



Reuse of Wood-Based Industrial Wastewater through Optimization of Electrocoagulation Process Using Aluminum and Iron Electrodes

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ABSTRACT

The wastewater treatment of Medium Density Fiberboard (MDF) is a harsh process because of its contents of high suspended solids, chemical oxygen demand, high molecular weight of lignin, and fatty acids. Electrocoagulation (EC) process was used for efficient removal of pollutants and reusing the water. The impact of aluminum and iron as sacrificial anodes on removal of Chemical Oxygen Demand (COD), Total Suspended Solid (TSS), turbidity, and Total Solid (TS) were investigated. A full quadratic model was deployed to optimize the EC process variables for pretreatment of MDF effluent through response surface methodology. The model results confirmed that the COD and TSS removal efficiency was enhanced upon increasing voltage and residence time; hence, other pollutants initially increased and then, decreased at higher levels. The comparison between aluminum and iron electrodes indicated that the polluted removal efficiencies of aluminum were higher than the iron electrode for MDF wastewater. The optimum values of voltage and residence time for electrocoagulation of MDF wastewater with aluminum were 33 V and 25 min, which resulted in 93 %, 89 %, 67 %, and 76 % for COD, TSS, turbidity, and TS removal, respectively. The implementation of electrocoagulation provided a possibility for reusing water and reducing water consumption in the MDF manufacturing process.

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1. INTRODUCTION

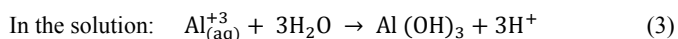
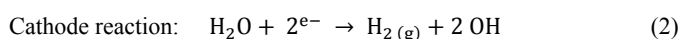
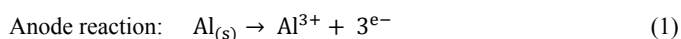
The Medium Density Fiberboard (MDF) industry is a promising industrial sector in Iranian wood-based industries. It may be susceptible to high capital energy, raw materials, and water demanding in the environment. The wood-based manufacturing may cause an increase in forming different kinds of pollutants. Therefore, it is of significance to be attentive to establishing pollution preventive facilities for formaldehyde emissions and wastewater treatment. The elevated pollutants of MDF wastewater emerging from suspended solids, Chemical Oxygen Demand (COD), Biochemical Oxygen Demand (BOD), fatty acids, and other soluble substances result in sludge thickening, dye problem, and environmental concerns [1, 2]. Therefore, noticeable concerns may arise in case of the release of the above untreated materials to the environment. The high fiber content derived from inefficient separation systems remains a problem for reuse of MDF wastewater [3, 4]. In general, the biological and chemical processes as well as their combination are the most common wastewater treatment processes. Biological treatment is sensitive to micro-organism, spaces, and high

residence time. Although chemical treatments tend to be low consumable, they remarkably expand the capacity of downstream mechanical plants. Chemical treatments have a low degree of robustness because the utilization of chemicals can be an expensive and inefficient attempt. The amendment of pretreatment allows for the reuse of treated water which will reduce water consumption in the MDF industry and facilitate an economical and efficient separation of solid from liquid. Wastewater is a valuable resource of liquid in the MDF industry and its efficient recycling brings about a number of economic and environmental advantages: minimizing the cost of waste consumption; mitigating the volume of generated wastewater, and resolving water crisis limitation.

Electrocoagulation (EC) is identified as new technology and an efficient industrial system for removing organic contaminants. EC involves in-situ generation of coagulants through dissolution of ions from metal-electrolyte (such as iron and aluminum) [5]. Thus, the mechanism of electrolytic oxidation forms coagulant during the pre-treatment of wood-based wastewater. Further, the aggregation of destabilized colloids results in flocs formation which makes purification system productive in this manufacturing. Therefore, appropriate selection of electrode materials is very important. Mechanisms that were proposed for the production of aluminum anode are as follows :

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A number of parameters with significant effect on the EC process include type of electrodes, electrolysis time, voltage, and wastewater characteristics. EC variables had different effects on the types of wastewater [6, 7]. In addition, a few studies have been explored to investigate the potential of reusing water through EC-treated effluent from industrial wastewater [8]. On the other hand, optimization of EC variables is an essential technique for high efficiency of wastewater treatment and minimizing power consumption and operation costs. Response Surface Methodology (RSM) is a useful statistical technique for optimization studies in several industrial processes [9-11]. RSM is an effective method for reducing the number of experimentals and assessing the importance of relative variable and their interaction with designing models. Optimization of wastewater process parameters by applying RSM methodology has been studied in the literature [12-14]. Thus, the aim of this study was to investigate the applicability of EC for pre-treatment and reusing water through EC process optimization to achieve maximum pollutant removal and minimum energy consumption. Removal efficiencies of COD, TSS, turbidity, and TS were measured and compared to the control group (i.e., sedimentation). Then, the possibility of reusing treated wastewater in MDF manufacturing process was analyzed.

