



Influence of operating conditions on the electrolytic treatment for the removal of color, TSS, hardness and alkalinity using Al-Al electrode combination

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Abstract: The present investigation observed the effect of current density (CD), operating time (OT), inter electrode distance (IED), electrode area (EA), initial pH and settling time (ST) using Al-Al electrode combination on the removal of color, total suspended solids (TSS), hardness (HR) and alkalinity (ALK) from biologically treated municipal wastewater (BTMW) of Sewage Treatment Plant (STP), Jagjeetpur, Haridwar, India. The maximum removal of color (99.86%), TSS (98.7%), HR (78.9%) and ALK (43.69 %) from BTMW was found with the optimum operating conditions of CD (2.65 A/m²), OT (40 mins.), IED (0.5 cm), EA (160 cm²), initial pH (7.5) and ST (60 min.). However, the maximum removal of ALK was found with the optimum operating conditions of CD (1.68 A/m²), OT (40 mins.), IED (1.0 cm), EA (80 cm²), initial pH (7.5) and ST (90 min.) The electrolytic treatment (ET) brought down the concentration of HR and ALK to the desirable limit of the BIS standards of drinking water. There was no need of pH adjustment of the BTMW during ET as the optimal removal efficiency was close to the pH of 7.5. Under optimal operating conditions, the operating cost was found to be 1.01 \$/m³ in terms of the electrode consumption (23.71x 10⁻⁵ kg Al/m³) and energy consumption (101.76 Kwh/m³). The study revealed that BTMW can be effectively treated with the Al-Al electrode combination for color, TSS, HR and ALK removal.

Keywords: Al-Al Electrode combinations, Electrolytic treatment, Operating conditions, Removal efficiency

INTRODUCTION

Water is an essential substance for living system as it allows the transport of nutrients as well as waste products in the living systems. However, sustainable water supply is becoming more challenging by the day due to ever increasing demand of growing population as well as increasing contamination of water resources. At the same time huge quantities of wastewater generated by industries of every hue and kind and also by exponential growth in the number of households are becoming a serious concern for society (Chopra and Sharma, 2013). Sewage treatment consists of an item that deserves ample documentation due to the environmental impact caused by such wastewater if directly discharged into water bodies. In addition, due to an increase in the scarcity of clean water Aiyuk *et al.* (2006), there is need for proper management of available water resources. Some of the goals of environmental protection and resource conservation concepts are the re-use of treated wastewater, residues emanating there from and other treatment by-products (Lettinga *et al.*, 2001 and Yi 2001).

Biological processes are the most common methods which are usually employed for the treatment of municipal wastewater (Iniesta *et al.*, 2002). However, due to the long detention time and large treatment

system, it results in higher treatment cost. Due to this reason, location of biological treatment utilities has become a big problem in the areas, where there is a scarcity of land and has high land value. In addition to that, biological processes are also susceptible to the changes of influent characteristics and require highly skilled labor. Therefore, there is a need for new and novel processes that could efficiently treat municipal wastewaters at relatively low operating cost. Research shows that electrochemical (EC) processes have the potential to be developed as a robust process, which can completely convert organic pollutants into gases such as N₂ and CO₂ (Feng *et al.*, 2003).

The role of electrochemistry in water and effluent treatment is relatively small since conventional electrode materials achieve only low current efficiencies due to the water electrolysis side reactions [Cominellis, 1994 and Simonsson, 1997]. Matteson *et al.* (1995) described a device, referred to as an "electronic coagulator" which electrochemically dissolves Al (from the anode) into the solution, reacting this with the OH⁻ ion (from the cathode) to form aluminum hydroxide. However, the use of sacrificial electrodes of metals which can give rise to multiple charged ions and their corresponding salts in the electrolytic systems, resulting in coagulation and flocculation of dissolved and undisclosed water

Table 1. Characteristics of BTMW (Values are Mean±SD of 5 replicates).

Parameter	Mean± S.D
pH	7.40±0.17
Conductivity (µS)	727.8±23.05
TDS (mg/l)	472.5±24.66
Colour	92.34±11.56
TSS (mg/l)	225.34±15.47
Hardness (mg/l)	336.78±21.45
Alkalinity (mg/l)	295.5±11.45

impurities. This helps in the removal of contaminants from wastewater. In the electrolytic process for the treatment of biologically treated municipal wastewater (BTMW), Al electrodes are dissolved during treatment and form their hydroxides and poly hydroxides which combine and subvert the organic and inorganic impurities in BTMW.

