be converted into current intensity (I) by multiplying in the surface area of electrodes (20 cm²).

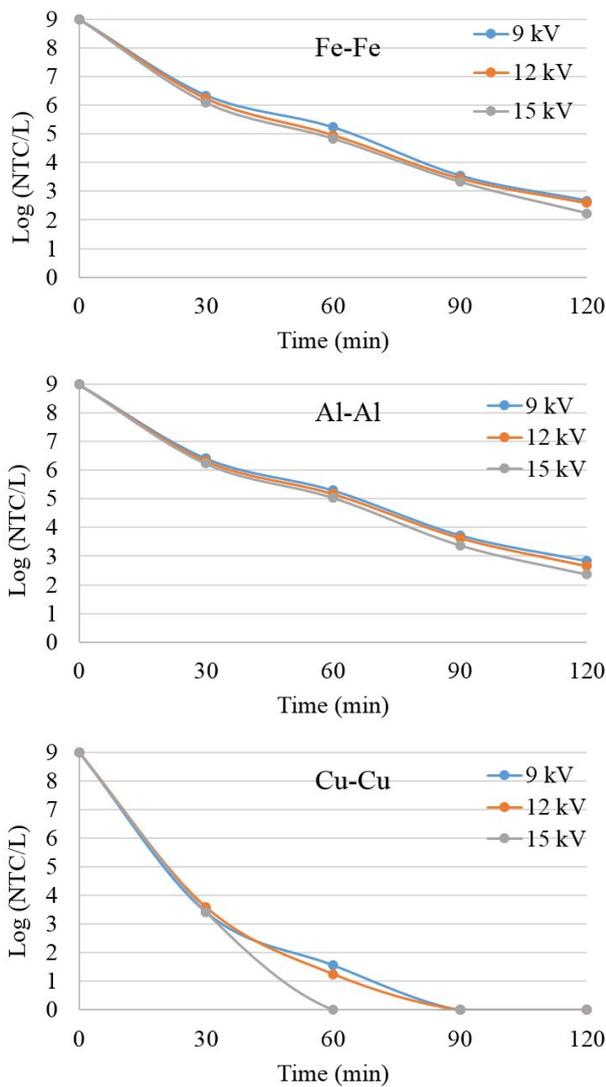


Fig. 9. The effect of voltage on the total coliform removal. *d* = 2 cm, Cd = 1.5 mA/cm². The final results of Cu electrodes were complete removal of total coliform (NTC = 0). Since the charts are logarithmic, the final NTC for Cu electrode is equal to 1 (NTC = 1).

from the data in Fig. 11, for Cu electrodes, after 60 min, the NTC reaches 0. So, with regard to Cu electrodes, the range of time is set up to 30–60 min in RSM designs. Models for calculating Log (NTC removal/L) are presented in Table 6.

