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Removal of COD From Landfill Leachate by Predication and Evaluation of Multiple Linear Regression (MLR) Model and Fenton Process

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MUNICIPAL Solid Waste (MSW) landfill leachate contains highly concentrated organic substances which are hazardous to the environment. Therefore, it must be treated before discharged into water bodies and suitable techniques are essential for effective treatment. In this regard, multiple linear regression (MLR) model has shown to be a favorable technique for optimization of landfill leachate treatment. In this study, four operational variables, H_2O_2:Fe^{2+} ratio, pH, reaction time, and Fe^{2+} concentration, were assessed using the model. The chemical oxygen demand (COD) removal was 96.43% by experiment, while the predicted assessment was 100% under the optimized settings; time = 33.87 min, concentration of Fe^{2+} = 749.64 mg/L, pH = 3, and ratio of H_2O_2:Fe^{2+} = 2 during Fenton treatment. The high value of the coefficient of determination, R^2 = 0.896, designates a resilient relationship between the experimental and model values. The residual study (residual plot) specified that the points were randomly distributed, confirming the appropriateness of the model. The MLR model established may possibly be used for assessment of other landfill leachate treatment.

Keywords: Leachate, Fenton, Multiple Linear Regression (MLR), Chemical Oxygen Demand (COD)

Introduction

Waste generation nowadays is rising in the world and is proving hard to prevent. Municipal Solid Waste (MSW) has been a major problem worldwide, especially in the fast growing cities and towns in the developing countries [1]. Landfill leachate comprises complex organic and inorganic compounds, high amounts of colour components, ammonia nitrogen and heavy metals [2]. Before leachate is discharged to the watercourse, it should be treated at on-site leachate treatment facility at a solid waste landfill. Each landfill treatment amenities are designed to fulfil the specific needs of the waste in landfill sites. This is because each landfill site is different in terms of characteristics of the waste disposed of and depends on the quality of leachate to be treated. Many factors determine the qualities and the quantities of the generated leachate e.g. moisture, available oxygen, temperature, hydrology, age, climate conditions, and waste stabilization. As a result, there are many types of leachate treatment such as the biological, physical, chemical and physio-chemical techniques [3,4]. The choice of the technology depends on characteristics of the leachate and discharge standards stipulated by the local authorities. Leachate is liquid formed from

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the percolation of rainwater. Leachate is generated due to waste decomposition together with rainwater percolation [5,6]. The toxic materials in the leachate may cause serious problems to both humans and the environment; thus, it is essential to monitor the concentration of certain parameters in leachate discharged from the landfill. Amongst the advanced oxidation processes, the Fenton process has been used extensively for leachate treatment. Studies have shown that this process has the capability to decrease organic contaminants and color in leachate treatment. The process has some advantages such as high organic matter removal, no toxic by-products, less energy requirement and simple experiment [7].

Multivariate statistics are being used widely to analyze data that arises from more than one outcome variable. This essentially models reality where each situation, product, or decision involves more than a single variable. In this regard, the multiple linear regression (MLR) is one example of the multivariate statistics used to examine the linear correlations between two or more independent variables and a single dependent variable [8,9]. The analysis is utilised to obtain data information, allow the extraction of useful information and improve data collection [10]. MLR model is used to demonstrate that the response variable varies with a set of independent variables. The variability that is exhibited by the response variable has two components; a systematic and random part. MLR equation is a weighted linear combination of the independent variables [11-14].Very few studies have explored the use of MLR model for landfill leachate treatment. Bhat et al. [15] developed equations for estimating biological oxygen demand (BOD) and chemical oxygen demand (COD) removal from landfill leachate. Kamyab et al. [16] reported that microalgae and macrophytes from agro-industrial waste were capable of removing nutrients only 4.4% of COD. The MLR model requires information such as the type of waste, temperature and precipitation rate for accurate estimation of the BOD and COD removal for any landfill. Brandstätter et al. [17] investigated MLR modeling for old landfills containing mixed waste. Wallace et al. [18] explored the use of multivariate statistical techniques such as PCA and PLS for the treatment landfill leachate. They have used eight parameters except for COD. Various research have been conducted to reuse the water treatment by using microorganism like microalgae in various substrates such as piggery, domestic, and mixed-kitchen wastes [19], textile industry [20] Agro-Industrial wastewater treatment and palm oil mill effluent (POME) by using macrophytes and microalgae [16, 21] As described by Abdel-Shafy et al. [22], membrane technology is rapidly developing new concepts for water and wastewater treatment. MBR leads to high hygienic standards of the treated effluent. Disposal of water treatment sludge by landfills can inhibit plant growth as its high organic and inorganic concentration can cause phosphorus fixation in the soils [23]. Due to these issues, studies have been conducted on treatment of landfill leachate by proposing an Artificial Neural Network in order to achieve zero discharge of sludge [24].