In an electrolytic reactor, generated Al^{3+} ions immediately hydrolyze to produce corresponding hydroxides and/or poly hydroxides in adequate pH. The Al hydroxides and poly hydroxides from the electrochemical dissolution have been reported to have stronger affinity to capture the pollutants in the wastewater, causing more coagulation than those from the conventional Al coagulants. Although the $Al(OH)_3$ produced by the anodic Al dissolution is more active to coagulate the pollutants in wastewater, the passivation of Al anodes and impermeable film formed on cathodes may interfere with the performance of electrocoagulation and electro flotation (Holt *et al.*, 2005). The destabilized particles are believed to be responsible for the aggregation and precipitation of the suspended particles and for the adsorption of the dissolved and/or colloidal pollutants which are subsequently removed by sedimentation and/or flotation (Bayramoglu *et al.*, 2004). The present study was focused to find out the treatability of BTMW in terms of color, TSS, HR and ALK removal by electro-

lytic treatment (ET) at various operating conditions using Al electrodes and also to assess the electrode and energy consumption during the ET.

MATERIALS AND METHODS

Collection of wastewater samples: The samples of BTMW were collected from the outlet of activated sludge process (ASP) of the sewage treatment plant (STP), Jagjeetpur, Haridwar (Uttarakhand), India and brought to the laboratory and then used for ET using Al-Al electrode combination. The pH of BTMW was adjusted before the electrochemical process and was maintained by adding the required amount of H_2SO_4 (1M) or NaOH (1M). The characteristics of BTMW are given in table 1.

Electrolytic experimental set up: A rectangular Reactor with external dimensions of height = 30 cm, width = 7 cm, length = 11cm and wall thickness = 10 mm constructed with glass with a capacity of 2.0 liters sample was used. Each time, the BTMW sample of 2.0 liters was collected and placed in an electrolytic Reactor. Al-Al electrode combination was connected to their respective anode and cathode leading to the D.C. Power supply (LMC electronics, India 0 -500 V and 0- 2 A) and energized for a required duration of time at different voltages and currents. All the experiments were performed at room temperature ($30\pm 2^\circ C$) and at a constant stirring speed (100 rpm) to maintain the uniform mixing of BTMW sample during the ET. Before conducting an experiment, the electrodes were washed with water, dipped into diluted HCl (5% v/v) for 5 mins., thoroughly washed with water and then finally rinsed twice with distilled water. Electrodes were dipped into BTMW sample with different surface areas (80 cm^2 , 120 cm^2 and 160 cm^2) and different electrode distances (0.5 cm, 1.0 cm, 1.5 cm, 2.0 cm and 2.5 cm). The different voltages (5 to 40 volts) were passed for different operating times (10 –

Table 2. Operating cost (OC) using Al-Al electrode combination at different electrode areas (EA) at constant operating conditions (Voltage: 40 V, OT:40 min., IED:0.5 cm,pH:7.5 and ST:30 min.).

EA (cm^2)	Al-Al electrode combination				
	CD (A/m^2)	Final pH	$C_{electrode}$ (kg/m^3)	C_{energy} (kWh/m^3)	Total OC ($$/m3)$
80	1.68	9.21	14.99×10^{-5}	64.32	0.64
120	2.09	10.89	18.68×10^{-5}	80.16	0.80
160	2.65	11.23	23.71×10^{-5}	101.76	1.01

Table 3. Rate constant (k (min⁻¹) values at variable current density and their correlation coefficients (r^2) using Al-Al electrode combination.

Voltage (V)	Color		TSS		HR		ALK	
	K (min^{-1})	r^2	K (min^{-1})	r^2	K (min^{-1})	r^2	K (min^{-1})	r^2
5	0.0045	0.9860	0.0018	0.9057	0.0012	0.9066	0.0006	0.7989
10	0.0083	0.9952	0.0053	0.9933	0.0018	0.9151	0.0020	0.9529
15	0.0120	0.9959	0.0080	0.9903	0.0029	0.9149	0.0026	0.9770
20	0.0135	0.9899	0.0094	0.9850	0.0039	0.9360	0.0034	0.9779
25	0.0157	0.9832	0.0103	0.9770	0.0058	0.9413	0.0042	0.9727
30	0.0174	0.9785	0.0130	0.9919	0.0075	0.9548	0.0053	0.9888
35	0.0203	0.9849	0.0148	0.9898	0.0099	0.9823	0.0056	0.9788
40	0.0217	0.9848	0.0217	0.9981	0.0119	0.9931	0.0062	0.9763

80 min). After passing the particular voltage for a particular time period i.e. after each batch experiment, the sample was taken for the different settling times (30, 60 and 90 mins.) as stated earlier (Chopra and Sharma, 2012).