2. EXPERIMENTAL

2.1. Electrocoagulation experiment by aluminum and iron electrodes

The samples were obtained from the plug screw feeder outlet that includes a chip converter machine, tapered compression zone, and chip/drain outlet from MDF manufacturing

(Golestan Cellulose Group; Gorgan). Iran products 1.5 million m³ of MDF per year through 8 large mills by consuming around 5 million m³ of water and discharging 3 million m³ of wastewater [15, 16]. The samples conducted in 20 l plastic containers were transferred immediately to the Environmental Health Engineering Laboratory (EHLE) of Medical Science, Gorgan, Iran and were preserved at 5 °C. According to standard methods, the characterization of wastewater samples was examined [17]. The properties of MDF wastewater are shown in Table 1.

Table 1. The typical medium density fiberboard wastewater

Component	Amount
COD (mg/l)	6400
BOD (mg/l)	2150
TSS (mg/l)	5360
Turbidity (NTU)	6170
TS (mg/l)	8000
pH	4.9

Figure 1 demonstrates the implementation of the EC process in WWTP. The primary influent from MDF wastewater was subjected to electrocoagulation with aluminum (Al-Al) and iron (Fe-Fe) in a batch reactor by optimizing two important process variables: voltage and residence time. The electrocoagulation experiment was conducted in an electrochemical reactor with four electrodes connected in parallel to a digital DC power supply (0-60 V, 5 A). A space of 4 cm between the electrodes and the 10 cm gap was maintained at the bottom of the reactor for the movement of magnetic stirrer. The dimension of the treated emulsion was 10 × 20 × 20 cm (the volume of 4 l) and the total effective electrode area was 150 cm². The experiments were performed at pH of 5.5 based on the characterization of wastewater effluent. The runs were performed at room temperature (20-25 °C).

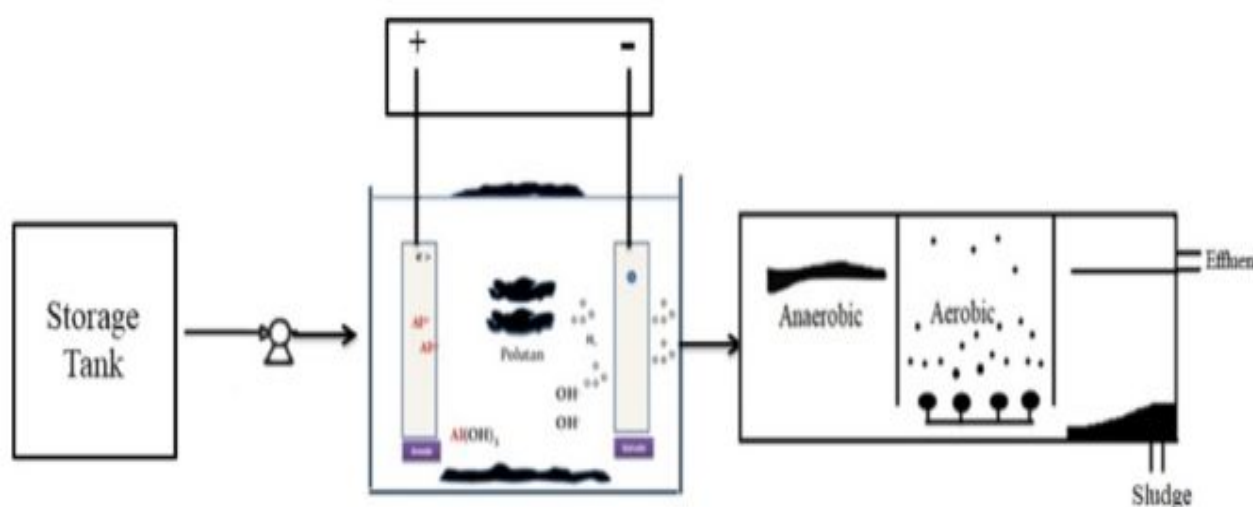


Figure 1. The overview of EC process implementation in MDF wastewater plant

2.2. Analytical measurement of COD and Turbidity

The turbidity was determined using a turbidimeter HANNA HI-98703 with a measurement range of 0 to 1000 NTU using the USEPA 180.1 procedure according to the EPA standards.

