Application of Response Surface Methodology for analysis and optimization of the operational parameters for turbidity removal from oily wastewater by electrocoagulation process

Thamer Jasim Mohammed¹ Hadeel Atiya Al-Zuheri²

¹Chemical engineering department, University of technology, Baghdad, Email: thamer_jasim58@yahoo.com
²Chemical engineering department, University of technology, Baghdad, Email: Che.80318@uotechnology.edu.iq

Abstract. The purpose of this work was to study the effectiveness of electrocoagulation process for turbidity removal from oily wastewater. Synthetic oily wastewater was used for conducting batch experiments, and the turbidity introduced to the wastewater by (0.3 gm) of Sodium Bentonite giving an initial turbidity of (200 NTU). Anode of aluminum and cathode of Iron were immersed in 1.5 L solution. An experimental design of (52 run) based on a central composite design (CCD), and the analyze of data was carried out using response surface methodology (RSM) in (Design Expert software) for evaluating the effect and interaction of several parameters including: initial pH (5-9), initial oil concentration (50-500 ppm), current density (5-25 mA/cm²), NaCl concentration (40-400 ppm), and electrolysis time (10-30 min). The response variable was Turbidity%. The experimental results were fitted to a quadratic equation and analysis of variance (ANOVA) was also performed. Results of the ANOVA showed that the model fitted well with turbidity% giving less than 0.05 of probability of error (p), and the variables had a significant effect on the removal. The high R-squared of 88% indicate the high correlation between the predicted and experimental values. In addition, the estimated optimum conditions of the treatment system were (pH= 6, [oil]= 387.5 ppm, Current density= 20 mA/cm², [NaCl]= 310 ppm, and time= 25 min) giving a predicted turbidity% of 98% and experimental of 97.4%.

Keywords: Electrocoagulation, Turbidity, Optimization, Response Surface Methodology Central Composite Design

1. Introduction
Availability of clean and safe drinking water is one of the main difficulties the world is facing. According to World Health Organization (WHO), approximately 1.1 billion people don’t have drinking water. Therefore, improved and more economic strategies of water management involving methods of treatment become needed especially with the population increasing dramatically. Electrocoagulation is one of these method for water treatment, which is capable of treating different types of contaminants in drinking water (Adapureddy et al., 2012). One of the main problems which is associated with water is turbidity. It is produced by the existence of clay in water and/or other colloids. Those colloidal components are stable and can’t be removed by sedimentation and clarification (Abuzaid et al., 1998).

The traditional method of treatment involves of metal salts addition (iron, aluminum etc.), colloidal destabilization (coagulation), finally flocculation and sedimentation. This method has
undeniable disadvantages as addition of chemicals in large quantities, and large amount of sludge is produced which cause problems of disposal and water losses (Rahmani et al., 2008). Electrocoagulation is a variant of conventional coagulation process in which coagulant is generated from electrolytic dissolution of electrodes on application of electric current. Advantages such as energy efficiency, versatility, ease of operation, environmental compatibility and cost effectiveness have made EC an attractive method in wastewater treatment and water purification (Giwa et al., 2012). Electrocoagulation has been applied successfully to treat different wastewater contaminants, such as phenol (Abdelwahab et al., 2009), oil (Tir and Moulai-Mostefa, 2008), boron (Yilmaz et al., 2007), petroleum hydrocarbons (Moussavi et al., 2011), fluoride (Behbahani et al., 2011), black liquor (Zaied and Bellakhal, 2009). Moreover, using the same method more than 99% turbidity removal from dairy wastewater has been reported (Tchamango et al., 2010; Kushwaha et al., 2010).

