Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2023.29660

299 (2023) 62–73
July

Robust statistical optimization study for pollutant degradation in landfill leachate using bio-coagulant

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Received 2 November 2022; Accepted 27 May 2023

ABSTRACT

Leachate contains huge amount of organic and inorganic substances which leads to environmental contamination. Coagulation study was performed with Taguchi experimental design using *Musa acuminata* Colla leaves (MACL) as a bio-coagulant for the removal of pollutants from leachate wastewater. Orthogonal arrays L_{16} plan and larger the better formula was employed for this optimization study. Scanning electron microscopy and Fourier-transform infrared spectroscopy studies were carried out to identify the changes in surface morphology, presence of functional groups and interaction effect between of *Musa acuminata* Colla leaves (MACL) coagulant and pollutants in leachate wastewater before and after the treatment. It was investigated how well *Musa acuminata* Colla leaves coagulant (MACLC) could remove color, chemical oxygen demand (COD), total suspended solids (TSS), Cr, Cd, and Cu from leachate wastewater. The pH, coagulant dosage, contact time and agitation speed were used as control variable and color, COD, TSS, Cr, Cd, and Cu were considered as responses. Optimum results were obtained at level 3 (pH = 6), level 2 (dosage = 1 g/L), level 4 (time = 60 min), and level 3 (agitation speed = 90 rpm), the maximum removal was occurred at the respective levels were color (91%), COD (77%), total suspended solids (83%), and heavy metals removal as Cr (89%), Cd (78%), and Cu (87%). Present study reveals that MACLC is a promising coagulant for treatment of leachate as an alternate to the synthetic coagulant and Taguchi experimental design is an efficient and cost-effective optimization tool for the treatment of landfill leachate.

Keywords: Bio-coagulant; Leachate; Taguchi experimental design; *Musa acuminata* Colla leaves

1. Introduction

Land filling is a simple and economical waste management method but nearly 95% of solid waste is dumped into landfills without any segregation of waste [1]. Occurring of physico-chemical and biological reactions in municipal solid waste results in the generation of leachate [2,3]. 0.2 m^3 of leachate generated an average of every one ton of municipal solid waste [4]. Leachate is a complex characteristics of ammonia–nitrogen (NH_3–N), heavy metals, chlorinated organic, and inorganic salts, dark brown color, chemical oxygen demand (COD), biochemical oxygen demand (BOD), and bad odours [5,6]. More than 200 organic compounds found in leachate including aromatic hydrocarbons, cyclic, bicyclic, alcohols, and ethers compounds like benzene, cyclohexenes, tetramethyltetralins, methanol, and butanol [7]. The concentration of the contaminants and its volumetric flow varies in every dumpsite because it depends on the composition of the wastes, waste age, geological location, moisture content, land filling method and also geological

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factors like rainfall, sunlight, and surface runoff coefficient [8,9]. Physical/chemical and biological treatment methods, aerated lagoons, activated sludge, anaerobic lagoons, reactors, air stripping, pH adjustment, chemical precipitation, oxidation, reduction, and coagulation methods were normally adopted for the treatment of leachate [8,9]. According to the complexity of the wastewater and the treatment processes, leachate will be treated using an integrated system. The ecological system is negatively impacted when contaminants in leachate percolate into the subsoil or a nearby water course, and the concentration of various minerals in groundwater increases over the permitted level. Proper treatment procedures and disposal techniques are necessary for this particularly difficult landfill wastewater in order to prevent surface water pollution, other environmental difficulties, human health risks, and comply with solid waste management rules and regulations.