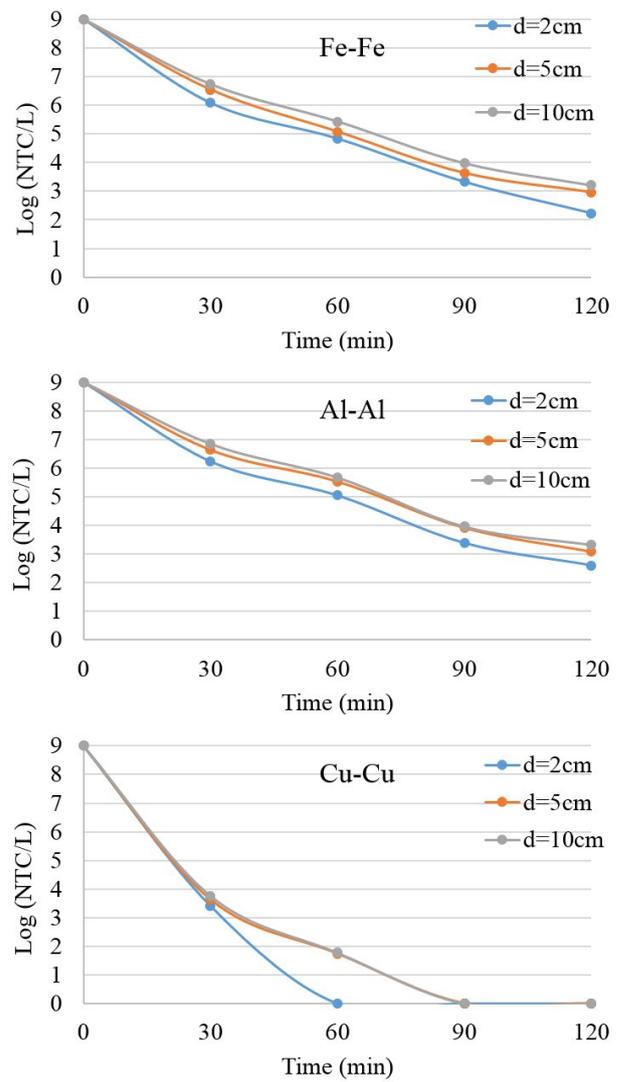


Fig. 10. Effect of distance between electrodes on the total coliform removal. *V* = 15 kV, Cd = 1.5 mA/cm². The final results of Cu electrodes were complete removal of total coliform (NTC = 0), but since the charts are logarithmic, the final NTC for Cu electrode is equal to 1 (NTC = 1).

Table 7 shows the results of optimization of total coliform removal:

Final temperature is almost equal to the room temperature and does not change during the experiments. Initial and

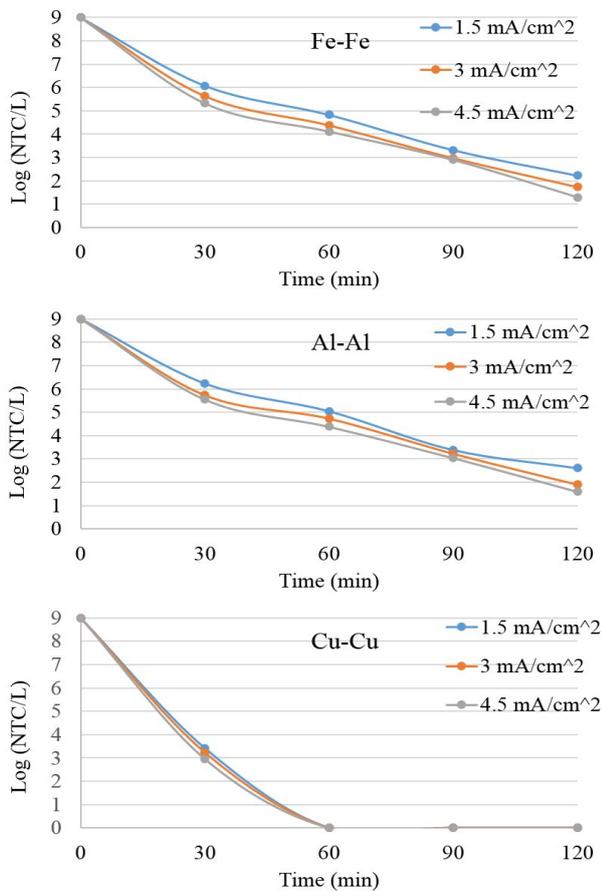


Fig. 11. Effect of current density on the total coliform removal. $V = 15$ kV, $d = 2$ cm. The final results of Cu electrodes were complete removal of total coliform (NTC = 0). But, since the charts are logarithmic, the final NTC for Cu electrode is equal to 1 (NTC = 1).

final pH also are almost the same, and the maximum pH variation is around ± 0.2 .

4. Discussion

Results show that the presented system has low cost and includes simple instruments. Non-pulsed high voltage

current is capable of removing both MB and total coliform effectively. The effect of electrode material and V , Cd , and t , on removing these two contaminants is also investigated, and accurate models are presented for calculation of the amount of removal.

Voltage and current density have direct impact on total coliform removal because more voltage can destroy the wall of coliform cells, and more current density means more electron flow that can lead to more coliform cells.

EEC values of the dye removal with electrocoagulation process are much higher than previous studies that employed EC for dye removal. For example, Kuokkanen et al. [7] reported EECs lower than 190 kWh/kg COD, in wastewater treatment applications with low voltage current.

Low efficiency of the presented system in dye removal could be explained with low current intensity of system. The efficiency of electrocoagulation process directly depends on the amount of the coagulant that is produced by dissolution of anode in the solution. In addition, as Eq. (1) implies that the amount of metal dissolution depends on the current intensity, and it is independent of voltage value. In this study, current intensities are very low (<90 mA) and the voltage is very high (>9 kV), resulting in high electrical power of system with low dye removal efficiency.