Electrocoagulation involve the production of coagulants in situ by metal ions dissolution from anode as current pass with the coincident hydroxyl ions H₂ bubbles formation at cathode (Fadali et al., 2016). In a process of electrocoagulation, the coagulants are produced in situ including three steps (a) the electrolytic reaction on surfaces of electrode, (b) coagulants formation and (c) contaminates adsorption on coagulants, which then removed by flotation or sedimentation. Sacrificial electrode (usually Iron (Fe) or aluminum (Al)) produce metal ions by dissolution from anode. The metals ions hydrolyze to form polymeric aluminum or iron hydroxides, which are the coagulants (Safari et al., 2016). When aluminum electrode is used, the main reaction at anode is as follows (Safari et al., 2016):

\[ \text{Al}_{(s)} \rightarrow \text{Al}_{(aq)}^{3+} + 3 \text{e}^- \]  \hspace{1cm} (1)

H₂ production arises from cathode provides floatation effect for the flocculated particles to the wastewater surface. by the following reaction.

\[ 2\text{H}_2\text{O} + 2 \text{e}^- \rightarrow \text{H}_2(\text{g}) + 2 \text{OH}^-_{(aq)} \]  \hspace{1cm} (2)

The generated Al³⁺ ions interact with water and OH⁻ to produce hydroxides and polyhydroxides as follows (Mehtap et al., 2009):

- Monomeric species such as Al(OH)²⁺, Al(OH)₃⁻, and Al(OH)₄⁻
- Polymeric species such as Al₂(OH)₄³⁻ and Al₃(OH)₅⁴⁻, and Al₆(OH)₁₅⁻, Al₇(OH)₁₇⁴⁺, Al₈(OH)₂₀⁴⁺, Al₁₂O₄(OH)₃₄⁴⁻, and Al₁₃(OH)₃₄⁵⁻, which transform primarily into Al(OH)₃(s) in the solution (Gengec et al. 2012) as in reaction (3):

\[ \text{Al}^{3+}_{(aq)} + 3 \text{H}_2\text{O} \rightarrow \text{Al(OH)}_{3(s)} + 3 \text{H}^+_{(aq)} \]  \hspace{1cm} (3)

Response surface methodology (RSM) is usually used in chemical engineering, bioengineering, and pharmaceutical engineering (Hwang et al., 2015). Multiple regression equations can be used for fitting values of variables and responses, estimating the coefficients of regression equations, selecting optimum condition for the process to achieve the best response. The main objective of this work was to remove turbidity by using electrocoagulation process. Using the response surface methodology (RSM) for experimental design, system optimization was performed by varying the independent parameters.

2. Materials and methods

2.1 Wastewater Sample

Synthetic wastewater was prepared by dissolving (0.3 gm) of Sodium Bentonite which was provided by Petroleum Research and Development Center, Iraq in distilled water to give water a turbidity of (200 NTU). Sodium Bentonite has been characterized by (FTIR) spectroscopy for chemical functional groups in the Ministry of Science and Technology, Iraq. Figure (1) shows the FTIR spectrum of the
Na-bentonite sample. Bands at 467, and 521 cm\(^{-1}\) are corresponded to bending vibrations of (Si–O–Si) and (Al–O–Si). And band at 621 cm\(^{-1}\) is ascribed to coupled (Al–O) and (Si–O) out-of-plane vibrations. The existing of quartz in sample confirmed as band at 795 cm\(^{-1}\) appeared. Band at 845 cm\(^{-1}\) is ascribed to (AlMgOH) bending vibration, while spectral band at 885 cm\(^{-1}\) represent (Al–O–(OH)–Al) stretching vibration (Tan et al., 2008). The (Si–O–Si) groups of the tetrahedral sheet represented by the strong band at 1038 cm\(^{-1}\). Bending of (H–OH) bond of water molecules is reflected at band 1635 cm\(^{-1}\), and vibration of (H–OH) of the water molecules is represented by the band at 3361 cm\(^{-1}\). The band at 3626 cm\(^{-1}\), is a result of (O-H) stretching vibrations of (Si-OH) groups (Yu et al., 2017).