Several treatment approaches were established for the treatment of landfill leachate. Coagulation–flocculation is the most economical, trustworthy, and effective treatment method available, and it is frequently used as a pretreatment process for leachate [10–12]. However, biological processes are ineffective in treating landfill leachate, which has a BOD_5 to COD ratio of less than 0.5 [13,14]. Heavy metals in leachate around 70%–90% are inorganic compounds or colloidal nature [15]. By adsorption, ion exchange, or surface precipitation, these heavy metals are bond to colloids [16]. Dark color in leachate is caused by colloidal particles smaller than 10 m in size, whereas turbidity in leachate is caused by bigger colloidal particles (3.5–6 m) in size [17]. Three different types of coagulants, including polymers, hydrolyzing metallic salts, and pre-hydrolyzed/pre-polymerized metallic salts, can be used in the coagulation process. The polymers can be natural or manufactured [18] but numerous studies have shown that both chemical and natural coagulants performed admirably well at removing heavy metals and non-biodegradable organic components from landfill leachate. Ferric chloride, polyaluminum chloride (PAC), aluminum sulfate (alum), and ferrous sulfate are widely applied chemical coagulants for leachate treatment [19]. Coagulant extracted or prepared from the naturally available materials such plant parts, animals skins, and easily biodegradable trash may be favorable for human well beings when compared with chemical coagulants [20]. Natural coagulants easily form larger flocs and it settle down very fast and also not harmful, easily biodegradable in nature [21]. The following natural coagulants were employed for the water treatment process because widely accessible, inexpensive, versatile, and environmentally acceptable: *Moringa oleifera*, nirmali seeds, tannin, chitosan, cactus, *Calotropis procera* bentonite dirt, *Strychnos potatorum* seeds [22].

The following natural coagulants were successfully employed for leachate treatment, *Hibiscus rosa-sinensis* [23,24], sago trunk starch [25], psyllium husk [26], oil palm trunk starch [27], *Artocarpus heterophyllus* seeds [28], cross-linked oil palm trunk starch, tobacco leaf [25], pine bark and chitosan [29], *Nephelium lappaceum* seed [30], barley [31], tannin-based natural coagulant [32], *Moringa oleifera* Lam, and *Abelmoschus esculentus* (L.) Moench (Okra) [33], lateritic soil [34], *Opuntia ficus* mucilage [35]. For the treatment of water and wastewater, various banana tree portions have been used as coagulants and adsorbents. Banana stem juice showed removal of 80.1% COD, 88.6% SS, and 98.5% turbidity at a pH 7 for spent coolant wastewater [36]. *Acacia mearnsii*, banana leaves, cactus, *Carica papaya*, *Cassia alata*, *Calotropis procera*, hyacinth bean, *Jatropha curcas*, and tamarind seeds were employed for the treatment of water among this nine coagulant banana leaves showed efficient turbidity removal percentage [37]. 1.0 g in 100 mL of banana stem adsorbent can remove 91.12% of the color from textile wastewater [38]; similarly, mercaptan cellulose made from banana leaf shows significant potential for sorbing arsenic [39]. Banana peels adsorbent exhibited 96% of chromium Cr(VI) removal under varied pH [40]. Using banana peels as a bio-flocculent can reduce 93.44% of the turbidity from synthetic turbid water at pH 12 [41]. Banana blossom floret and peel adsorbents removed 81.1% and 77.7% of the manganese(Mn) from groundwater, respectively [42].

Taguchi method is widely used for the optimization of the wastewater treatment design parameters owing to the fact that it gives the well-balanced minimum number of experimental designs to analysis the multiple factors by orthogonal arrays (OA) [43]. This Robust experimental design was combined using statistical and mathematical methods. The planning, conducting, analysis, and validation phases make up this design. Each step has a particular goal and is connected to the ones before it to complete the total optimization process [44]. Based on the desired performance responses, the signal-to-noise ratio is a measured. The nominal-the-better, the smaller-the-better, and the larger-the-better are the three sorts of features that make up the signal-to-noise (S/N) ratio [44]. Taguchi design is simple to use, economical and requires fewer trails [45]. In a very small number of research investigations, Taguchi optimization study was used to treat landfill leachate. These studies focused on electrocoagulation, microwave oxidation, advanced oxidation, ultrasonic activated persulfate, and catalytic ozonation. In the current study, the Taguchi statistical optimization method was utilized to remove color, COD, total suspended solids (TSS), Cr, Cd, and Cu from landfill leachate using *Musa acuminata* Colla leaves (MACL) as a natural coagulant. The orthogonal array L_{16} plan was used to investigate the optimal pH, coagulant dose, contact time, and agitation speed for the removal of color, COD, TSS, chromium, copper, and cadmium. Each control parameter (pH, coagulant dosage, contact time, and agitation speed) was examined for its influence on each response using the S/N ratio, analysis of variance (ANOVA), two-dimensional contour maps, and probability plots (color, COD, suspended solids, chromium, cadmium, and copper).

