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The leachate treatment system of combined O_3 oxidation, and activated carbon adsorption

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Abstract

Treatment of organic leachate is one the most controversial topics around the world which led this study to assess the efficiency of the combined oxidation and adsorption treatment (COAT) process in the treatment of leachate by considering local experiments. The removal of effluent parameters (TDS, COD, BOD) was enhanced by oxidizing the GAC surface as a catalyst with NaOH before the process and by ozone within the procedure as well. Assessing the interacting effect of operating variables (i.e., ozone concentration, GAC density, reaction time and pH) provides valuable information for optimization. Response Surface Methodology (RSM) was employed. The optimized model's circumstances are the reaction time of 30.77 min, ozone dosage of 141.29 mg/l, pH of 7.2, and the GAC density of 1.29 gr/cm3 with the predicted removal percentage of 51.63%,62.84% and 56.13% for TDS, COD and, BOD respectively.

key words

Catalytic ozonation

Organic wastewater treatment

Ozone-Activated Carbon adsorption

Organic leachate

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1.Introduction

Nowadays, one of the hot topics of world is the deliberation of leachate, which is challenging to purify for many experts. Considerable organic and inorganic matters are included in leachate such as xenobiotic organic compounds, refractory constituents, heavy metals, ammonia nitrogen, and other toxicants (Cassano et al., 2011; Pivato et al., 2006). Through soil and subsoil, untreated leachate could percolate leading to the adverse effects on receiving waters (H.-S. Li et al., 2009; S.Q. Aziz et al., 2011). Large sums of organic wastes are created daily in large cities. Thus, the beneficial disposal of these biodegradable compounds is controversial. Leachate involves a high sum of pollutants with complex and expensive treatments, which needs different and blended processes. Thus, disposal and treatment of leachate should be handled carefully. In sustainable advancement, organic wastewater treatment becomes a severely critical problem. However, it includes several toxic and refractory organic pollutants. Industrialization as well as inappropriate waste management leads to the huge deal of accumulated kitchen and foods waste (Sindhu R et al., 2019). The vegetable and fruit industry has been rapidly expanded with the economic development and fast structural reform of agriculture worldwide, making major problems of disposing of a heavy deal of fruit and vegetable waste for several countries. This waste is generally caused by production, transport, storing, distribution, and consumption of vegetables and fruits (Ji C et al., 2017). In Tunis, the whole waste is 6 tons per day in general, in Mercabarna it is 90 tonnes per day near Barcelona, Spain, while in India, it is 15,000 tons per day (Bouallagui H et al., 2005). In Central de Abasto, in Mexico City, the total of waste is 895 tonnes per day (Garcia-Peña et al., 2011). Leachate includes several organic matter, ammonia, nitrogen, inorganic salts, and metal ions (Zhao J et al., 2013). This is mainly true for numerous urban settings such as Tehran with food waste as the main proportion of municipal solid waste. Now, it poses heavy pressures on the depleting landfill space (Tsui TH et al., 2020). The biological treatment, as one of the conventional chemical, biological, and physical treatment methods (B.P. Naveen et al., 2017), is extensively utilized for the effectively removing the nutrients since it is a cost-effective method, with recalcitrant organic fractions as well as heavy metals left behind (J. Wiszniowski et al., 2006; L. Miao et al., 2019). Though, the recalcitrant organic matter can be oxidized and mineralized by chemical oxidation systems, particularly advanced oxidation procedures (AOPs) (A. Gupta et al., 2014; F.J. Rodríguez et al., 2016). However, AOPs are high cost leading to the secondary pollution (Cassano et al., 2011; C. Di Iaconi et al., 2006). The requirement for eco-friendly technologies creates no more hazardous by-products. Thus, it resulted in an incremented interest in using AOP like Fenton's oxidation. Thus, for effectively treating mature leachates with higher strength at an affordable cost, the best treatment system is a combination of physical approaches and chemical procedures (A.I. Gomes et al., 2019; T.F.C.V. Silva et al., 2013; V.J.P. Vilar et al., 2011; Z. Liu et al., 2015, Nabavi et al., 2021). The combined oxidation and adsorption treatment (COAT) technique through Ozone oxidation integrated with GAC (Granular Activated Carbon) adsorption is a mature technology to recover energy and various resources from different organic waste streams.