The COD was measured using a Merk Spectroquant NOVA 60 with a measurement range of 25 to 1500 mgO₂L⁻¹ using the EPA 410.4 procedure with a standard method 5220 according to the ISO 15,705 standard.

2.3. The design of experiment

The central composite design was applied in order to investigate the significance of variables in the response manner. Two following important reactions are identified as independent variables: voltage with a range of 20-40 V and residence time (10-30 min). The initial experiments were performed to choose the ranges of variables. Based on the response surface method, the CCRD was selected to fit the number of experimental runs with RSM second-order model. Furthermore, the CCRD, as an intellectual technique, was employed to select the prominent model based on the lack of fit test [18, 19]. The CCRD in the experimental design consisted of 8 factorial points, four axial points, and two blocks (aluminum and Iron electrodes). Five iterations at the center of the design were performed to predict the error sum of squares. The yield of response function helped predict the correlation between dependent and independent variables with a center point value. After measuring the response function, the second-order equation expressed the optimal point based on the full quadratic model as follows [20, 21].

$$Y = \beta_{k0} + \sum_{i=1}^{i=4} \beta_{ki}x_i + \sum_{i=1}^{i=4} \beta_{kii}x_i^2 + \sum_{i<j=2}^{i=4} \beta_{kij}x_i x_j \quad (4)$$

where Y is the predicted response (LA); β_{k0} , β_{ki} , β_{kii} and β_{kij} represent regression coefficients; x_i x_j are the coded independent factors. The quality of models was assessed by coefficient of determination (R^2), adjusted coefficient of determination (R^2 -adj), and predicted coefficient of determination (R^2 -pred). Moreover, the Root Mean Square Error of Prediction (RMSEP) and Absolute Average Deviation (AAD) were employed to minimize the relationship of responses with actual experimental values [2]. The analysis of variance (ANOVA) was accomplished to analyze the importance of response based on the accurate model.

$$RMSEP = \sqrt{\frac{\sum_{i=1}^N (y_{pre} - y_{exp})^2}{N}} \quad (5)$$

$$ADD = \left\{ \frac{\sum_{i=1}^N \left(\frac{|y_{exp} - y_{pre}|}{y_{exp}} \right)}{N} \right\} \times 100 \quad (6)$$

where y_{pre} , y_{exp} , and N are the predicted data, observed data, and number of treatments, respectively.

Optimization of multiple responses was simultaneously performed through numerical and graphical optimization execution of Design-Expert software. The optimum conditions were considered all independent variables within the studied ranges and the COD removal, the TSS removal, the turbidity removal, and TS removal were maximized. The range of 0-1 as desirability values from responses determined the reliability of optimum conditions. The most desirable optimum point was closer to 1. In order to ensure the overall process improvement, the desirability function was used as one of the most useful techniques for the optimization of multi-responses processes through independent variables series (Eq. 6).

$$D(x) = (d_1 \times d_2 \times \dots \times d_n)^{\frac{1}{n}} \quad (6)$$

where n (1, 2, ..., n) represents the number of independent variables. If any of the dependent variables is out of the desirable range, the function becomes zero. In this situation, the numerical optimization identifies a point that maximizes this function. The regression equation of the accurate model confirmed that the predicted values were in close agreement with actual values. The distribution of the predicted and actual values is illustrated in Figure 2, whose data mainly align. Thus, the underlying assumption of normality in this study was appropriate and therefore, the developed models were validated.