The above review shows that there is a lack of studies on the use of the MLR model for landfill leachate treatment. This study attempted to use MLR model to evaluate and optimize the oxidative performance of Fenton process for landfill leachate treatment. In our previous study [24], we have demonstrated the use of an artificial neural network (ANN) technique for landfill leachate treatment. In the current study, we have attempted to use the MLR model to compare the good quality predictions.

Materials and Methods

Fenton experiments

The leachate used in this study was taken from Jeram Landfill, Kuala Selangor, Malaysia. The leachate has the following characteristics; pH=7.5, COD=10,516 mg/L, total suspended solids (TSS)=810 mg/L and oil and grease (O & G) =9.5 mg/L. The experiments were performed by the means of a randomized experimental design in the laboratory. The variables investigated in this study were pH, Fe^{2+} concentration, reaction time, and the ratio of H_2O_2:Fe^{2+}. The experiments were conducted by using a jar-test glass reactor with 1L capacity. The pH of the leachate was adjusted to 3, 4.5, 6, 7.5, and 9 using 95%–97% H_2SO_4. Fenton reaction was performed at 500, 750, 1000, 1,250, and 1,500 mg/L by adding powdered ferrous sulphate ($FeSO_4.7H_2O$). The ratio of H_2O_2:Fe^{2+} was prepared at 2, 4, 6, 8, and 10. Rapid mixing was performed at 250 rpm for 80 s and then slow mixing at 50 rpm. The selected contact times were 5, 18.75, 32.5, 46.25 and 60 min. This phase was conducted to determine the effective variables and their proper range in this study. A full-factorial design effect plot for each of the three variables was prepared by using Design Expert software. A full-factorial approach was used in this study

[25]. The screening phases included four steps and 20 runs in each set of experiments (there were a total of 80 runs, designed by using 2^k, where k = number of variables).

Analytical methods

COD is the most broadly utilized indicator of leachate natural contamination. This parameter is observed consistently within the procedure of leachate treatment. It can be used to perceive the solid waste stabilization organize in landfills. Consideration of the conduct of COD indicator versus age of landfill by means of numerical models would help in predicting the future grade of leachate natural contamination and, consequently, the most productive method for working the leachate treatment facility. This is where advanced equations for anticipating the leachate's COD substance would be helpful. However, few research have been founded on information from a solitary landfill or from regionally explicit landfills. The few attempts to display leachate quality/attributes utilizing measurable techniques or programming have likewise centered around a single landfill or couple of area landfills. This investigation utilizes a complete lab-scale reactor plan that covers wide scopes of contact time, pH, H_2O_2:Fe^{2+} ratio and Fe^{2+} concentration [15,17]. Table 1 shows the preliminary full-factorial design in four steps. Effect plots were prepared in order to examine each variable and how it affects the response variables. The selection of full-factorial design was based on the ratio of H_2O_2: Fe^{2+} (2, 5, 6, and 10) as the variable factor. Several concentrations of Fe^{2+} were based on previous studies. Several runs were conducted at each concentration as follows:

- Step 1: 500 mg/L Fe^{2+}
- Step 2: 4,000 mg/L Fe^{2+}
- Step 3: 1,000 mg/L Fe^{2+}
- Step 4: 2,000 mg/L Fe^{2+}

Experimental Procedure

MLR is one of the traditional measurable devices used to depict quantitative relations between a dependent and an independent variable [26, 27]. Through univariate regression study, the relations between a dependent variable and an independent variable are examined, and the equation representing the linear relations between the dependent and independent factors is formulated. The regression models with one dependent variable and more than one independent variable are known as multivariate regression analysis [26]. The routine form of the linear regression model is:

Table 1. Preliminary full factor design for COD removal.