Owing to its adsorption features, a large porous volume is extensively used for water treatment along with a vast surface area within the range 1000 to 1300 m^2/g, granular activated carbon (GAC). Within adsorption, a material is moved from the liquid phase to a solid's surface and becomes bound by physical or chemical interactions. It is cost-effective and easy to operate as a result of the lower energy demand. Moreover, leachate treatment through GAC may be achievable for meeting the strict discharge standards increasingly for tenacious pollutants (Kurniawan, T.A.et al., 2006b). Ozone can convert contaminants into harmless materials in a short time. Dissimilar to chlorination, secondary contaminants are not produced by ozonation in the environment since lower molecular weight compounds like acetic acid is caused by the ozonation of organic compounds in wastewater (J.J. Wu et al., 2004).

2. Materials and methods

2.1. Materials

After sampling in polyethylene carboys (20 L), they were tightly closed and kept in the refrigerator at 4°C to minimize the consequent possible alterations in its physical-chemical and biological properties before examination. The leachate was immediately characterized in terms of the standard approaches (L.S. Clesceri et al., 1998) and the parameters of pH, NH_3-N, COD, BOD5, alkalinity (as $CaCO_3$), total nitrogen, alkaline metal cations,

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NO_3-N, total organic carbon (TOC), and conductivity. Before treating, to measure pH of the raw leachate specimens, a pH meter model Orion 710A (Texas, US) was used. Its impacts on the elimination of NH_3-N and COD during treatment was studied by setting the pH via 0.1 M HCl and 0.1 M NaOH. A spectrophotometer model Spectronic 4001 (Nevada, US) was used to analyze the concentrations of COD and NH_3-N. However, conductivity of the leachate and the total organic carbon (TOC) were determined utilizing a conductivity meter type Lutron CD4303 (California, US) and a TOC analyzer type Shimadzu 5000A (Minnesota, US), respectively. Otherwise, all the reagents and chemicals with analytical grade were supplied by Aldrich (Missouri, US). Using distilled-deionized water, all the reagents and working solutions were prepared. Standard solutions were freshly prepared from 30% (v/v) H_2O_2 without pH adjustment, by dilution of the stock solution to the prestated concentrations.

2.2. Experimental setup

The same glass columns (h: 200 cm; i.d: 5 cm) was packaged in a fixed bed evaluation, with various adsorbent values. Using 1 cm of glass wool and a layer of glass beads, the below part of the tank was fitted. The leachate was relocated from the upside of the tank to the down with no pre-treatment. The purified effluent was stored periodically for COD or TDS analysis. After reaching the saturation point, column operations were ended; i.e., $Ce/C_0 = 1$. C_0 and Ce represent the initial concentration and equilibrium of TDS and/or COD, respectively in leachate (mg/l). For the next ozonated leachate treatment, the same method was utilized (B. Morawe et al., 1995). The NaOH-modified GAC was then utilized in the combined process. The leachate first moves to the Ozonation tank via a pump, while inserting ozone from the bottom of the tank via the ozone generator. A good opportunity is provided for ozone using a stirrer along with homogenizing the leachate, to integrate and decompose the leachate. Then, through the pipes, the oxidized leachate composition moves to the retention tank to permit suitable contact time for reactions. The composition is inserted in the GAC reactor via a pump (NaOH modifies GAC particles to enhance performance) for completing the purification processes and adsorption in this tank.

2.3. Statistical analysis

The reliability, precision, and repeatability of the collected data were guaranteed through experiments in at least 3 issues to provide an average amount of 3 data sets. The data were overlooked by trespassing the error of 1.0%. Thus, the fourth experiment was conducted to achieve the admissible error limit. RSM was utilized in terms of CCD design for establishing a numerical model with interaction terms for COAT procedure at a probability level of 5%. Then, investigation was performed on the impacts of four independent variables in the COAT process including, inlet ozone dose (X_1) (25-200 mg L^{-1}), reaction time (X^2) (0-60 min), the GAC density (X3) (0.2-2 g/cm^3), and pH (X_4) (3-9). Three response variables included TDS (Y_1), COD (Y_2), and BOD (Y_3). Thirty-nine performances were included in the experiments with eight axial points and 6 central points ($\alpha = 1$). The RSM used specific experimental design combinations to look for optimum efficiencies from a particular sort of response factors and variables. Then, fitting a second-order polynomial (Eq. 1) was considered for the experimental data, as:

$$y= \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_4 x_4 + \beta_{11} x_1^2 + \beta_{22} x_2^2 + \beta_{33} x_{32} + \beta_{44} x_4^2 + \beta_{12} x_{12} + \beta_{13} x_{13} + \beta_{14} x_{14} + \beta_{23} x_{23} + \beta_{24} x_{24} + \beta_{34} x_{34} \quad \text{Eq. (1)}$$

where y denotes one of response variables and β_0 is constant. The linear effects regression coefficients are represented by β_1, β_2, and β_3. The quadratic coefficients are represented by β11, β22, and β33. Moreover, β_{12}, β_{13}, and β_{23} denote the interaction coefficients.

3. Results and discussion

3.1. Comparison between methods

To better compare different methods and find the optimal interval to remove the output parameters (TDS, COD, BOD), three methods were compared, namely integrated ozone-GAC adsorption, ozonation alone, and COAT process, taking into account all 39 experiments performed. Figure 1 compares the performance of these three methods in COD removal. Based on using ozonation alone, the removal of COD with an initial COD of 12175 mg/l improved from 13.30% to 34.76% with incrementing the dose of ozone from 25 to 200 mg/l. This is caused by the fact that in the leachate, the recalcitrant organic compounds became less available with continuing oxidizing of ozone, since it was oxidized. Therefore, decomposing the remaining organic compounds after ozone oxidation

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was difficult. No considerable growth was found in removal of COD after utilizing a particular ozone dose. The reason is the humic substances mostly in the fixed wastewater with less susceptibility to ozonation. Moreover, the substances are less aromatic and more aliphatic (F. Wang et al., 2004). By improving the performance of these methods between different tests, removal by the ozone-GAC technique has improved from 21.06% to 54.72%, indicating its relatively weaker performance than the COAT method by improving the removal percentage from 30.84% to 68.37%, indicating efficiency Suitable for COAT method in COD removal. The purified wastewater must possess fewer than 500 mg/l COD, based on the local laws for the maintenance of groundwater against pollution by wastewater. It was indicated that the COAT process treatment cannot create effluent after local regulations. Figure 2 compares the performance of these methods in TDS removal for each test separately. As shown in Figure 2, the COAT method performed best with a removal interval of 32.09-59.47%, while the integrated ozone-GAC adsorption method came in second with a removal interval of 20.13-39.57%. In the meantime, removal of TDS by Ozone alone represented a 10.79% to 24.57% improvement by rising the ozone concentration from 25 to 200 mg/l. Figure 3 shows the performance of these three methods in BOD removal. In the BOD removal procedure for ozonation method, the removal improvement of this parameter changes from 5.44% to 24.64% beginning from an ozone dose of 25-200 mg/l. Considering all the tests and displaying them as a plot, the integrated ozone-GAC adsorption method, with an improvement of BOD removal percentage from 20.36% to 49.05%, is in the second place after the COAT method with an improvement removal percentage from 22.04% to 67.20%.

A closer look at the plots reveals that the COAT and Ozone-GAC plots are very close in Figures 1 and 3 (COD and BOD removal). Having more points of intersection than the same plots in Figure 2, they show a more similar performance. From these figures, it can be seen that the performance of COAT and Ozone-GAC methods in BOD and COD removal has been much closer to each other, while these two methods are located at a greater distance from each other in the TDS removal plot. Thus, the COAT method has outperformed the Ozone-GAC method in TDS removal compared to other parameters (BOD and COD). Figure 4 compares the performance of the Ozone-GAC and Ozone alone methods. For this purpose, the percentage change created for all experiments between Ozone-GAC and Ozone alone methods was calculated and also the output parameters (COD, BOD, TDS) were compared. As shown in Figure 4, the most changes and differences between Ozone-GAC and Ozone alone can be seen in the TDS parameter. According to Figure 4, the Ozone-GAC method performed better in competition with Ozone alone in removing TDS (compared to