However, in comparison with studies that made attempts to remove coliform with pulsed high voltage, the EEC value of this study is very low. For example, Shimizu et al. [23] employed 8 kV, ~ 7 A current for removal of 7 log of coliform in 50 mL that equals to 5.7 log in 1 L in 20 min. The optimum EEC for their study is equal to 3,275 kWh/Log(NTC).L, which is around 50 times more than our optimum results.

Present study indicates that Fe electrodes are more effective than Al, and Al is more effective than Cu electrodes in dye removal. These results are in accordance with the study by Barrera Díaz and González-Rivas [16]. They compared the effect of these electrode materials on COD removal by low voltage (lower than 15 V [48]) electrocoagulation [16].

This study implies that the amount of voltage has a direct effect on dye removal. More voltage will lead to more dye removal, as also approved by Zidane et al. [6] that studied the effective parameters such as voltage on dye removal efficiency.

Results also reveal that more current density results in more dye removal. This statement is in agreement with the other similar studies, such as Abu Ghalwa et al. [13].

Table 6
Total coliform removal models

Log (NTC removal/L) with Fe electrodes:	R^2
$(0.878 + 0.013 \times V + 0.233 \times Cd - 0.023 \times d + 0.014 \times t - 0.0271 \times Cd^2 - 0.00003 \times t^2)^2$	0.990
Log (NTC removal/L) with Al electrodes:	
$(0.953 + 0.013 \times V + 0.065 \times Cd - 0.023 \times d + 0.018 \times t - 0.00006 \times t^2)^2$	0.976
Log (NTC removal/L) with Cu electrodes:	
$(2.926 - 0.024 \times V + 0.046 \times Cd - 0.014 \times d - 0.035 \times t + 0.00078 \times V \times t + 0.00046 \times t^2)^2$	0.923

Table 7
Optimum conditions for total coliform removal

Electrode type	V (kV)	Cd (mA/cm ²)	d (cm)	t (min)	Log (NTC removal/L)	Desirability	EEC (kWh/Log(NTC))	m_{anode} (mg)
Fe	9	2.1	2	87	5.324	0.57	206	63
Al	9	1.5	2	66	4.229	0.71	140	11
Cu	9	1.5	2	67	9	0.87	67	40

Total coliform removal with Fe electrodes is slightly more than that of Al electrodes. However, Cu electrodes are significantly more effective than Fe and Al in total coliform removal. These results are in line with the results of Varkey [44]. The paper worked on the effect of explosion of microorganism to several metal plates without any electrical current. This study proves that Cu is much more effective than Al in removing total coliform [44].

Temperature and pH changes during experiments are negligible. Consistency of temperature can be explained with low current intensity of system that does not generate much heat. Hence, the ambient temperature overcomes the small temperature changes.

The low pH changes also can be explained by low metal dissolution during the processes. In addition, majority of dissolved metal is separated from solution through the coagulation process. Therefore, initial and final pH are almost the same.

5. Conclusions

An effective system was designed and built in the present study. The system has several innovations in comparison with previous studies. Both positive and negative poles are made here, while in other studies the earth is used as the negative pole. The system is designed with cheap and simple instruments, which is effective on both dye removal and disinfection of total coliform in one reactor. This is a significant advantage that could reduce the required time, area, equipment and cost of the treatment process. Hence, the system is simple and has low cost. The interaction of the operational parameters was modeled by using RSM with very high accuracy ($R^2 > 0.92$). Presented models can be used to predict and increase the efficiency of the process in practical applications. The process was also optimized by RSM to minimize the duration of the process.

The output current of the system is non-pulsed high voltage current, while the other studies used pulsed discharge produced by capacitors, which generate spark to remove biological waste. However, the presented method has a negligible effect on the ambient temperature, and it does not produce heat pollution. The results showed that pH variations during the process is negligible.

The capability of removing both dye and total coliform in one reactor without using any additional chemical and/or physical procedures can be very useful in water/wastewater treatment processes. Since additional chemical procedures are not used, the amount of generated sludge is minimum that reduces the cost of sludge disposal. Therefore, this method can be employed for slaughterhouse, tannery, and hospital wastewater treatment.