Analytical methods: The color, TSS, HR and ALK of wastewater were analyzed before and after the ET following the standard methods for examination of water and wastewater (APHA,2005). The calculation of color, TSS, HR and ALK of removal efficiencies after ET was carried out using the formula:

$$CR \% = \frac{C_0 - C}{C_0} \times 100$$

Where, C_0 and C are concentrations of wastewater before and after electrolysis.

Kinetic study: The rate of removal of color, TSS, HR and ALK are represented by the following first-order mechanism (Ashtoukhy and Amin, 2010).

$$\ln \left(\frac{C_0}{C_t} \right) = kt$$

Where, C_0 - initial concentration (mg/L), C_t - final concentration with respect to time, and t - the time (min) and k - the rate constant (min^{-1}) for color, TSS, HR and ALK for ET using Al- Al electrode combination.

RESULTS AND DISCUSSION

The results on color, TSS, HR and ALK removal efficiency of BTMW by ET at different operating conditions like voltage/current density (CD), operating time (OT), inter electrode distance (IED), electrode area (EA), pH and settling time (ST) are shown in figs. 1- 6.

Effect of current density (CD): The effect of CD on the removal percentage of color, TSS, HR and ALK increased progressively with an increase in the CD from 0.16 to 1.68 A/m^2 corresponding to its constant voltages (5 to 40V). As indicated in Fig. 1, the maximum removal of color (82.54%), TSS (76.76%), HR (59.7%) and ALK (40.28%) was with the operating conditions of OT 30 min., IED 1.0 cm, EA 80 cm^2 , pH 7.5 and ST 30 min. This is ascribed to the fact that as the current increased, more amount of $\text{Al}(\text{OH})_3$ was produced, which could have contributed for the removal of color, TSS, HR and ALK from BTMW by precipitation and flotation.

The present finding is in support of Bukhari (2008) who stated that 0.05 A and 0.1 A of current applied for 5 mins. of electrolytic time resulted in 40–50% and 50–60% of suspended solids removal efficiency respectively and that the removal efficiency was obtained in proportion to the amount of dissolved materials against each of the working times ranging between 10 and 50 mins. Golder *et al.* (2007) investigated that more bubbles are generated at higher current and this improves the degree of mixing of Al

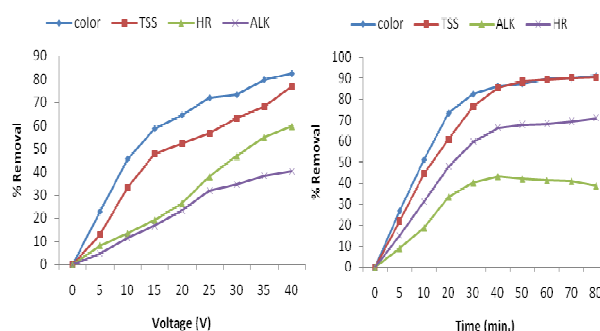


Fig. 1. % removal of color, TSS, HR and ALK using Al-Al electrode combination with different voltages at constant operating conditions (OT: 30 min., IED: 1.0cm, EA: 80 cm^2 , pH: 7.5, ST:30 min.).

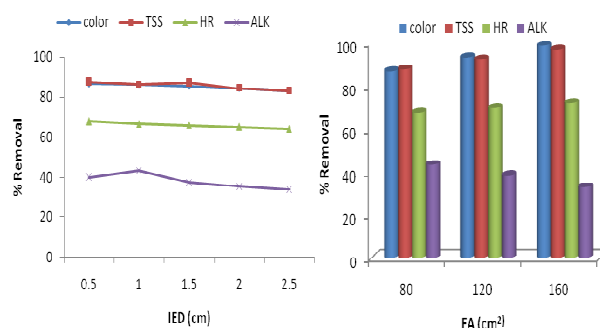


Fig. 3. % removal of color, TSS, HR and ALK using Al-Al electrode combination with different IED at constant operating conditions (Volt:40 V, OT: 40 mins., EA: 80 cm^2 , pH: 7.5, ST:30 min.).