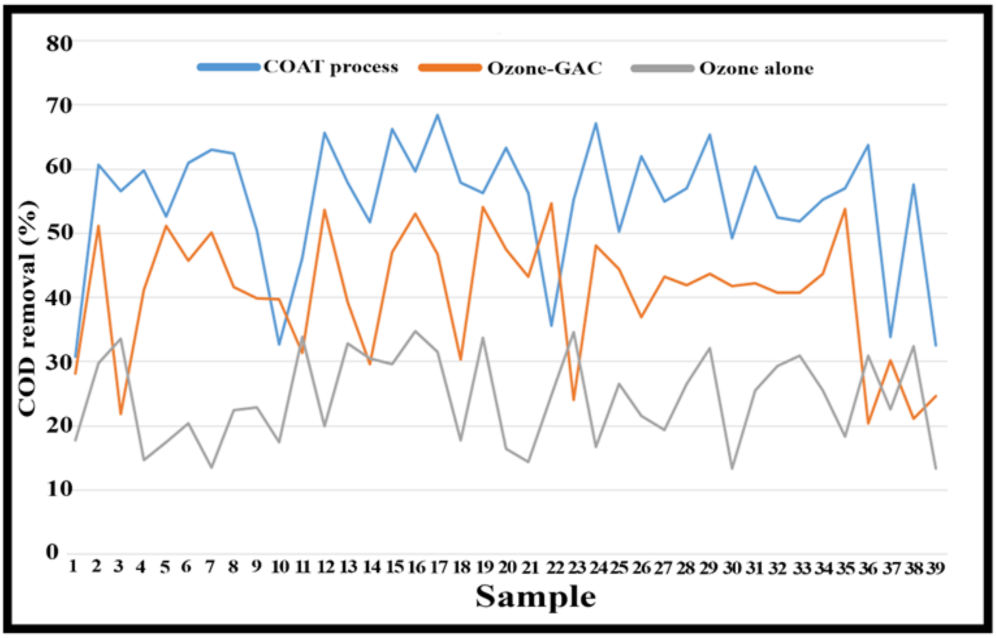

Fig. 1. Comparing three methods (Ozone-GAC, Ozone alone, COAT process) in the COD removal (Time: 60 min, flow rate:1.8 L/min, pH:7.2).

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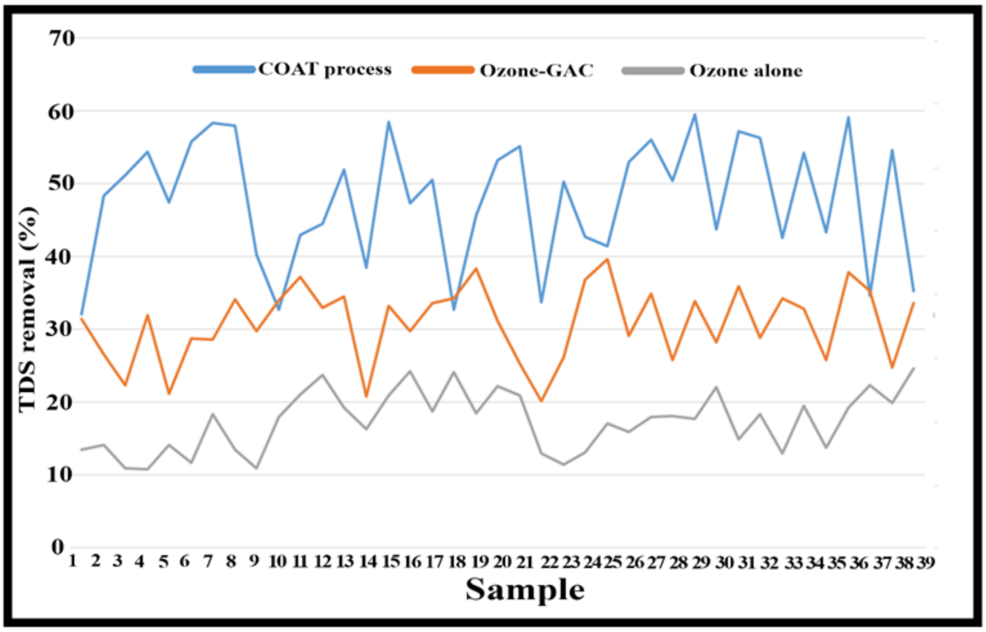

Fig. 2. Comparing three methods (Ozone-GAC, Ozone alone, COAT process) in the TDS removal (pH:7.2, flow rate:1.8 L/min, Time: 60 min).