2. Material and methods

2.1. Materials

2.1.1. Site description, collection and characterization study of leachate wastewater

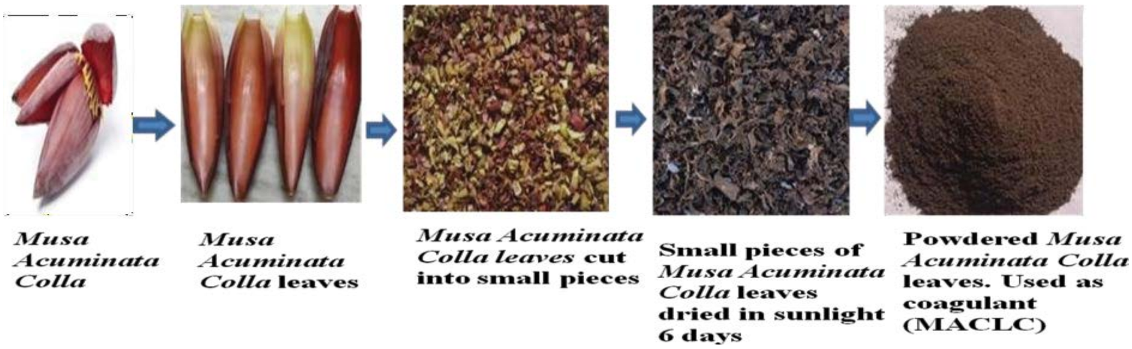
leachate samples were taken from the sanitary landfill at Dindigul, Tamil Nadu, India. The collected leachate samples were stored in plastic bottles in the lab at 4°C and used for further research to avoid any chemical and biological interactions. The collected samples were subjected to physico-chemical characterization analyses both before and after treatment process using APHA Standard Methods (American Public Health Association, 2012). Analytical graded chemicals were utilized in this study. The heavy metals identified in the leachate were examined using atomic absorption spectroscopy (Model Name – Shimadzu Atomic Absorption Spectrophotometer AA-6300; ROM version-1.03). Leachate's turbidity was measured with a portable turbidity meter (Model: 331, Make: Deep Vision), and the results were reported in nephelometric turbidity units (NTU). Removal of color was identified by UV-Visible Spectrophotometer (Model: R6000, Make: Hach Co., USA), COD digester (Make: Hach Co., USA) is used to analyze the COD which is coupled with UV Spectrophotometer (Model: DR6000, Make: Hach Co., USA). The leachate samples were taken at various time intervals from the same point source. In each experiment, physico-chemical characteristics were assessed three times and the average results were shown in Table 1 along with a comparison to the CPCB standards. Leachate had a pH that was alkaline and within the CPCB limit. The BOD and COD readings exceeded the CPCB's recommended limits so it is possible that there are more organic compounds present in the landfill leachate that was collected, and the unpleasant scent of the sample that was taken also suggests there are more organic compounds present. The presence of inorganic mineralization and high levels of dissolved organic salts may be caused by the high total dissolved solids value. The physico-chemical characteristics of the receiving water bodies are changed, and the toxicity is increased. The BOD_5/COD ratio 0.22 reveals that the collected leachate sample is not easily biodegradable. The amount of heavy metal in the leachate sample was under the prescribed limit, but it was also a secondary factor to be taken into consideration.

2.1.2. Preparation of *Musa acuminata* Colla leaves coagulant

Fresh *Musa acuminata* Colla leaves were acquired from local vegetable markets at Dindigul, Tamil Nadu. The preparation process of *Musa acuminata* Colla leaves coagulant (MACLC) is shown in Fig. 1. Manual removal of *Musa acuminata* Colla leaves from blossoms followed by cutting into numerous little pieces. To get rid of the dirt, dust and other pollutants, small pieces of *Musa acuminata* Colla leaves were washed with distilled water about five times. The *Musa acuminata* Colla leaves were cut into small pieces and dried in the sun for 6-d before being crushed in a mixer grinder. *Musa acuminata* Colla leaves were powdered and passed through a sieve size of 75 μ. The distilled water and powdered MACLC were mixed for about 10 min at room