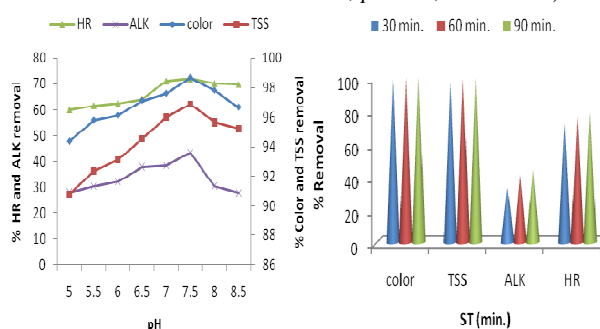


Fig. 5. % removal of color, TSS, HR and ALK using Al-Al electrode combination with different pH at constant operating conditions (Volt: 40 V, OT: 40 mins., IED: 0.5 cm, EA:160 cm^2 ,ST:30 min.).

$(\text{OH})_3$ and phenol which enhances floatation ability of the cell with a consequent increase in the phenol removal efficiency. Moreover, as shown by Khemis *et al.* (2006), higher production rates of hydrogen allowed by higher currents, favor the floatation of the flocculated matter. Zaroual *et al.* (2006) investigated that the efficiency of COD removal of textile effluent

depended on the quantity of hydroxide flocs, which was bound with time and cell current of electro coagulation. When one of the two parameters increased, the distribution of the coagulation agent density was more effective. Song *et al.* (2008) established that the rate of bubble-generation increases and the bubble size decreases with an increase in CD. Both of these trends are beneficial in terms of high pollutant removal efficiency by H₂ floatation.

Effect of operating time (OT): In the present study, OT also influenced the treatment efficiency of ET. With an increase in OT, the anodic electrode dissolution led to release of metal ions and the cathode released OH⁻ which formed their hydroxides into BTMW. Fig. 2 depicts that the removal of color, TSS, HR and ALK increased progressively with an increase in the OT from 5 to 40 min. with the operating conditions of CD 1.68 A/m², IED 1.0 cm, EA 80 cm², pH 7.5 and ST 30 min.. The maximum removal of color (86.4%), TSS (85.6%), HR (66.5%) and ALK (43.33%) was observed at optimum OT of 40 min. beyond which there was no significant removal. This may be due to the fact that the dissolved metal ions and their hydroxides in the BTMW achieved the saturation stage for the floc formation. However, ALK was found to decrease gradually with an increase in OT up to 80 mins. at the constant operating conditions. It has been observed that as the electrolysis period increases, the concentration of metal ions and their hydroxide flocs increases (Zaroual *et al.*, 2006, Daneshvar *et al.*, 2006 and Daneshvar *et al.*, 2007). Holt *et al.* (2002) investigated that more electro coagulation time (>30 min) and high voltage result in floatation of particles attached to hydrogen gas bubbles released from the cathode which are important for the turbidity removal. Kobya and Delipinar (2008) reported that an increase in the OT from 10 to 60 min. in the treatment of the Baker's yeast wastewater by electro coagulation resulted in an increase in the removal efficiency of COD, TOC and turbidity. Vijayaraghavan *et al.* (2008) also observed that an increase in the electrolysis period resulted in a decrease in residual COD and BOD concentrations irrespective of the CD during the ET of latex wastewater. It was established that the anodic electro dissolution leads to release of coagulating species in the wastewater during electrolysis. Aoudj *et al.* (2010) observed that anodic electro dissolution led to the release of coagulating species at constant CD of 2.5mA/cm² and initial pH of 6. They found that an increase in the time of electrolysis from 10 to 60 min. yields an increase in the dye removal efficiency from 52.5% to 98.28%.

Effect of inter electrode distance (IED): During the present study, IED was an effective factor in the ET of BTMW. The removal percentage of color, TSS, HR and ALK increased progressively with decrease in IED from 2.5 to 0.5 cm, whereby it exhibited the maximum

removal of color, TSS and HR at a short distance of 0.5 cm between the electrodes, each with an electrode area of 80 cm² and with the operating conditions of CD 1.68 A/m², OT 40 min, EA 80 cm², pH 7.5 and ST 30 min. (Fig. 3). After that, there was not much difference in the removal efficiency of color (86.9%), TSS (87.80%) and HR (67.8%) as the IED increased from 0.5 to 2.5 cm., whereas the ALK (40.25%) was decreased due to the increase in amount of Al(OH)₃ which enhanced the alkaline character of BTMW. Similar observations have also been reported by Li Xu. (2008) for the removal of COD from tridecane dicarboxylic acid wastewater which decreases with the decrease in distance between electrodes of the same composition. This is because the shorter distance speeds up the anion discharge on the anode and improves the oxidation. It also reduces resistance, electricity consumption and the cost of the wastewater treatment. Ghosh *et al.* (2008) have also observed that with the increase of IED, the percentage removal of dye products from waste water decreases. At a lower IED, the resistance encountered by the current flowing in the solution medium decreases, thereby, facilitating the EP resulting in enhancement of dye removal.