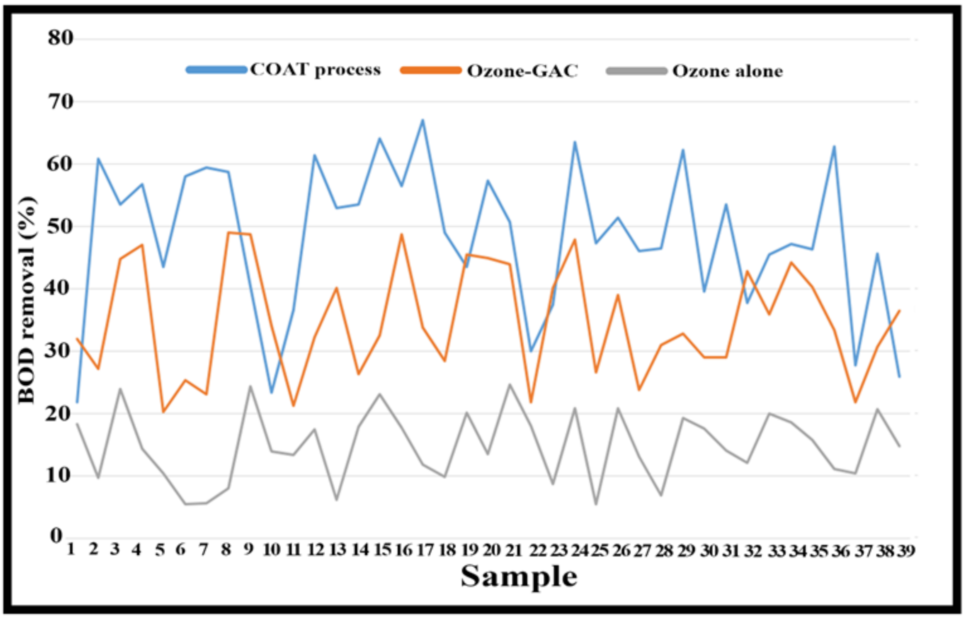

Fig. 3. Comparing three methods (Ozone-GAC, Ozone alone, COAT process) in the BOD removal (Time: 60 min, flow rate:1.8 L/min, pH:7.2).

COD and BOD). On the other hand, this means that by adding GAC adsorbent to the Ozone alone process, a much higher percentage of TDS removal can be expected.

Ozone dose effects were investigated by keeping other parameters constant. In the oxidation procedure (pH=7.2, GAC density=1 gr/cm3), the efficiency of COD/TDS/BOD removal is changed according to the dose of oxidant. Hence, the optimum dose of ozone was investigated to attain the maximum of output parameters removal under equilibrium circumstances. The effect of ozone dose on TDS, BOD, and COD removal performance followed by ozone-GAC method, and the COAT process were measured. It was also measured that integrating the ozone-GAC adsorption treatment significantly enhanced the COD removal from 18.38% to 48.19% and the COAT process enhanced the COD removal from 25.32% to 53.44% at the concentration the same as COD