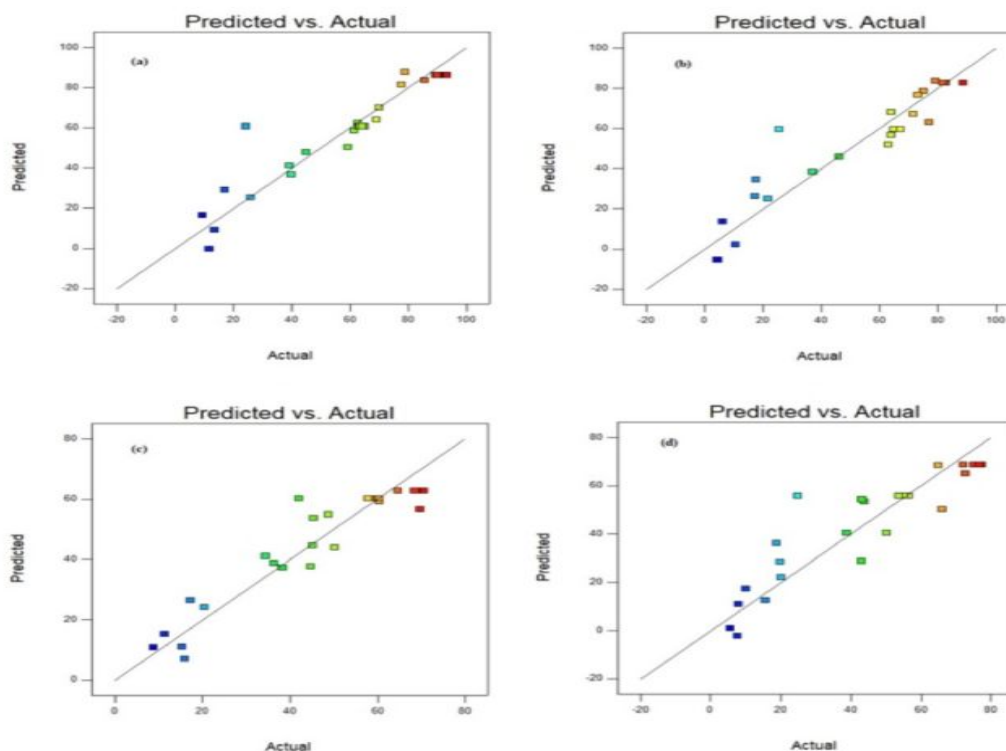


Figure 2. Predicted vs. actual values: (a) COD removal, (b) TSS removal, (c) Turbidity removal, and (d) TS removal

3. RESULTS AND DISCUSSION

3.1. The importance of electrodes on pretreatment of MDF wastewater

The variation in the pollutant removal efficiency using aluminum and iron electrodes is shown in Figure 3. The maximum COD removal efficiency rates experienced for aluminum and iron electrodes at around 30 V and 25 min were 93 % and 65 %, respectively. However, the maximum turbidity removal efficiency was around 70 % for aluminum and 60 % for iron. The results demonstrated that there were strong differences in the electrocoagulation of iron and aluminum electrodes. In [22], it was indicated that the COD and color removal by aluminum electrode was more efficient than that for the electrode at high voltage in the textile-industry wastewater. Amorphous hydroxides are the primary species in both cases. However, aluminum electrode dissolution yields significant concentrations of monomeric and even polymeric ionic species.

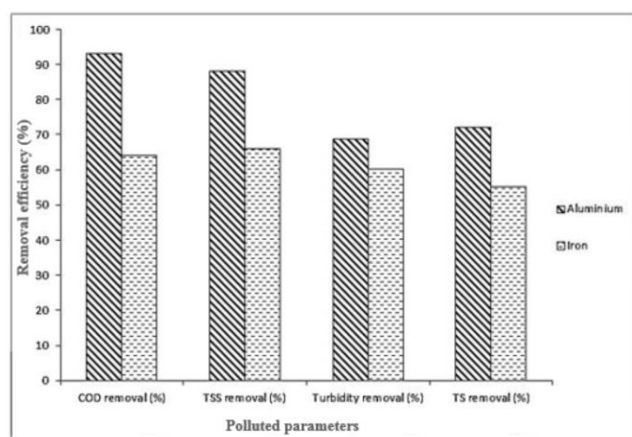


Figure 3. The effects of type of electrode on removal efficiency at voltage 30 V and time 20 min

3.2. The independent effects of voltage in electrocoagulation process

The independent influence of voltage was assessed to obtain the initial optimum condition of electrocoagulation variables (COD, TSS, Turbidity, and TS removal). Effect of voltage was investigated, ranging from 10 to 40 mA and the initial pH was kept at 5.5 in this study (Figure 4). The COD removal was found as the most significant variable when the aluminum electrode was used, while the Iron electrode exhibited remarkable responses to TSS removal. The maximum COD removal efficiency was obtained at nearly 93 % at voltage of 33 mA using aluminum electrode, whereas the COD removal of 69 % was detected by only Iron electrode in the same condition. On the other hand, the higher desirability for pretreatment of MDF wastewater through electrocoagulation process was considered with aluminum electrode when the voltage of 33 mA was relatively used in this process.