2.2 Electrocoagulation cell

Turbidity removal from synthetic wastewater was performed in a batch electrocoagulation cell with 2.5 liters’ volume and made of Perspex material. consists of two vertical plate electrodes (cathode made of Fe and anode made of Al), anode have effective surface area of 89.29 cm\(^{2}\)and a gap between 3 cm (Esraa et al., 2013). Batches of 1.5 litres of synthetic wastewater were introduced to the electrocoagulation cell with mixing speed of 150 rpm. Then the electrodes were connected to the DC power supply (MODEL: APS3005S). Experiments were performed by varying cell current (0 – 5 A; 0-30 V).

2.3 Evaluation of Removal Efficiency

The removal efficiency of turbidity in OWW after treatment with electrocoagulation is evaluated as follows:

\[
\eta\% = \left( \frac{C - C'}{C'} \right) \times 100
\]  

Where:

- \(\eta\%\) = turbidity removal efficiency.
- \(C\) = initial turbidity at (NTU).
- \(C'\) = Final turbidity at any time (NTU).

![Figure 1. FTIR spectrum of sodium bentonite](Image)
2.4 Experimental design

The CCD, which is the standard RSM, was selected in order to study and find the relationship between the response and studied factors, and for optimization. Knowing the constraint of the variables owing to their difference in units and/or difference in limits of variation, the variables were coded according to the following equation:

\[ X_i = \frac{x_i - X_{cp}}{\Delta x} \]  

(5)

Where X: corresponds to code level; Xpc: is the un-coded value at the central point; xi is the un-coded value, and Δxi is the change value between levels (Montgomery et al., 2005). The factors levels and codes are presented in Table (1). The response variable for the statistical analysis was turbidity removal efficiency. For the response surface method, the experimental results were adjusted to a second order multivariable polynomial as presented in Equation (6):

\[ Y_i = \beta_0 + \sum_{i=1}^{4} \beta_i X_i + \sum_{i=1}^{4} \beta_{ii} X_i^2 + \sum_{i=1}^{4} \sum_{j=i+1}^{4} \beta_{ij} X_{ij} \]  

(6)

Where, \( \beta_0 \), \( \beta_i \), \( \beta_{ii} \) and \( \beta_{ij} \) are the coefficients of regression for intercept, linear, quadratic and interaction terms, respectively; and \( X_i \), and \( X_{ij} \) are the independent factors. Response (\( Y_i \)) represents the percentage removal of turbidity (\( Y_1 \)).

The Design Expert Software (version 9) was used to generated (52) runs of experiments, build regression, and graphical analysis. The interactive effects of the factors on the response were illustrated by 3D surfaces plots. From these three-dimensional plots, the simultaneous interaction of two factors on the response was studied. Additional experiment was performed for verification of the model validity.