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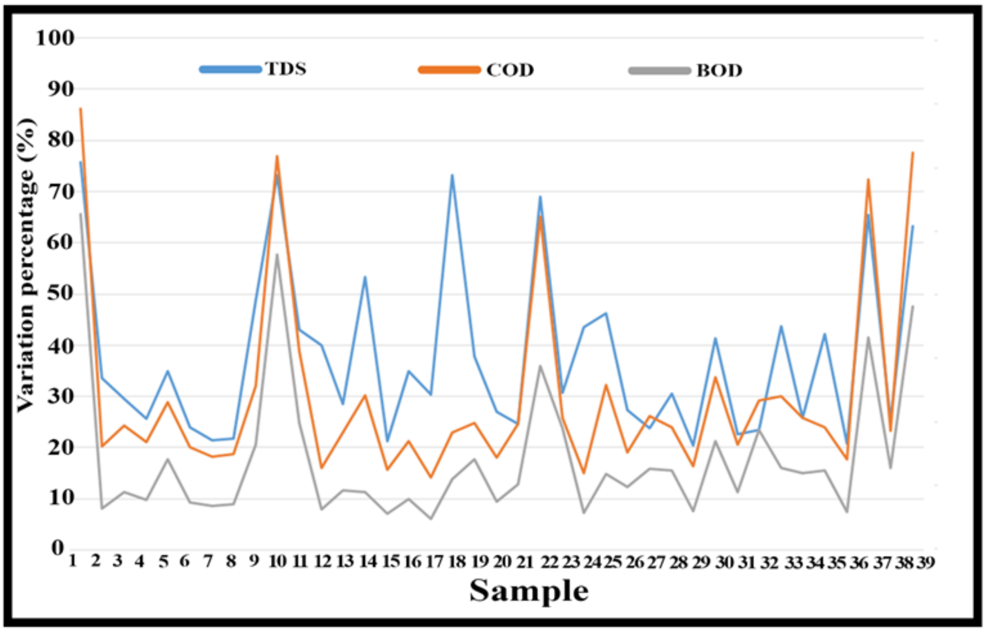

Fig. 4. Percentage of variation in output parameters (TDS, COD, BOD) between two approaches (Ozone GAC and Ozone alone).

(12175mg/L).

Likewise, the removal of TDS was significantly improved from 17.04% to 31.81% and from 28.23% to 45.69% respectively when the dose of ozone in the ozone-GAC adsorption treatment and the COAT procedure incremented from 25 to 200 mg/l.

In the meantime, the Ozone-GAC method via the same dose varies in the BOD removal rate from 17.73% to 43.29%, and the COAT method shows the rising rate from 18.69% to 58.19%, indicating the combined method's better performance.

3.2. Analytical techniques in RSM

Using the Design Expert Software, experiments were designed statistically and data analysis was performed. The effects of four independent variables on the response performance were detected using the second-order CCD and RSM designs. Experimental data were obtained in terms of 39 normalized observations. The ozone concentration (A), time (B), GAC density (C), and pH (D) were the evaluated variables. Using CCD, the interaction between various factors was recognized (Shi, X et al., 2020). Five levels of -α, -1, 0, +1, and +α were considered for the independent variables. Considering the previous studies and pilot investigations, the range was recognized. According to Bianco et al. (Bianco, B et al., 2011), such codes could be largely used to fit regression models leading to the variables within the range of –α to +α . The regression parameters ANOVA for the approximated response surface quadratic models as well as other statistical parameters for COD, BOD, and TDS are presented in Table 1. The total response variation estimated by this model is represented by R2 coefficient (COD=0.9105, TDS=0.8538, and BOD=0.8108). It shows the ratio of the summated squares gained through regression in comparison to their total sum.

3.2.1. TDS removal

TDS removal of 32.1-59.47% was obtained by the COAT procedure. The results of ANOVA were utilized for assessing the findings and investigating the "goodness of fit". Using the empirical results, an empirical formula was developed associated with the variables' response for TDS removal through the COAT process. According to the results, the TDS prediction's overall error with the RSM technique was 3.9% in comparison to the experimental data. Furthermore, it was indicated that the TDS removal through the experimental data and formula has better consistency. It is indicated that the existing experiential formula was can estimate the removal of TDS and provide a gentle rational consistency. For prediction of the TDS removal, an experimental association was developed based on all variables. The present association can be used for quickly assessing the TDS removal in organic leachates via the CAOT process. For this purpose, Eq. (2) is proposed.

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Table 1 Regression analysis in RSM model.

TDS			
Regression parameters	Magnitudes	Regression parameters	Magnitudes
Std. Dev.	612.29	R-Squared	0.8538
Mean	7297.18	Adj R-Squared	0.8262
C.V. %	8.39	Pred R-Squared	0.7885
PRESS	1.907E+007	Adeq Precision	14.107
COD			
Regression parameters	Magnitudes	Regression parameters	Magnitudes
Std. Dev.	397.44	R-Squared	0.9105
Mean	6190.77	Adj R-Squared	0.8515
C.V. %	6.42	Pred R-Squared	0.7983
PRESS	9.054E+006	Adeq Precision	15.981
BOD			
Regression parameters	Magnitudes	Regression parameters	Magnitudes
Std. Dev.	380.89	R-Squared	0.8108
Mean	4190.77	Adj R-Squared	0.8019
C.V. %	9.09	Pred R-Squared	0.7698
PRESS	9.686E+007	Adeq Precision	15.366