RUN	Factor A: pH	Factor B: Time (min)	Factor C: H_2O_2: Fe^{2+} Ratio
1	6	105	6
2	3	105	6
3	6	105	6
4	9	180	10
5	6	60	6
6	3	180	2
7	9	105	6
8	3	180	10
9	6	60	6
10	3	30	2
11	6	105	5
12	6	105	6
13	6	105	6
14	6	105	5
15	9	30	2
16	6	105	6
17	9	180	2
18	3	30	10
19	9	30	10
20	6	105	6

$$y_i = \beta_0 + \beta_1 x_{1i} + \beta_2 x_{2i} + \ldots + \beta_k x_{ki} + \varepsilon_i \; i = 1,2,\ldots N \quad (1)$$

Where y is the dependent variable, $x_1, x_2,\ldots, x_k$ is the independent or explanatory variables, i index the N sample observations, and ε is a random error term [28,29]. The regression formula were established by using Microsoft Excel version 10. Three mathematical relations were developed by using MLR analysis method. The ranges for independent variables were as follows: reaction time (x_1) of 5 min –60 min, Fe^{2+} concentration (x_2) of 500–1,500 mg/L, pH (x_3) of 3–9, and H_2O_2:Fe^{2+} ratio (x_4) of 2–10. The dependent variable (or objective function) was COD (Y_1) removal. R-squared value, p-value, and residuals were analysed when each multiple regression model was formed. The data analysis of the optimization process were conducted using Design-Expert Version 7.0. The adequacy of the model was checked using analysis of variance (ANOVA) and diagnostics nodes. Optimum leaching conditions for oxidative performance of Fenton process for landfill leachate treatment was gained by accomplishing mathematical optimization process coupled with the prestige method [30].

Results and Discussion

Model development for COD removal

Table 2 describes the experimental and predicted values of COD removal efficiency. The experimental variables x_1, x_2, x_3, and x_4 represent reaction time, the concentration of Fe^{2+}, pH of the leachate, and H_2O_2:Fe^{2+} ratio, respectively. The observed response (Y_1) represents the COD removal percentage. From the table, the highest COD reduction effectiveness was at run 40 (94.41%) and the lowest was at 26 (12.4%). As described by Ghanbari and Moradi [31], COD removal followed pseudo-second order kinetic model where the textile wastewater was treated by electro-Fenton and electrochemical Fenton, in which hydrogen peroxide was superficially implemented while an iron anode was used. Kang and Hwang [32] reported that the efficiency of hydrogen peroxide obtained from the removed COD values by oxidation in landfill leachate was observed to be about 45%.

The highest and lowest efficiency of 88.89% and 10.11%, respectively, were observed for the predicted values. Figure 1 illustrates the assessment of COD removal efficacy between experiment and prediction. Wang et al. [33] reported that only 42.4% of COD removal was obtained at the H_2O_2 dosage of 0.078 mol/L, corresponding to 0.5 of stoichiometric value theoretically required to completely remove COD from the leachate. Another report was indicated that COD removal was only 65% when hydrogen peroxide alone was applied to the electrolytic reactor, and the presence of ferrous ion greatly improved COD removal from landfill leachate [33]. An overall prediction error of -2.87% was observed in the MLR method. These results indicated that the COD removal efficiencies from the experiment were close to the predicted values for all the runs in the experiment. A relationship between the response and the variables were developed for the treatment system and the following equation describes the COD removal efficiency:

$$\text{COD Removal} = 0.715\times x1 - 0.039\times x2 - 10.89\times x3 - 1.77\times x4 + 141.39 \quad (2)$$