The variations in removal efficiencies for COD, TSS, turbidity, and TS removal are shown in Figure 5. The interaction between the voltage and residence time on the COD removal when pH was kept at 5.5 indicated that the COD removal gently increased upon increasing voltage. On the basis of Faraday's law, the electrolytic cell at a specific time directly corresponds to the number of coagulants at dissolved anodic metal. Therefore, the amounts of aluminum and iron hydroxide flocs increase by increasing the voltage,

which results in the enhancement of colloidal particles removal [23]. The enhancement of hydroxide ion led to increase in the pH which results in decrease in the TSS, TS, and turbidity removal, as can be seen below.

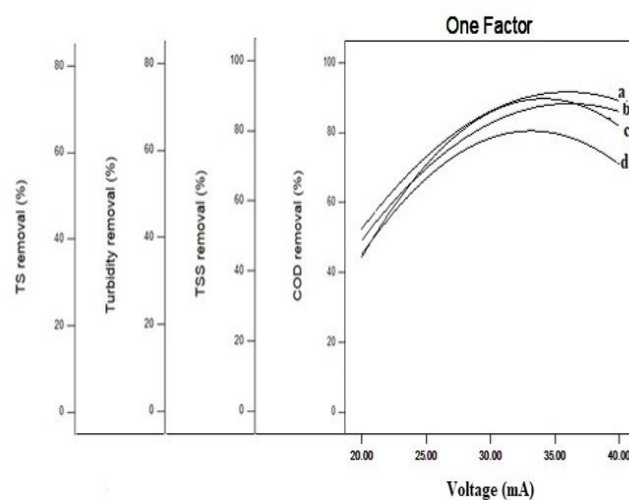


Figure 4. The independent effect of voltage on a) COD, b) TSS, c) TS, and d) turbidity removal

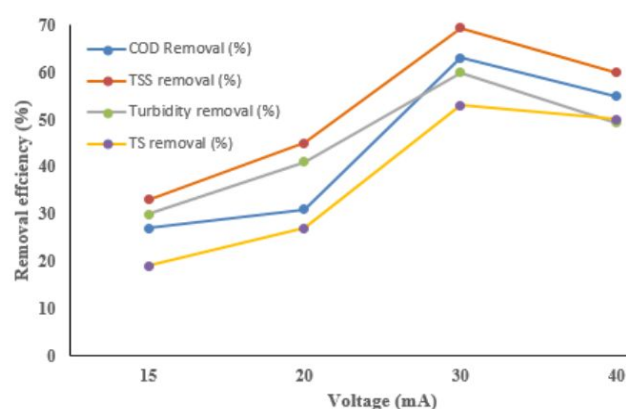


Figure 5. The effect of voltage on removal efficiency (%), Iron electrode, time 20 min

3.3. The independent correlation between residence time and the response variables

The effect of residence time on electrocoagulation is presented in Figure 6. All pollutant removal efficiencies increase with respect to the residence time. The TS removal was preliminarily enhanced by extending the time; notwithstanding, it decreased in the case of longer residence time. This evidence is presumably due to the great content of hydroxide ion from aluminum electrode at cathode, which results in the final pH rise. Mouldi-Mostafa et al. determined that the optimum reaction time for treatment was approximately 20 min [6].

In comparison of residence times, voltage has noticeably significant impact on the pollutant removal in the electrocoagulation of MDF manufacturing wastewater (Figures 4 and 7). For instance, the maximum TS removal was observed at nearly 75 % when the voltage was increased, while up to 60 % TS removal was obtained at maximum time. The optimal electrocoagulation time was found among 20 to 26 min when the maximum COD, TSS, turbidity, and TS removal was obtained at around 94 %, 89 %, 67 %, and 75 %, respectively.

respectively (Figure 7). The response variables experienced a decline at refining time of 30 min. The reduction of removal efficiency found here can be explained by the high concentration of color and colloidal particles in the samples

after the treatment. The independent relationship for residence time ascertained that the COD and TSS underwent the most significant pollution reduction rather than turbidity and TS.