Effect of electrode area (EA): The greater electrode area increases the rate of floc's formation, which in turn influences the removal efficiency of color, TSS and HR. According to Daneshava *et al.*, (2005), an increase of electrode area causes a corresponding increase of coagulants. The entire effectiveness of the coagulation process depends on the appropriate amount of coagulant. The logistical relationship between electrode geometric area (AG) and Cu removal efficiency and concluded that an increase in Cu removal was related to an increase in AG, reaching an optimal value of 35 cm², with an asymptotic value of near 80 % (Escobara *et al.*, 2006). Chopra and Sharma (2013) observed that with a fourfold increase in the electrode area of Al-Fe from 40 to 160 cm², the CD increased from 0.24 to 0.58 A, which resulted in an increase in the removal percentage of TD, COD and BOD. The maximum removal efficiencies of 81.51 % (TD), 74.36 % (COD) and 70.86 % (BOD) of BTMW were achieved with the use of electrode area of 160 cm² and IED of 2.5 cm.

Similarly in the present study, it was observed that with an increase in EA from 80 to 160 cm², the CD density increased from 1.68 to 2.65 A/m² corresponding to its constant voltage of 40V that resulted in an increase in the removal percentage of color, TSS and HR of BTMW. The maximum removal of color (98.7%), TSS (96.89%) and HR (72.12%) was achieved at maximum EA of 160 cm² with the operating conditions of voltage 40 V, OT 40 min., IED 0.5 cm., initial pH 7.5 and ST 30 min., while the removal of ALK (43.33%) was with the 80 cm² (Fig. 4). However, the ALK of BTMW was decreased gradually with an increase of the electrode area of

Al-Al electrode combination due to increase in the formation of Al (OH)₃ in the BTMW.

Effect of pH: As pointed out by several authors [Lin and Chen, 1997; Chen *et al.*, 2000; Gurses *et al.*, 2002 and Kobya *et al.*, 2003], pH is an important operating factor influencing the electrocoagulation process. pH of the medium changes during the process depending on the type of electrode material and the initial pH. Gurses *et al.* (2002) observed an increase in removal efficiency of colloidal particles in the pH range of 4–7 that leads to the formation of amorphous hydroxide precipitates and other aluminum hydroxo complexes with hydroxide ions and polymeric species. On the other hand, the electro coagulation process exhibits some buffering capacity, especially in alkaline medium, which prevents large changes in pH and shows a decrease of the pollutant removal efficiency (Bayramoglu, 2004). The difference between maximum pH for COD and turbidity can be attributed to the formation of more stable flocs when pH is about 7, although it does not correspond to the optimum for dye removal (Essadki *et al.*, 2008).

In the present study, the removal efficiency of color, TSS, HR and ALK with the different initial concentrations of pH 5-8.5 of BTMW and at operating conditions of CD 2.65 A/m², OT 40 min., IED 0.5 cm, EA 160 cm² and ST 30 min. indicated that the maximum removal of color (98.7%), TSS(96.89%), HR (72.12%) and ALK(43.33%) was at the initial pH of 7.5 (Fig. 5). After that the removal efficiency of these parameters decreased gradually with the increase in pH of more than 7.5 at constant operating conditions using Al-Al electrode combination. It was found that the pH of BTMW did not have significant influence on the removal of color, TSS, HR and ALK.

Effect of settling time (ST): The effect of ST on removal efficiency of the parameters after ET has not been given due consideration so far. There appears to be no work with regard to the ST on the BTMW treatment during electrolysis. During the present study, it was interesting to note that the removal efficiency of color, TSS, HR and ALK with the constant operating conditions of CD 2.65 A/m², OT 40 min., EA 160 cm² and pH 7.5 was improved with an increase of settling time from 30 to 90 mins. of treated BTMW. The maximum removal of color (99.86%), TSS (98.7%), HR (75.89%) and ALK (40.59%) was found at the ST of 60 min. beyond which there was no significant removal of these parameters (Fig. 6). The present study indicated that the ET of BTMW brought down the concentration of HR from 336.78 mg/l to 81.19 mg/l and ALK from 295.5 mg/l to 175.55 mg/l i.e. to within desirable limit of BIS (1991) standard of drinking water.