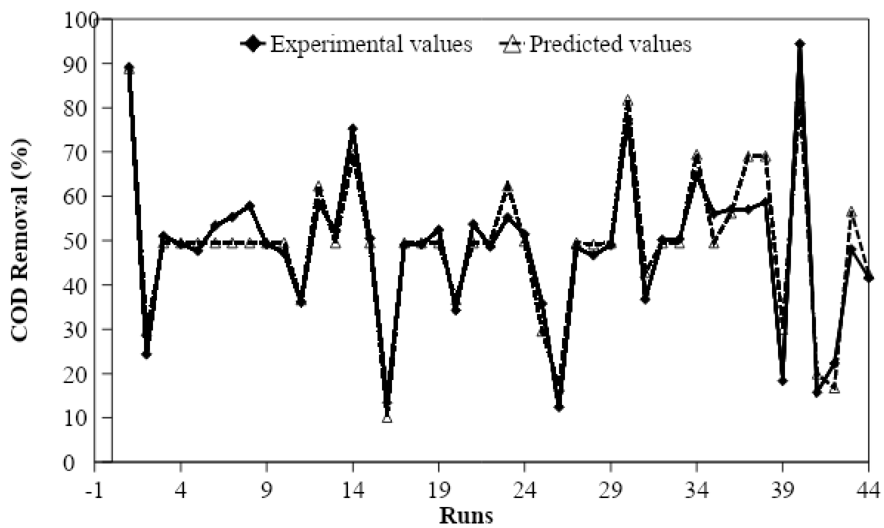


Fig. 1. Assesemment of COD removal efficiency between experiment and prediction.

TABLE 2. Data used in the regression modeling

Run	Time (min.)	Fe^{2+} Concentration (mg/L)	pH	H_2O_2: Fe^{2+} Ratio	COD Removal (%)	Predicted COD Removal (%)	Error (%)
1	46.25	750	4.5	4	89.16	88.894	0.298
2	46.25	1250	7.5	8	24.4	29.644	-21.492
3	32.5	1000	6	6	51	49.506	2.927
4	32.5	1000	6	6	49.2	49.506	-0.623
5	32.5	1000	6	6	47.7	49.506	-3.787
6	32.5	1000	6	6	53.4	49.506	7.290
7	32.5	1000	6	6	55.3	49.506	10.476
8	32.5	1000	6	6	57.81	49.506	14.363
9	32.5	1000	6	6	49.15	49.506	-0.725
10	32.5	1000	6	6	47.2	49.506	-4.887
11	18.75	750	7.5	4	36.2	36.699	-1.379
12	18.75	750	4.5	8	58.4	62.289	-6.659
13	32.5	1000	6	6	52.3	49.506	5.3408
14	46.25	1250	4.5	4	75.3	69.394	7.842
15	32.5	1000	6	6	50.5	49.506	1.966
16	18.75	1250	7.5	8	13.4	10.119	24.483
17	32.5	1000	6	6	49	49.506	-1.034
18	32.5	1000	6	6	49.13	49.506	-0.766
19	32.5	1000	6	6	52.4	49.506	5.521
20	46.25	1250	7.5	4	34.31	36.724	-7.036
21	32.5	1000	6	6	53.7	49.506	7.808
22	32.5	1000	6	6	48.67	49.506	-1.719
23	46.25	1250	4.5	8	55.17	62.314	-12.949
24	18.75	1250	4.5	4	51.4	49.869	2.978
25	18.75	750	7.5	8	35.8	29.619	17.264
26	18.75	1250	7.5	4	12.4	17.199	-38.703
27	32.5	1000	6	6	48.3	49.506	-2.498
28	46.25	750	7.5	8	46.7	49.144	-5.233
29	32.5	1000	6	6	49	49.506	-1.034
30	46.25	750	4.5	8	77.41	81.814	-5.689
31	18.75	1250	4.5	8	36.79	42.789	-16.306
32	32.5	1000	6	6	50.15	49.506	1.282
33	32.5	1000	6	6	50.36	49.506	1.694
34	18.75	750	4.5	4	64.6	69.369	-7.382
35	32.5	1000	6	6	56	49.506	11.595
36	46.25	750	7.5	4	57	56.224	1.360
37	32.5	500	6	6	57	69.006	-21.064
38	60	1000	6	6	58.6	69.031	-17.801
39	32.5	1500	6	6	18.4	30.006	-63.080
40	32.5	1000	3	6	94.41	82.176	12.957
41	5	1000	6	6	15.8	20	-26.582
42	32.5	1000	9	6	22.3	16.836	24.498
43	32.5	1000	6	2	48	56.586	-17.889
44	32.5	1000	6	10	41.6	42.426	-1.987