3. Results and discussion

3.1. Analysis of Design of Experiments

The design matrix with experimental and predicted turbidity removal efficiencies are listed in Table 2.
| 5 | 6 | 162.5 | 20 | 130 | 15 | 6.22 | 96.89 |
|---|---|-------|----|-----|----|------|------|
| 6 | 8 | 162.5 | 20 | 130 | 15 | 7.91 | 96.05 |
| 7 | 6 | 387.5 | 20 | 130 | 15 | 6.78 | 96.61 |
| 8 | 8 | 387.5 | 20 | 130 | 15 | 7.76 | 96.12 |
| 9 | 6 | 162.5 | 10 | 310 | 15 | 8.7  | 95.65 |
| 10| 8  | 162.5  | 10 | 310  | 15 | 9.5  | 95.25 |
| 11| 6 | 387.5 | 10 | 310 | 15 | 9.05 | 95.48 |
| 12| 8 | 387.5 | 10 | 310 | 15 | 11.35| 94.33 |
| 13| 6 | 162.5 | 20 | 310 | 15 | 7.04 | 96.48 |
| 14| 8 | 162.5 | 20 | 310 | 15 | 8.65 | 95.68 |
| 15| 6 | 387.5 | 20 | 310 | 15 | 7.55 | 96.23 |
| 16| 8 | 387.5 | 20 | 310 | 15 | 9.82 | 95.09 |
| 17| 6 | 162.5 | 10 | 130 | 25 | 6.14 | 96.93 |
| 18| 8 | 162.5 | 10 | 130 | 25 | 7.61 | 96.2  |
| 19| 6 | 387.5 | 10 | 130 | 25 | 6.7  | 96.65 |
| 20| 8 | 387.5 | 10 | 130 | 25 | 8    | 96    |
| 21| 6 | 162.5 | 20 | 130 | 25 | 5.14 | 97.43 |
| 22| 8 | 162.5 | 20 | 130 | 25 | 6.28 | 96.86 |
| 23| 6 | 387.5 | 20 | 130 | 25 | 5.83 | 97.09 |
| 24| 8 | 387.5 | 20 | 130 | 25 | 7.25 | 96.38 |
| 25| 6 | 162.5 | 10 | 310 | 25 | 7.12 | 96.44 |
| 26| 8 | 162.5 | 10 | 310 | 25 | 7.2  | 96.4  |
| 27| 6 | 387.5 | 10 | 310 | 25 | 6.54 | 96.73 |
| 28| 8 | 387.5 | 10 | 310 | 25 | 8.93 | 95.54 |
| 29| 6 | 162.5 | 20 | 310 | 25 | 5.92 | 97.04 |
| 30| 8 | 162.5 | 20 | 310 | 25 | 6.99 | 96.51 |
| 31| 6 | 387.5 | 20 | 310 | 25 | 6.54 | 96.73 |
| 32| 8 | 387.5 | 20 | 310 | 25 | 8.37 | 95.82 |
| 33| 5 | 275  | 15  | 220 | 20  | 9.06 | 95.47 |
| 34| 9 | 275  | 15  | 220 | 20  | 16.1 | 91.95 |
| 35| 7 | 50   | 15  | 220 | 20  | 6.99 | 96.51 |
| 36| 7 | 500  | 15  | 220 | 20  | 5.34 | 97.33 |
| 37| 7 | 275  | 5   | 220 | 20  | 13.56| 93.22 |
| 38| 7 | 275  | 25  | 220 | 20  | 5.34 | 97.33 |
| 39| 7 | 275  | 15  | 40  | 20  | 9.16 | 95.42 |
| 40| 7 | 275  | 15  | 400 | 20  | 9.57 | 95.22 |
| 41| 7 | 275  | 15  | 220 | 10  | 9.97 | 95.02 |
| 42| 7 | 275  | 15  | 220 | 30  | 3.75 | 98.13 |
| 43| 7 | 275  | 15  | 220 | 20  | 6.89 | 96.56 |
| 44| 7 | 275  | 15  | 220 | 20  | 8.12 | 95.94 |
| 45| 7 | 275  | 15  | 220 | 20  | 8.37 | 95.82 |
| 46| 7 | 275  | 15  | 220 | 20  | 8.05 | 95.98 |
| 47| 7 | 275  | 15  | 220 | 20  | 7.02 | 96.49 |
| 48| 7 | 275  | 15  | 220 | 20  | 4.99 | 96.51 |
| 49| 7 | 275  | 15  | 220 | 20  | 5.12 | 97.44 |
| 50| 7 | 275  | 15  | 220 | 20  | 5.04 | 97.48 |
| 51| 7 | 275  | 15  | 220 | 20  | 5.34 | 97.33 |
The following quadratic regression model for turbidity removal efficiency in terms of coded factors was obtained:

\[
\text{Turbidity\%} = +96.38 - 0.40*A - 0.13*B + 0.34*C - 0.16*D + 0.47*E - 0.088*AB - 6.562e-3*AC - 0.017*AD + 0.035*AE - 7.812e-3*BC - 0.047*BD - 7.812e-3*BE - 0.048*CD - 0.098*CE + 0.047*DE - 0.46*A^2 + 0.14*B^2 + 0.094*C^2 - 0.22*D^2 + 0.18*E^2
\]