$$TDS(Y) = +6149.45 - 1189.01\ A + 56.40\ B - 485.03\ C - 126.30\ D - 206.25\ (AB) - 1525.82\ (AC) + 1595.09\ (AD) + 442.27\ (BC) + 1040.87\ (BD) - 1525.82\ (CD) + 1376.43\ A^2 + 527.67\ B^2 - 331.27\ C^2 + 250.36\ D^2 \quad \text{Eq. (2)}$$

where Ozone dose, time, GAC density, and pH are denoted by A, B, C, and D respectively, and parameter of Y shows the response (the predicted removal of TDS percentage). The model was evaluated at a confidence level of 95% for the p-value. The fit polynomial model value was represented by R^2 and Adj R^2, and the actuarial importance was confirmed by Fisher's F-test (Umar M et al., 2010). The regression was significant statistically (F-value: 8.70), for the strongly less possibility for the organic wastewater's degradation value (p-value less than 0.0001). The punctuality of the second-order multinomial credibility as well as the model were represented by higher R2 values. Moreover, according to the satisfactory accuracy over 4, the model can be used to plan the design space by the CCD. Based on the F-value of 8.70, the model was significant statistically (p-value less than 0.0001). Such a "Model F-Value" has the occurrence probability of only 0.01% owing to the noise element.

3.2.2. COD removal

The effects of various parameters on the COD removal are measured using Design-Expert software, 3D plots, and contour plots. The COAT method recorded a range of 30.84% -68.37% for the COD removal. An experimental equation was made using experimental results, to solve the COD removal variables via the ozone-GAC process. The overall error of COD estimation was found 4.1% as the RSM method in comparison to the experimental data. The experimental equation is:

$$COD(Y) = +4814.16 - 1434.90\ A + 132.22\ B - 302.59\ C + 359.36\ D + 733.20\ (AB) + 101.77\ (AC) + 787.93\ (AD) - 1303.05\ (BC) + 1183.20\ (BD) - 1187.75\ (CD) + 1622.63\ A^2 + 623.99\ B^2 + 452.38\ C^2 + 213.09\ D^2 \quad \text{Eq. (3)}$$

where A, B, C, and D are Ozone dose, time, GAC density, and pH, respectively, and the response represents Y (the predicted COD removal percentage). The regression was significant statistically (F-value: 10.33) based on the lower R^2 values and probability for the organic leachate degradation (p-value < 0.0001). A (Ozone dose), C (GAC density), A^2 (Ozone dose were significant model terms, AB (Ozone dose: time), AC (Ozone dose: GAC density), and BC (time: GAC density). The model's significance was revealed by "Lack of Fit F-value" of 4.22. For "Lack of Fit F-value", a probability of 5.77% was found owing the noise element.

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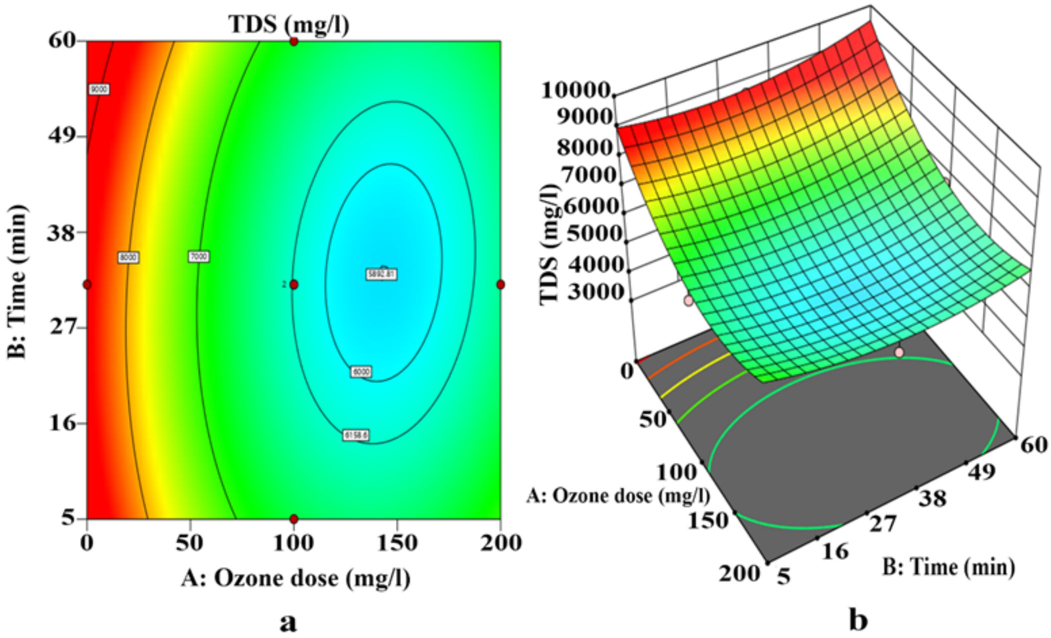