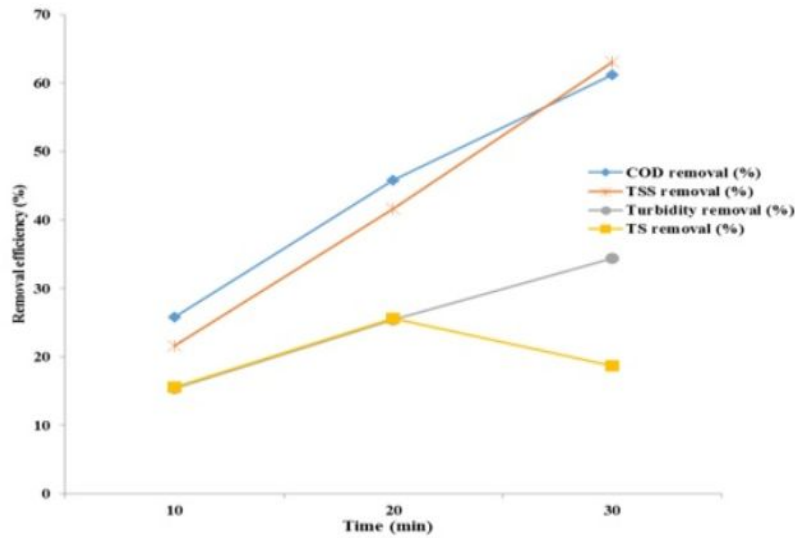


Figure 6. The effect of residence time on removal efficiency (%), aluminum electrode, voltage 20 V

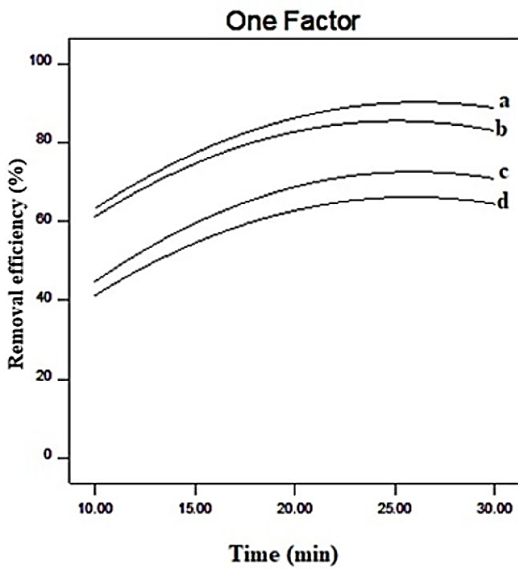


Figure 7. The independent effect of Time on a) COD removal, b) TSS removal, c) TS removal, and d) Turbidity removal

3.4. Optimization of EC variables as pre-treatment of MDF wastewater

The optimum experimental design was created and it comprised 52 sets of coagulation experiments. The full quadratic regression model was satisfied for all tests. The significance of full quadratic models to confirm the validity of models based on the ANOVA analyses is illustrated in Table 2. The importance and significance of each variable shows a large F-value and a small p-value for the responses. The values of the adjusted R² respectively measured at 0.88 and 0.83 for COD removal and turbidity removal are large enough to supported the significance of the model. The RSM plots derived from quadratic models for the variables are shown in Figure 8.

Of note, the COD and TSS removal efficiencies increased upon increasing current voltage and residence time; however,

the turbidity and TS removal efficacies initially increased with increasing voltage and residence time, followed by a decrease at higher levels. The colloidal particles present in MDF wastewater had mostly a negative superficial charge and they were destabilized by the hydroxides generated during EC. Notwithstanding this evidence, the COD and TSS removal occurred easily due to efficient destabilization of charge [24, 25]. There was a remarkable difference between COD and TS removal efficiency rates for aluminum and Iron. Aluminum electrode was found to be more efficient than Iron electrode due to its higher charge as well as high solubility during EC process. Moreover, use of iron electrodes often results in the formation of very fine brown particles which are less prone to settling than the gel floc formed with aluminium [26]. The optimum amounts of voltage and residence time for electrocoagulation of MDF wastewater with aluminum were 33 V and 25 min, respectively. The maximum COD, TSS, turbidity, and TS removal rates were acquired 93 %, 89 %, 67 %, and 76 %, respectively.