where (A, B, C, D, and E) are coded values for the factors (pH, initial oil concentration, current density, NaCl concentration, and time) respectively. An empirical relationship was developed by the RSM where the response variable (Turbidity\%) was evaluated as a function of the five factors; effects of first-order (linear terms), and second-order (quadratic terms) and; interaction effects (interactive terms). The result of Analysis of Variance (ANOVA) for turbidity\% is shown in Table (3). ANOVA gives statistical results and diagnostic checking tests that enable the evaluation of adequacy of the models. The table demonstrates that the suggested quadratic model was significant since the p-value was less than 0.05 (p-value < 0.0001). A non-significant lack of fit (used for comparison of the deviation of real points from the fitted surface relative to pure error) is required to demonstrate the applicability of the model in order to predict the response variables (Mook et al. 2013). The calculated p-value of lack-of-fit was (0.7025) for turbidity\% is more than 0.05; thus, there are no statistically significant evidences that the model doesn’t represent the results at confidence level of 95%. Furthermore, high R² values of 88\%, express a high correlation between the predicted and actual values.

**Table 3.** The results of ANOVA for Response Surface Quadratic model turbidity\%
Also the “Predicted R2” is in reasonable agreement with the “Adjusted R2”. The low standard deviation of (0.37) is certainly another evidence of the adequacy of quadratic model. Coefficient of variance (C.V) (ratio of the standard error of estimated to the mean value of the observed response) describes the model reproducibility. If the model C.V is less than 10%, then the model is considered reproducible (Chowdhury et al., 2012). The results in Table 3 show that the model has C.V= 0.3, thus its considered to be reproducible. Figure (2a) show the normal probability plot. It can be seen in this plot that the points give fairly straight line. While Figure (2b) is a plot of the residuals versus predicted. In this figure, there is no patterns such as increasing residuals with increasing fits or domination of negative or positive residuals. Thus, Figure (2) shows that the model is adequate to describe turbidity% by RSM. In addition, Fig. (3) shows the actual versus predicted values for turbidity%. The figure indicates good agreement between the predicted and observed values.

3.2 Effect of the Factors on turbidity removal

The effect of the main variables and their interactions on the turbidity% response can be understood by ANOVA. It can be seen in Table 3 that all the studied parameters had significant effect on turbidity removal including (pH, initial oil concentration, current density, NaCl concentration, and time) as their P-value as shown in the table is less than 0.05. Additionally, it was found that square terms of (pH, initial oil concentration, and time NaCl concentration, and time) were significant to the response and the interaction terms have negligible effect.
Also, these effects are represented by the coefficients in the quadratic model (Eq. (7)). When coefficients have negative sign (-) means that the factors have antagonistic effect (the response decreases by varying factors), and a plus sign (+) shows a synergistic effect factors (the response increases with varying factors). It was observed that current density (C), and time (E), have positive effects on turbidity removal, and the other parameters have a negative effect (Kermet-Said et al., 2015). For a better explanation of the main and the interaction effects, 3D plots are also presented in Fig. (4). 3D plots of the RSM are drawn as a function of two factors at a time, holding all other factors at fixed levels. The 3D surface graph, of pH versus (initial oil concentration, time, current, and NaCl) is shown in Fig. (3 (a, b, c, d)) respectively. It is clear from the figures that the turbidity% increase at the condition where pH range from 5 to 6. It was found that colloidal particles in the pH range 6-7 contribute to the formation of amorphous hydroxide precipitates and other iron hydroxo complexes with hydroxide ions and polymeric species (Ahmadi et al., 2012). As pH increased to 8, a decrease in turbidity% is observed due to the less formation of the reactive flocs of aluminum hydroxide and its solubility increases as the solution becomes more acidic (Tezcan et al., 2009). In the basic range, aluminum hydroxide ions may form negatively-charged ions which allow less effective coagulations (Ahmadi et al., 2012).