Table 1
Physico-chemical characteristics of landfill leachate

Parameters	Values	CPCB standards for disposal (India)
pH	8.5	5.5–9.5
Color	Dark brown color	Unobjectionable
Odour	Foul smell	Unobjectionable
BOD_5 (mg·O_2/L)	562	10
Turbidity (NTU)	960	–
COD (mg·O_2/L)	2,521	250
BOD_5/COD	0.22	–
Total solids (mg/L)	5,513	–
Total dissolved solids (mg/L)	4,327	2,100
Total suspended solids (mg/L)	1,214	20
Cr (mg/L)	1.198	2
Cd (mg/L)	0.2	2
Cu (mg/L)	0.77	3



Musa Acuminata Colla

***Musa Acuminata Colla* leaves**

***Musa Acuminata Colla leaves* cut into small pieces**

Small pieces of *Musa Acuminata Colla* leaves dried in sunlight 6 days

Powdered *Musa Acuminata Colla* leaves. Used as coagulant (MACLC)

Fig. 1. Preparation of coagulant from *Musa acuminata* Colla leaves.

temperature before being filtered through muslin cloth because distilled water is used as a solvent to extract very active coagulation elements and avoid unnecessary organic elements [46]. Drying the wet coagulant (MACLC) in an oven for 48 h at 100°C was followed by storing it in airtight metal containers. Pollutant removal from landfill leachate wastewater was accomplished using this MACLC. Fourier-transform infrared spectroscopy (FTIR) and scanning electron microscopy (SEM). Characterization studies were accomplished for MACLC before and after the treatment to understand the surface morphology and any variations in the functional group of this natural coagulant.

2.2. Coagulation–flocculation experimental procedure

A coagulation–flocculation investigation was carried out using a jar test apparatus (Enkay Enterprises, Model: KEJ16, India) attached with revolving six paddles (stainless steel). 1,000 mL beakers each held a sample of 500 mL leachate. In order to determine and set the ranges of each control factor, a one-time classical method experiment was done using 500 mL of leachate samples placed into six 1,000 mL beakers at room temperature. The pH of leachate samples were varied from 2 to 11, the coagulation dosages were adjusted from 0.5 to 2.5 g/L, contact duration was modified from 10 to 90 min, and the quick mixing speed was varied as 30, 60, 90, and 120 rpm for leachate samples. NaOH or HCl solutions were used for the pH alteration. Each sample were allowed to settle for 90 min, after 90 min, the supernatant samples were filtered by Whatman Filter Paper No. 40 and pollutant removal efficiency of color, COD, TSS, and heavy metals (Cr, Cd, and Cu) were tested at room temperature. The Taguchi robust statistical experimental design was used to validate the experimental findings, and the maximum pollutant removal efficiency was determined. After the one-time classical method experimental investigation, control factor levels were fixed for pH 2, 4, 6, 8; coagulant dose of 0.5, 1.0, 1.5, and 2 g/L; contact time of 30, 40, 50, and 60 min, and agitation speed of 30, 60, 90, and 120 rpm. Leachate sample was utilized for further research after being diluted with 30% distilled water. The removal efficiency of color, COD, TSS, Cr, Cd, and Cu were calculated by Eq. (1):

$$\% \text{ of Removal}\left(R\right)=\frac{\left(P_i-P_f\right)}{P_i}\times 100 \tag{1}$$

where P_i – initial concentration of color, COD, TSS, and heavy metals (Cr, Cd, and Cu) in leachate; P_f – final concentration of color, COD, suspended solids, and heavy metals (Cr, Cd, and Cu) in leachate; R – removal efficiency in percentage (%).

2.3. Taguchi optimization experimental design and analysis

The pollutant removal efficiency of the MACLC in the leachate was validated using the Taguchi statistical experimental method. Taguchi statistical experimental design is a cost-effective technique since it may quickly achieve 70%–90% of the necessary parameter optimization. L_{16} orthogonal arrays with 4 columns and 4 rows were used in this statistical design to analyze the optimization parameters for the coagulation process. MINITAB (Version 21.1.0) analytical software was used to examine the data. For this investigation, pH, coagulant dosage, contact time, and agitation speed were chosen as the optimization parameters. Process parameter for every level was quantified with S/N ratio. Higher value of S/N ratio represents the higher quality characteristic values. The S/N ratio minimizes the impact of unwanted or noisy effects on process variables [33,47]. Performance characteristics for optimization study were analyzed with three categories. In this present study, "larger-the-better" is used to evaluate the removal efficiency of COD, color, TSS, Cr, Cd, and Cu using Eq. (2).