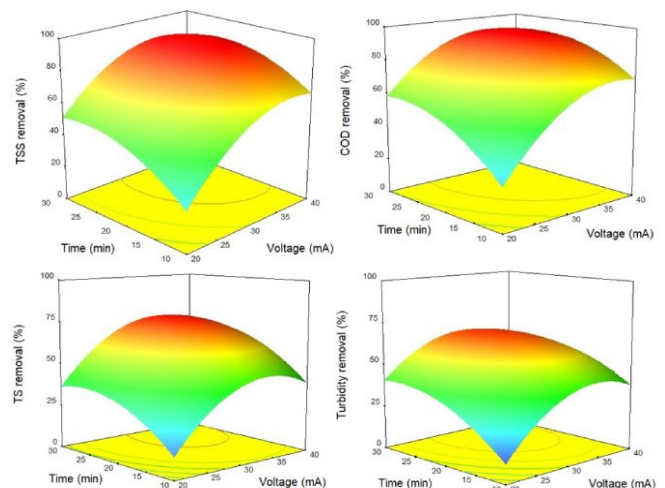


Figure 8. The interaction of voltage and residence time on removal efficacies (%)

Table 2. ANOVA for the models for COD removal

Source	Sum of squares	df	Mean of square	F value	P-value	Prob>F
Model	16567.20	8	2070.90	16.79	0.0001	Significant
A (voltage)	4335.83	1	4335.83	35.15	0.0001	Significant
B (time)	3476.60	1	3476.60	28.18	0.0001	Significant
C (electrode)	4229.43	1	4229.43	34.28	0.0001	Significant
AB	121.99	1	212.99	0.99	0.3340	
AC	61.12	1	61.12	0.50	0.4910	
BC	65.92	1	65.92	0.53	0.4747	
A2	3320.94	1	3320.94	26.92	0.0001	Significant
B2	1456.31	1	1456.31	11.81	0.0032	Significant
Residue	2097.16	17	123.36			
Lack of fit	825.97	9	91.77	0.58	0.7849	Not significant
Pure error	1271.19	8	158.90			
Total	18664.36	25				

3.5. Reuse of MDF wastewater

In this study, treatment of MDF wastewater through optimization of the EC process is essential to reusing the water. EC aimed to make the reusing of the water possible for MDF industrial process by removing the pollutants. The results point to the considerable difference between the turbidity and TSS removal efficiency rates for common design with and without the EC process for MDF wastewater treatment plant. The high concentration of suspended solid and turbidity inevitably led to prevention of reusing the water in common conventional systems of MDF wastewater. It is anticipated that high concentration of iron ionic allowing to oxidize the functional group of cellulosic fibers would result in negative impact on flocculation of solid wastes, while the aluminum ionic was not observed. Otherwise, the aluminum electrode was confirmed to have significant impact on pollutant removal, even at a low level of concentration. The high level of pollutant removal helps reuse the water for MDF manufacturing process, which has not been feasible in previous systems.

4. CONCLUSIONS

This study was carried out with aim of reusing the water through Electrocoagulation Process (EC) in manufacturing MDF wastewater. The colloidal particles were a major problem associated with the MDF wastewater. EC coupled with wastewater treatment plant helped compensate the sludge generation and reduce the cost of operations. The effects of two important independent variables, namely voltage and residence time, with iron and aluminum electrodes on responses including the COD, TSS, turbidity, and TS were evaluated. It was anticipated that the amounts of aluminum hydroxide flocs would increase upon increasing the voltage, which led to the enhancement of colloidal particle removal. The aluminum electrode was more efficient because of better electrolysis which generated higher charge and was easily soluble to destabilize the colloidal particle in MDF wastewater. RSM was successfully used for optimizing the EC process variables. The optimum values of voltage and residence time were determined at 33 V and 25 min. Under optimum values of process variables, 93 % removal COD, 89 % removal TSS, 67 % removal turbidity, and 76 % removal TS were obtained.

Therefore, the pretreatment of MDF wastewater through electrocoagulation confirmed that the EC system could

perform biological treatment better than the current conventional physio-chemical treatments. The effluent of EC represented a significant difference in quality of water for reusing the water. In addition, it reduced energy consumption as well as process residence time. Removal of the chemical additives and low sludge production characterized a cost-effective approach of industrial wastewater management. It was also demonstrated that the EC process managed to reuse the water in the MDF manufacturing process.

5. ACKNOWLEDGEMENT

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