Where x_1, x_2, x_3, and x_4 are reaction time, Fe^{2+}concentration, pH, and H_2O_2:Fe^{2+} ratio, respectively, and Y is the response, *i.e.,* the predicted COD removal percentage. The empirical formula developed in this research may be utilised to quickly check the COD removal efficiency and obtain an initial educated guess about the percentage of COD removal when Fenton process is used. Wang et al. [33] elaborated that over 70% of COD removal was achieved under optimal conditions. This is because advanced Fenton reagent was coupled with electrochemical process to generate more hydroxyl radicals. Pai et al. [34] found the prediction accuracy of 48.22% for COD by employing artificial neural network (ANN) in the wastewater treatment plant.

Statistical analysis

For statistical analysis, the coefficient of determination (R^2) is an important criterion for model evaluation [35]. The R-squared coefficient represents the total variation of the predicted response and indicates the ratio of the sum of squares for regression (SSR) to the total sum of squares (SST). An R^2 value close 1 shows a satisfactory agreement of the linear model with the experimental data. In the present study, the R^2 value was 0.89. This number tells how much of the variance of output variable is explained by the variance of input variables. Ideally, this is at least 89%. Another important criterion for model evaluation is the adjusted R and in the current study, the value of adjusted R was 0.88. It shows the accuracy of the regression equation. The regression output values are given in Table 3.

Table 4 shows the ANOVA decomposition for the COD removal efficiency. The first row of Table 4 indicates the significance of the multiple regression models. The much larger mean square for the regression (2855.03) as compared to the residual error (33.827) indicates that the model is highly significant with zero probability of error. The F statistic for linear regression indicates that the statistical probability of the partial regression coefficients for a multiple linear regression is equal to zero. From ANOVA analysis, it can be seen that the F value of 1.15563E-18 is significant at $p < 0.05$, which signifies that the model is well determined by the factors.

The process of multiple linear modeling yielded the value of R^2 and the ANOVA test accepted the proposed linear modeling. To determine the significance of the coefficients, the p-value and t value are considered. The t-test is used to eliminate the least significant interaction variable. The regression coefficient values, standard error, and Prob. > F-value (probability) are given in Table 5. A larger t value together with a smaller p-value indicates that the parameter is of higher significance [36]. Results of the t-test ($p < 0.05$) showed the signs of the coefficients, confirming the existence of a linear relationship between the response and the explanatory variables.

TABLE 3. Regression statistics

Regression Statistics	
Multiple R	0.946
R Square	0.896
Adjusted R Square	0.885
Standard Error	5.816
Observations	44

TABLE 4. ANOVA decomposition for COD removal

ANOVA					
	df	***SS***	***MS***	***F***	***Significance F***
Regression	4	11420.126	2855.0316	84.399	$1.15563E^{-18}$
Residual	39	1319.269	33.827		
Total	43	12739.396			

Figure 2 shows the relation between the predicted and observed values. It can be seen that there is a strong correlation between the predicted and observed values from the coefficient of determination, $R^2 = 0.896$. The high value for the adjusted coefficient of determination (adjusted $R^2 = 0.885$) indicates that 88.5% of the total variation can be explained by the suggested model. In addition, the high value of the *F*-test (*F* model = 84.39) and the very low probability value (p-value $> F = 0.0001$) demonstrated the significance of this model and demonstrates the model's ability to predict COD removal efficiency.