The tests which were conducted in the present study revealed that voltage and current intensities have direct impacts on dye and total coliform removal, and increasing these parameters will decrease the duration time of removal. However, the distance between electrodes has an inverse impact on dye and coliform removal, which increased the duration time of removal reaction. Fe electrodes are the best material for MB removal. Cu is the best material for total coliform removal and can decrease the number of total coliform faster than the other materials. Cu electrodes eliminate the whole of total coliform. Al and Fe materials can reduce the number of total coliform by 99.99%. However, they cannot remove all the total coliform.

The system that is designed and made in this study has a relative low EEC value for total coliform removal with non-pulsed high voltage in comparison with others that used pulsed high voltage for disinfection purposes. The current study also found that the EEC value for dye removal is relatively high in comparison with low voltage electrocoagulation process. Hence, the proposed system is suitable for both dye and total coliform removal in one reactor or just total coliform removal. However, it is not proper to use the presented system for just dye removal by electrocoagulation.

As future works, we suggest investigating the effects of other effective parameters such as properties of solution, initial concentration, type and concentration of electrolyte, and higher current densities on the results of dye removal and disinfection by using the presented practical system.

References

- [1] A.R. Khataee, V. Vatanpour, A.R. Amani Ghadim, Decolorization of C.I. acid blue 9 solution by UV/Nano-TiO₂, Fenton, Fenton-like, electro-Fenton and electrocoagulation processes: a comparative study, *J. Hazard. Mater.*, 161 (2009) 1225–1233.
- [2] M.S. Mahmoud, J.Y. Farah, T.E. Farrag, Enhanced removal of Methylene Blue by electrocoagulation using iron electrodes, *Egypt. J. Pet.*, 22 (2013) 211–216.
- [3] A. Anouzla, Y. Abrouki, S. Souabi, M. Safi, H. Loukili, H. Rhabal, R. Slimani, Optimization of coagulation process with SIWW is coagulant for colour and COD removal of acid dye effluent using central composite design experiment, *Int. J. Environ. Monit. Anal.*, 2 (2014) 1–5.
- [4] R.V. Kandisa, K.V.N. Saibaba, S.K. Beebi, R. Gopinath, Dye Removal by Adsorption: A Review, *J. Bioremediat. Biodegrad.*, 7 (2016) 1–16.
- [5] H.N.J. Hoong, N. Ismail, Removal of dye in wastewater by adsorption-coagulation combined system with *Hibiscus Sabdariffa* as the coagulant, *MATEC Web Conf.*, 152 (2018) 1–7.
- [6] F. Zidane, P. Drogui, B. Lekhlif, J. Bensaid, J.F. Blais, S. Belcadi, K. El Kacemi, Decolorization of dye-containing effluent using mineral coagulants produced by electrocoagulation, *J. Hazard. Mater.*, 155 (2008) 153–163.