Increasing current density or time caused increasing turbidity removal efficiency as seen in the figures. This is expected, as by increasing the current or time, rate of anode dissolution increases based on Faraday’s law, thus concentration of metal ions in the solution increases too, and consequently the turbidity removal rate is enhanced. In addition, an increase in bubble density occurs at high current, which also results in a greater flux and faster removal of pollutants (El-naas et al., 2013). It’s clear from figures that Increasing initial oil concentration led to decrease in turbidity%. This may be ascribed to the interface between oil removal and turbidity removal since at higher concentration efficiency of electrocoagulation to remove oil increase because of the increasing of the neutralized oil drops collision frequency with the increasing of oil concentration which results in the formation of larger and easy to float drops (El-Ashtoukhy et al.,2011).

As NaCl increased turbidity% increased too as seen in Fig. (4) but when [NaCl] increased to 220 ppm turbidity removal efficiency decreased which mean (220 ppm) is the optimum concentration of NaCl for turbidity removal is 130 ppm. As NaCl increased Cl- concentration in the solution increased which help in destroying the oxide film of Al2O3 that covers the Al anode and preventing Al3+ dissolution into the solution (El-Ashtoukhy et al., 2010). But higher NaCl concentration, cause improper dissolution of aluminum due to ‘corrosion pitting’, causing undesired interaction between coagulants and particles (Sánchez-Calvo et al., 2003). The 3D surface graphs for turbidity% in Fig. (4 (a, b, c, e, i, j)) give a saddle shape which indicate that the highest turbidity% is obtained generally at the intersection of fixed levels of all of the factors. While figures (4 (f, g, h)) illustrate a falling rigid with minimum outside the experimental region. A mound shape contour with maximum response at the central demonstrated in Fig. (4d) of (pH Vs. NaCl). interaction, the circular shape contours suggest the independence of factors effect (Kuehl et al., 2000).
Figure 4. 3D surface plots of interaction between factors: (a) pH and oil concentration, (b) pH and time, (c) current density and pH, (d) NaCl and pH, (e) NaCl and time, (f) Time and current density, (g) current density and oil concentration, (h) Time and oil concentration, (i) Time and NaCl, (j) NaCl and oil concentration. Hold values: (initial pH =7, initial oil concentration =275 ppm, current density = 20 mA/cm², and time = 15 min).
Based on these figures maximum turbidity% of 97% was obtained in at pH range (5.8- 7.5), current range (21-25 mA/cm²), oil concentration (50 ppm), NaCl range (160-220 ppm), and time (25-30 min)

4. Optimization Analysis
Additional verification experiments were conducted at optimum conditions to confirm the validation of the experimental model. The optimum conditions of investigated factors were determined by solving the regression equation in (Design Experts software). The optimum values of the test variables selected were as follows; The optimum condition was found to be: (pH= 6, [oil]= 387.5 ppm, Current density= 20 mA/cm², [NaCl]= 310 ppm, and time= 25 min). While the response predicted was turbidity% = 98%. This model prediction from the regression equation agreed reasonably well with the data from the verification experiments. Based on optimized experimental condition turbidity% of 97.4% was achieved under optimized condition.

5. Conclusion
In this study the process of electrocoagulation was used for the removal of turbidity from oily wastewater using aluminum anode. Central composite Design was effectively used for the experimental design and analyzing the results. The obtained correlation coefficient $R^2$ was found equal to 0.88 for turbidity removal, which indicated that the actual results fit relatively good with the predicted results by the application of the quadratic models. Furthermore, the ANOVA shows the significance of the model, since the probability value was very low. Moreover, 3D response surface graphs were presented to investigate the optimum condition of the studied variables. The study shows that turbidity removal reached a turbidity% up to 97% at optimum condition (pH= 6, [oil]= 387.5 ppm, Current density= 20 mA/cm², [NaCl]= 310 ppm, and time= 25 min).