Residual Plots

A residual plot is used to check whether the MLR is suitable for the data. Both variables, the residuals and the independent, are shown in the plot. A linear regression model is appropriate if the points are randomly dispersed. In the present study, the residual plots were examined to check the suitability of the model (Fig. 3). Figure 4 illustrates the residuals plot as a function of the projected values. Based on the results of the residual analysis, the points in the residual plot were randomly distributed around the horizontal axis. As a result, a linear regression model is appropriate for the data. As described by Larsen and McCleary [37], partial residual plot shows the extent and direction of linearity while displaying deviations from linearity, such as outliers, inhomogeneity of variance, and curvilinear relationships.

Normal probability plot

A normal probability plot is used to check the normality of error and shows whether data is approximately normally distributed. A plot that is nearly linear specifies normality is satisfied. A plot which departs substantially from linearity indicates that the error distribution is not normal. The normal probability plot after the transformation is presented in Fig. 5. It was observed that the normal probability plot was a straight line. Based on MATLAB, the most accurate estimations of the artificial neural networks were obtained with log sigmoid transfer function [38].

TABLE 5. The regression coefficient values.

	Coefficients	Standard Error	t-Stat	P-value	Lower 95%	Upper 95%
Intercept	141.3	8.150	17.34	$6.2E^{-20}$	124.9	157.87
Time	0.715	0.086	8.284	$3.96E^{-10}$	0.54	0.88
Fe^{2+} concentration	-0.039	0.004	-8.398	$2.81E^{-10}$	-0.049	-0.03
pH	-10.89	0.791	-13.76	$1.48E^{-16}$	-12.49	-9.29
H_2O_2: Fe^{2+}ratio	-1.77	0.593	-2.98	0.0048	-2.97	-0.57

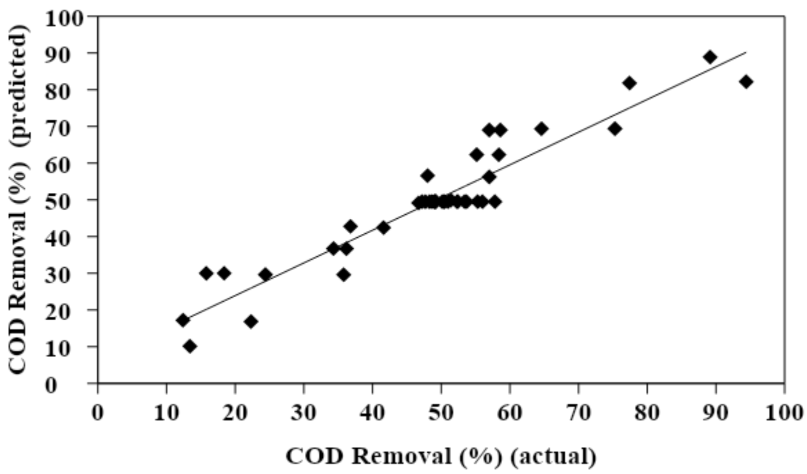
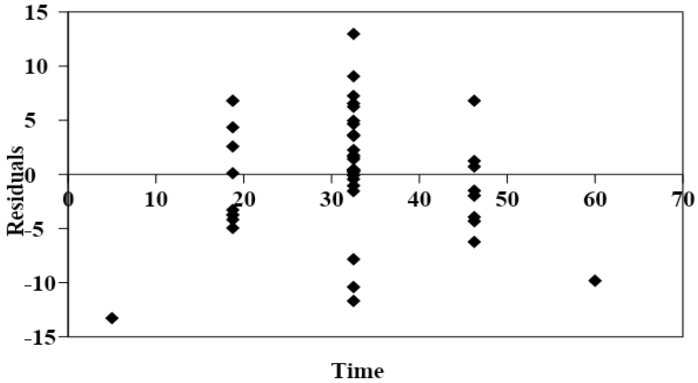
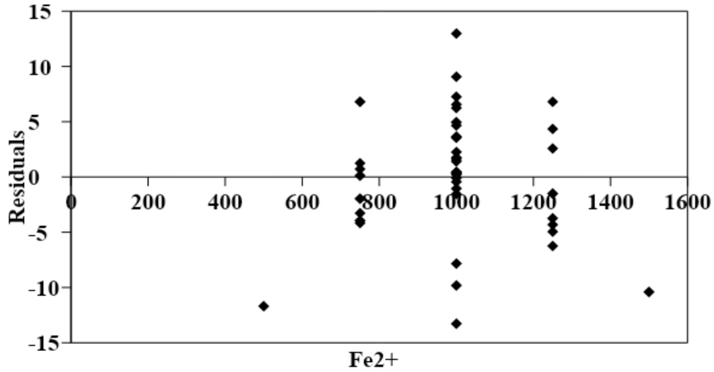