- [7] V. Kuokkanen, T. Kuokkanen, J. Rämö, U. Lassi, Recent applications of electrocoagulation in treatment of water and wastewater—a review, *GSC*, 3 (2013) 89–12.
- [8] A.S. Naje, S. Chelliapan, Z. Zakaria, M.A. Ajeel, P.A. Alaba, A review of electrocoagulation technology for the treatment of textile wastewater, *Rev. Chem. Eng.*, (2016) 1–30.
- [9] D. Rabiataladawiyah, C.A. Luqman, S. Shafreeza, Potential of copper electrodes in electrocoagulation process for glyphosate herbicide removal, *MATEC Web Conf.*, 103 (2017) 1–12.
- [10] R. Katal, H. Pahlavanzadeh, Influence of different combinations of aluminum and iron electrode on electrocoagulation efficiency: application to the treatment of paper mill wastewater, *Desalination*, 265 (2011) 199–205.
- [11] C. Guohua, Electrochemical technologies in wastewater treatment, *Sep. Purif. Technol.*, 38 (2004) 11–41.
- [12] R.G.B. Akanksha, K.S. Lokesh, Comparative study of electrode material (iron, aluminum and stainless steel) for treatment of textile industry wastewater, *Int. J. Environ. Sci.*, 4 (2014) 519–531.
- [13] N.M. Abu Ghalwa, A.M. Saqer, N.B. Farhat, Removal of reactive red 24 dye by clean electrocoagulation process using iron and aluminum electrodes, *J. Chem. Eng. Process Technol.*, 7 (2016) 1–7.
- [14] E. Butler, Y.T. Hung, R.Y.L. Yah, M.S. Al Ahmad, Electrocoagulation in wastewater treatment, *Water*, 3 (2011) 495–525.
- [15] A.I. Adeogun, R.B. Balakrishnan, Electrocoagulation removal of anthraquinone dye Alizarin Red 5 from aqueous solution using aluminum electrodes: kinetics, isothermal and thermodynamics studies, *Int. J. Electrochem. Sci. Eng.*, 6 (2016) 199–213.
- [16] C.E. Barrera Diaz, N. González-Rivas, The use of Al, Cu, and Fe in an integrated electrocoagulation-ozonation process, *J. Chem.*, 2015 (2015) 1–6.
- [17] J.A. Impa, D.P. Nagarajappa, K. Gouda, N.T. Manjunath, Domestic wastewater treatment by electrocoagulation using copper and aluminum electrodes, *Int. J. Innov. Res. Sci. Eng. Technol.*, 4 (2015) 3844–3850.
- [18] E. Bazrafshan, K.A. Ownagh, A.H. Mahvi, Application of electrocoagulation process using iron and aluminum electrodes for fluoride removal from aqueous environment, *Electron. J. Chem.*, 9 (2012) 2297–2308.
- [19] Z. Atashzaban, A. Seidmohammadi, D. Nematollahi, G. Azarian, S.O. Heidary, A.R. Rahmani, The efficiency of electrocoagulation and electroflotation processes for removal of polyvinyl acetate from synthetic effluent, *Avicenna. J. Environ. Health Eng.*, 3 (2016) 1–7.
- [20] M.A. Ahangarnokolaie, H. Ganjidoust, B. Ayati, Optimization of parameters of electrocoagulation/flotation process for removal of Acid Red 14 with mesh stainless steel electrodes, *J. Water Resuse Desalin.*, 8 (2018) 278–292.
- [21] B.M.B. Ensano, L. Boreal, V. Naddeo, V. Belgiorno, M.D.G. De Luna, F.C. Ballesteros Jr, Removal of pharmaceuticals from wastewater by intermittent electrocoagulation, *Water*, 9 (2017) 1–15.
- [22] D.C. Johnson, D.S. Dandy, V.A. Shamamian, Development of a tubular high-density plasma reactor for water treatment, *Water Res.*, 40 (2006) 311–322.
- [23] K. Shimizu, S. Muramatsu, T. Sonoda, M. Blajan, Water treatment by low voltage discharge in Water, *Int. J. Plasma Environ. Sci. Technol.*, 4 (2010) 58–64.
- [24] A. Hazmi, R. Desmiarti, E.P. Waldo, A. Darwison, Removal of microorganisms in drinking water using a pulsed high voltage, *J. Eng. Technol. Sci.*, 45 (2013) 1–8.
- [25] J. Wang, H. Tan, Y.L. Zhang, Y.Z. Pan, The treatment of high concentration dyeing wastewater with pulsed current electrocoagulation, *Mod. Appl. Sci.*, 10 (2016) 87–97.
- [26] High-Voltage Water Purification for Water Recycling or Point-of-Use Applications, NASA, 2015 NP-2015-04-1498-HQ.
- [27] M. Jourshabani, A. Badiei, N. Lashgari, G.M. Ziarani, Application of response surface methodology as an efficient approach for optimization of operational variables in Benzene hydroxylation to Phenol by V/SBA-16 nanoporous catalyst, *J. Nanostruct.*, 6 (2016) 105–113.