Fig. 5. a) Contour plot and b) RSM analysis in terms of the effects of Ozone dose and time on the TDS removal.

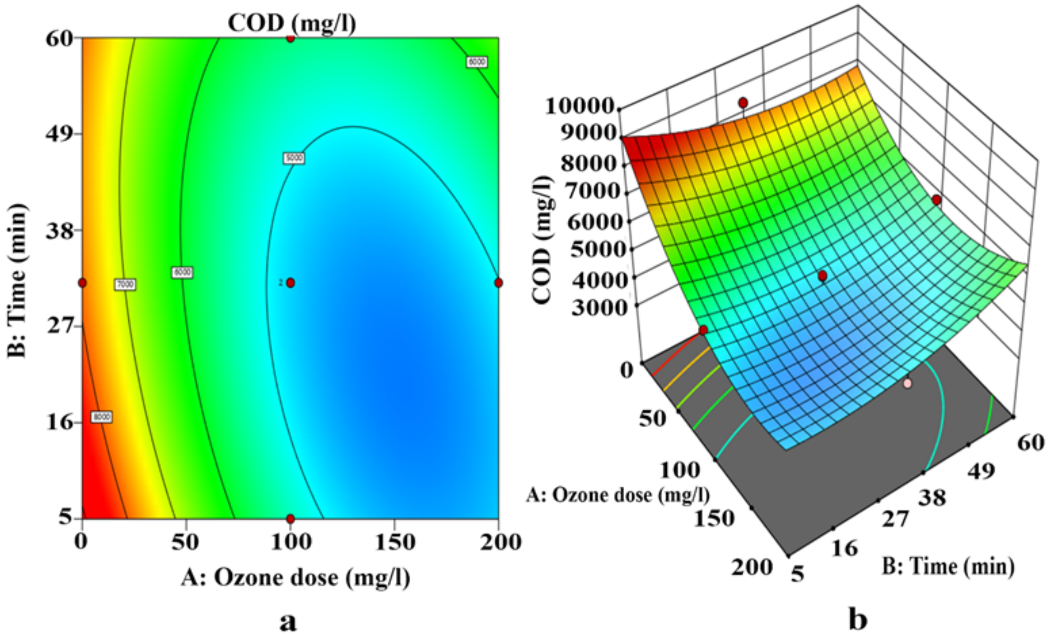

Fig. 6. a) Contour plot and b) RSM analysis in terms of the effects of Ozone dose and time on the COD removal.

4. Conclusion

Pursuant to the results, the performance of the COAT method is acceptable to treat the leachate, and utilizing this process is totally recommended to purify all kinds of wastewaters that carry the same characteristics of this study. RSM model used in the optimization of the production process caused increasing energy efficiency and made the process energy-efficient and sustainable by considering operating variables. The optimal conditions of the RSM-based model are the reaction time of 30.77 min, ozone dosage of 141.29 mg/l, pH of 7.2, and the GAC density of 1.29 gr/cm^3 with the forecasted removal percentage of 51.63%,62.84% and 56.13% for TDS, COD and, BOD respectively. In local condition experiments the COAT process showed an acceptable efficiency by the removal percentage of TDS=59.47%, COD=68.37%, and BOD=67%.

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