Fig. 2. Relationship between empirical and predicted values.



(a)



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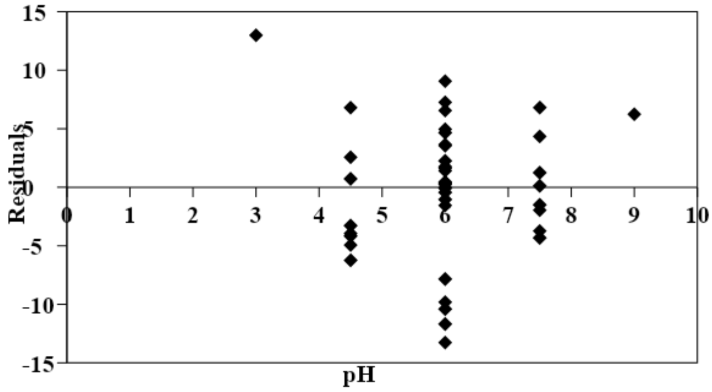

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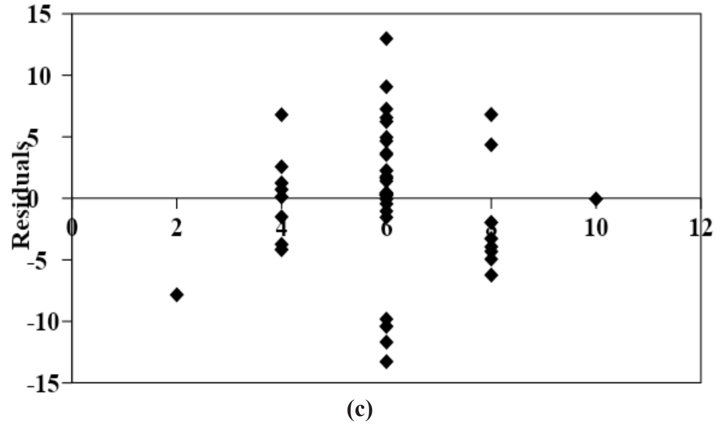

(c)

Fig. 3. Partial residual plots for a) Time, b) Fe2+ concentration, c) pH and d) H2O2:Fe2+ Ratio.

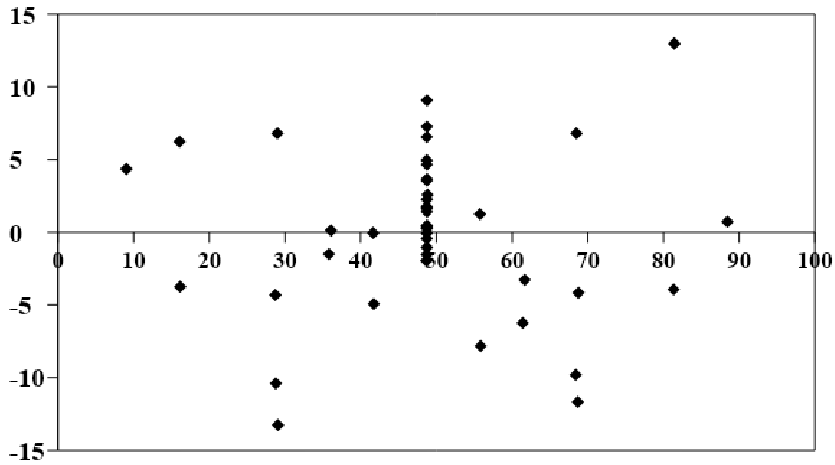

Fig.4. Residuals vs. predicted values for linear fit.