6. References
[1] Abdelwahab, O., Amin, N. K., & El-ashtoukhy, E. Z., (2009), “Electrochemical removal of phenol from oil refinery wastewater”, 163, 711–716.
[2] Adapureddy, S. M., & Goel, S. (2012). Optimizing Electrocoagulation of Drinking Water for Turbidity Removal in a Batch Reactor. International Conference on Environmental Science and Technology IPCBEE, 30, 97–102.
[3] Ahmadi, S., Sardari, E., Javadian, H. R., Katal, R., & Sefti, M. V. (2013). Removal of oil from biodiesel wastewater by electrocoagulation method. Korean Journal of Chemical Engineering, 30(3), 634–641.
[4] Behbahani, M., Alavi Moghaddam, M.R., Arami, M., “Techno-economical evaluation of fluoride removal by electrocoagulation process: Optimization through response surface methodology”, Desalination, 217, 209-218, 2011.
[5] Chowdhury, S.R., Yanful, E.K., (2010), “Arsenic and chromium removal by mixed magnetite-magnehemite nanoparticles and the effect of phosphate on removal”, J. Environ Manage 91:2238–2247.
[6] El-Ashtoukhy, E., & Yasmine, F., (2014) “Oil Removal from Oil-Water Emulsion by Electrocoagulation in a Cell with Rotating Cylinder Anode”, The Electrochemical Society of Japan, 82(11), 974–978.
[7] El-naas, M. H., Al-zuhair, S., & Al-labaney, A. (2013). Treatment of Petroleum Refinery Wastewater by Continuous Electrocoagulation, 2(10), 2144–2150.
[8] Esraa, R.A., (2015), “A Comparative Study of Coagulation-Flocculation and Electrocoagulation in the Treatment of Produced Water”, MSc Thesis, University of Technology, Chemical Engineering Department, Iraq.
[9] Fadali, O. A., Ebrahem, E. E., El-Gamil, A., & Altaher, H. (2016). Investigation of the electrocoagulation treatment technique for the separation of oil from wastewater. Journal of Environmental Science and Technology, 9(1), 62–74.
J. R., Kocakerim, M. M. (2007). "Removal of Petroleum Hydrocarbons from Oil Water Sludge Using Response Surface Methodology by Electrocoagulation". Desalination, 209, 321-324.

[22] Tan, X.L., Hu, J., Zhou, X., Yu, S.M., Wang, X.K., (2008). "Characterization of Linan Montmorillonite and its Application in the Removal of Ni(II) from Aqueous Solutions". Radiochim Acta, 96, 487-495.

[23] Tchamango, S., Nanseu-Njiki, C.P., Ngameni, E., Hadjiev, D., "Treatment of Dairy Effluents by Electrocoagulation Using Aluminum Electrodes". Science of the Environment, 408, 947-952, 2010.

[24] Tezcan, U., Uğur, S., Koparal, A. S., Bakir Oguşveren, U., (2006) "Electrocoagulation of Olive Mill Wastewaters", Sep. Purif. Technol., 52 (1), 136-141.

[25] Tir, M., and Moulai-Mostefa, N. (2008) “Optimization of Oil Removal from Oily Wastewater by Electrocoagulation Using Response Surface Method.” Journal of Hazardous Materials, Vol. 158, No. 1, pp: 107-115.

[26] Yılmaz, A.E., Boncukoğlu, R., Kocakerim, M.M. 2007. “An empirical model for parameters affecting energy consumption in boron removal from boron-containing wastewaters by electrocoagulation”, Journal of hazardous Materials, 149, 475-481, 2007.

[27] Yu, X., (2017), “Impact of environmental conditions on the sorption behavior of Pb (II) in Na-
bentonite suspensions”. Journal of Hazardous Materials, 183(1–3), 632–640.
[30] Zaied, M., Bellakhel, N., “Electrocoagulation treatment of black liquor from paper industry” Journal of Hazardous Materials, 163, 995–1000, 2009.