Economic evaluation: Electrical energy and electrode consumption are the important parameters in the economic evaluation of EC process. In EC process, the operating cost includes material mainly electrodes and electrical energy cost as well as labor, maintenance,

sludge dewatering and its disposal. In the present study, energy and electrode material costs have been taken into account as major cost items in the calculation of the operating cost (US \$/m³) (Ghosh *et al.*, 2008 and Chopra and Sharma, 2012) as follows:

$$\text{Operating cost} = aC_{\text{energy}} + bC_{\text{electrode}}$$

Where, C_{energy} (KWh/m³) and C_{electrode} (kg Al/ m³) are the consumption quantities for the color, TSS, HR and ALK removal.

“a” electrical energy price 0.1 US\$/ kWh; “b” electrode material price 3.4 US\$/kg for Al electrode. Cost due to electrical energy (KWh/m³) is calculated as:

$$C_{\text{energy}} = \frac{U \times I \times t_{\text{EC}}}{v}$$

Cost for electrode (Kg Al/m³) was calculated as follows using the equation:

$$C_{\text{electrode}} = \frac{I \times t \times M_w}{z \times F \times v}$$

Where, U - cell voltage (V), I- current (A), t_{EC}- time of electrolysis (s) and v- volume (m³) of STS water, MW - molecular mass of aluminum (26.98 g/mol), z- no. of electrons transferred (z =3) and F-Faraday's constant (96487C/mol) .

The energy consumption increased from 64.32 kwh/ m³ to 101.76 kWh/m³ with an increase in EA (80-160 cm²) and CD (1.68 to 2.65 A/m²) that resulted in increase of the electrode consumption from 14.985 x 10⁻⁵ to 23.71x10⁻⁵ kg/m³ for the removal of color, TSS, HR and ALK from BTMW during ET .The cost for ET of BTMW was found to be 1.01 \$ in terms of energy and electrode consumption with the optimum operating conditions of CD (2.65 A/m²), OT(40 min.), IED(0.5 cm),pH(7.5) and ST(90 min.) of EC process (Table 2).

Kinetic evaluation: In the present study, it was revealed that the slope of the plot of C₀/C_t versus time gives the value of the rate constant at the various voltages for color, TSS, HR and ALK (Figs. 7 and 10). The study indicated that the pseudo-first-order abatement kinetic was relatively fitted. The increase in the voltages form 5 to 40 V increased the rate constant from 0.0045 to 0.0217min⁻¹ (Color), 0.0018 to 0.0217min⁻¹ (TSS) and 0.0006 to 0.0062min⁻¹ (ALK) and 0.0012 min⁻¹ to 0.0119 min⁻¹ for the ET of BTMW using Al-Al electrode combination. The increase in the rate constant may be ascribed to the decrease of color, TSS, HR and ALK of the BTMW. The removal of color, TSS, HR and ALK exhibited pseudo first order kinetic with the significant correlation coefficients (>.79) at maximum voltage of 40 V (Table 3).

Conclusion

The removal of color, TSS, HR and ALK from BTMW using Al electrodes was found to be dependent on

voltage/CD, OT, IED, EA and initial pH during the ET. The maximum removal of color, TSS and HR was with the optimal operating conditions of CD 2.65 A/cm², OT 40 min., EA 160 cm² and pH 7.5. However, the maximum removal of ALK was found with the optimum operating conditions of CD (1.68 A/m²), OT (40 mins.), IED (1.0 cm), EA (80 cm²), initial pH (7.5) and ST (60 min.) The interesting thing was that there was no need of pH adjustment of the BTMW during ET as the optimal removal efficiency was close to the pH of 7.5. The ET brought down the concentration of HR and ALK to the desirable limit of the BIS standards of drinking water. The kinetic rate constants for color, TSS, HR and ALK removal at various voltages indicated that pseudo first-order kinetic were in good agreement with the significant correlation coefficients (>.79) at maximum voltage of 40V. The operating cost for the ET of BTMW was found to be 1.01 US\$/m³ in terms of energy and electrode consumption.

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