- [28] K.A.M. Said, M.A.M. Amin, Overview on the response surface methodology (RSM) in extraction processes, *J. Appl. Sci. Proc. Eng.*, 2 (2015) 8–17.
- [29] S.A. Younis, W.I. El-Azab, N.S. El-Gendy, S.Q. Aziz, Y.M. Moustafa, H.A. Aziz, S.S. Abu Amr, Application of response surface methodology to enhance Phenol removal from refinery wastewater by microwave process, *Int. J. Microwave Sci. Technol.*, (2014) 1–12.
- [30] Z. Derakhshan, M.A. Baghapour, M. Ranjbar, M. Faramarzian, Adsorption of methylene blue dye from aqueous solutions by modified pumice stone: kinetics and equilibrium studies, *Health Scope*, 2 (2013) 136–144.
- [31] D.B. Jirekar, A.A. Pathan, M. Farooqui, Adsorption studies of methylene blue dye from aqueous solution onto *Phaseolus aureus* biomaterials, *Orient. J. Chem.*, 30 (2014) 1263–1269.
- [32] J.M. Salman, M.A. Baiee, A.R. Omran, Experimental study to removal of methylene blue dye from aqueous solution by adsorption on eco-friendly materials, *Int. J. Chem. Technol. Res.*, 9 (2016) 560–566.
- [33] X. Duan, P. Wu, K. Pi, H. Zhang, D. Liu, A.R. Gerson, Application of modified electrocoagulation for efficient color removal from synthetic methylene blue wastewater, *Int. J. Electrochem. Sci.*, 13 (2018) 5575–5588.
- [34] DR5000 Spectrophotometer Procedures Manual, Hach Company, 2005, November 05 Edition 2, Catalog Number DOC082.98.00670.
- [35] Y. Rohmah, A. Rinanti, D.I. Hendrawan, The determination of ground water quality based on the presence of *Escherichia coli* on populated area (a case study: Pasar Minggu, South Jakarta), *IOP Conf. Series: Earth Environ. Sci.*, 106 (2018) 1–6.
- [36] A. Rompre, P. Servais, J. Baudart, M.R. Roubin, P. Laurent, Detection and enumeration of coliforms in drinking water: current methods and emerging approaches, *J. Microbiol. Methods*, 49 (2002) 31–54.
- [37] K.N. Nicholson, K. Neumann, C. Dowling, S. Sharma, *E.coli* and Coliform Bacteria as indicators for Drinking Water Quality and handling of Drinking Water in the Sagarmatha National Park, Nepal, *Environ. Manage. Sustain. Dev.*, 6 (2017) 411–418.
- [38] A.H. Divya, P.A. Solomon, Effects of some water quality parameters especially total coliform and fecal coliform in surface water of Chalakudy River, *Procedia Technol.*, 24 (2016) 631–638.
- [39] N.D. Pant, N. Poudyal, S.K. Bhattacharya, Bacteriological quality of bottled drinking water versus municipal tap water in Dharan municipality, Nepal, *J. Health, Popul. Nut.*, 35 (2016) 1–6.
- [40] S. Shafi, A.N. Kamili, M.A. Shah, S.A. Bandh, Coliform bacterial estimation: a tool for assessing water quality of Manasbal Lake of Kashmir, Himalaya, *Afr. J. Microbiol. Res.*, 7 (2013) 3996–4000.
- [41] Standard Method for the Examination of Water and Wastewater, 20th ed., 2018, pp. 47–58.
- [42] Microbiology- Cosmetics-General Instructions for Microbiological Examination [in Persian], Institute of Standards and Industrial Research of Iran, (2007) 48–50, ISIRI: 11068.
- [43] E.G. Gonzalez, J.C. Mirza-Rosca, Study of the corrosion of titanium and some of its alloys for biomedical and dental implant applications, *J. Electroanal. Chem.*, 471 (1999) 109–124.
- [44] A.J. Varkey, Antibacterial properties of some metals and alloys in combating coliforms in contaminated water, *Sci. Res. Essay*, 5 (2010) 3834–3839.
- [45] A. Godymchuk, G. Frolov, A. Gusev, O. Zakharova, E. Yunda, D. Kuznetsov, E. Kolesnikov, Antibacterial properties of copper nanoparticle dispersions: influence of synthesis conditions and physicochemical characteristics, *Mater. Sci. Eng.*, 98 (2015) 1–10.
- [46] R. McWeeny, More physics: electric charges and fields – electromagnetism, *Universit' a di Pisa*, (2011) 1–142.
- [47] H.G. Kwatny, K. Miu-Miller, *Power System Dynamics and Control*, Springer Science and Business Media, 2016, pp. 32–56.
- [48] C. Barrera-Díaz, P. Cañizares, F.J. Fernández, R. Natividad, M.A. Rodrigo, Electrochemical advanced oxidation processes: an overview of the current applications to actual industrial effluents, *J. Mex. Chem. Soc.*, 58 (2014) 256–275.