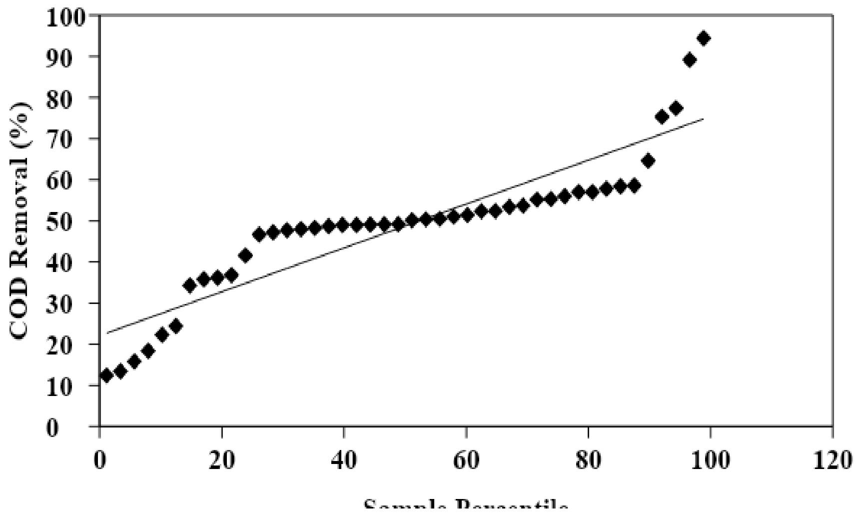

Fig. 5. Normal probability plot of residual when fitting by a line.

Response optimisation and validation of the experimental model

The response optimisation of COD removal effectiveness by Fenton process was evaluated in this part. From the results, the optimised conditions for the highest COD removal efficiency was observed at pH= 3, H_2O_2: Fe^{2+} ratio= 2, Fe^{2+} concentration = 749.64 mg/L, and reaction time= 33.87 min. During this period, a COD removal efficiency of 100% was predicted. Then, a further experiment was performed at optimum conditions to verify the model and experiment. Results showed a value of 96.43% for the experiment, which was very similar to the value predicted by the model (100%). A previous report by Zhang et al. [39] showed COD removal from landfill leachate was only 65% when hydrogen peroxide alone was applied, with the presence of ferrous ion greatly improving COD removal. Singa et al. [40] observed that the maximum COD removal from landfill leachate by using photo Fenton was 68% under optimum operating conditions. Also, Singa et al. [41] found that under favorable experimental conditions, maximum COD removal from landfill leachate by using Fenton process was 56.49%. Sruthi et al. [42] indicated that heterogeneous Fenton process was capable of removing 88.6% COD from landfill leachate at the optimal conditions.

Conclusion and Future Work

MLR model can be utilised to predict and optimise landfill leachate treatment and it is a useful tool that provides a practical complement to the knowledge gained from experimental and theoretical studies. The MLR model used in this study validated significant effects of four operating variables (contact time, pH, H_2O_2:Fe^{2+} ratio and Fe^{2+} concentration). The optimum COD removal efficiency of 94.4% was experimentally shown at optimum values of contact time = 33.87 min, pH = 3, H_2O_2:Fe^{2+} ratio = 2, and Fe^{2+} concentration = 749.64 mg/L. The R^2= 0.896 indicates a strong correlation between the predicted and observed values. From ANOVA analysis, the F value for COD removal was 1.15563E-18 and the results were significant at $p < 0.05$. However, economical related aspect was not taken into consideration in this study. Therefore, cost-benefit analysis of such process should be considered in the future studies. In addition, planned future work and improvements include performance in a real plant and the classical steps of model identification and validation approach (testing robustness with a new dataset).

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