The larger-the-better S/N ratio formula for the using base 10 log is:

$$\frac{S}{N}=-10\log\left(\frac{\sum\left(1/Y^2\right)}{n}\right) \tag{2}$$

where Y = responses for the given factor level combination and n = number of responses in the factor level combination.

The Taguchi optimization method provides an economical experimental design matrix while reducing the complexity of the traditional one-time classical method. The factors and their level in the orthogonal array L_{16} are displayed in Table 2. Orthogonal arrays make it easier to analyze the impact of different process parameters effectively [48]. 16 experimental runs and 4 parameter levels make up an orthogonal array. The interaction between the independent and dependent variables was made clear by the ANOVA. The effect of mean square variance, adjusted sums of squares, adjusted mean squares, F-test of significance, probability value, total degrees of freedom, S value, and R^2 value were all examined. The number of observations in the experiments can be used to determine the total DF value. The variation among the different components like variables and responses can be identified through the adjusted mean squares measures (Adj. MS). The model's design efficiency is proven by the F and p values.

3. Results and discussion

3.1. SEM and FTIR studies of MACLC

The surface morphology of the MACLC was examined using SEM to determine the differences between the two states of the coagulation–flocculation process. The good interaction effect between the MACLC and pollutants in leachate is clearly observed by the SEM image of before and

Table 2
Orthogonal array operating factors and levels

Factors	Level 1	Level 2	Level 3	Level 4
pH	2	4	6	8
Dosage (g/L)	0.5	1	1.5	2
Time (min)	30	40	50	60
Agitation speed (rpm)	30	60	90	120

after the coagulation process through the high changes in MACLC surface. The surface structure of MACLC was analyzed in 2 μm. Fig. 2a and b illustrate it. Large size porous structures of MACLC were filled by the pollutants of leachate wastewater after the process. The results of an FTIR study were used to analyze the presence of functional groups in the treated and untreated natural coagulant MACLC, as shown in Fig. 2c. The peak values at 3,400–3,500 cm^{-1} frequency indicated the presence of hydroxyl groups [42,49,50]. Before and after the treatment, there was a peak of strong O–H stretching (3,434.76 and 3,447.08 cm^{-1}). The presence of carboxylic/carbonyl functional groups is indicated by the peak values between 1,400 and 1,700 cm^{-1}. The carboxylic/carbonyl functional groups (C=O) were found at the peak values of 1,642.98; 1,644.2; 1,561.33; 1,450.66 and 1,419.08 cm^{-1}. Following the coagulation procedure, aromatic (C=C) molecules were also found in the peak values of 1,561.33 cm^{-1} this could be due to the binding of benzene groups. Leachate showed an aromatic compound content of 1.21 mg/L, and C–OH stretching frequency ranged among 1,250–1,300 cm^{-1}. After the coagulation process, 1,269.02 cm^{-1} was detected, which could be the result of acidic groups binding with the coagulant of leachate [49]. The C–H group bonded to the amino group may be responsible for the peak value of 2,854.04 cm^{-1} [51].

3.2. Taguchi experimental optimization design for coagulation–flocculation process

In order to investigate the potential of MACLC for the removal of color, COD, TSS, Cr, Cd, and Cu in the coagulation and flocculation process, an OA L_{16} design matrix was developed with 16 runs of experiment. The efficiency of MACLC's pollutant removal was evaluated using analytical software Minitab (Version 21.1.0). The list of independent variables and the choices made for this research work are explained in Table 3. Experimental design variables, responses, their levels and results of removal efficiency of color, COD, TSS, Cr, Cd, and Cu are furnished in Table 4. All the studies were conducted with three times of repetition and average outcomes were recorded. All experiments were carried out at room temperature and pollutant removal effectiveness was given as a percentage of removal.

3.3. Signal-to-noise ratio

The significance of each independent variable (pH, dosage, contact period, and agitation speed) on the elimination of each of the six distinct responses (color, COD, TSS, Cr, Cd, and Cu) is explained by the S/N ratio. The study selects a larger-is-better, therefore the peaks of all the plots were

Table 3
Independent variables and responses

S. No.	Independent variables	Responses
1.	pH (*A*)	Color removal
2.	Dosage concentration (*B*)	COD removal
3.	Contact time (*C*)	TSS removal
4.	Agitation speed (*D*)	Heavy metals removal (Cr, Cd, Cu)

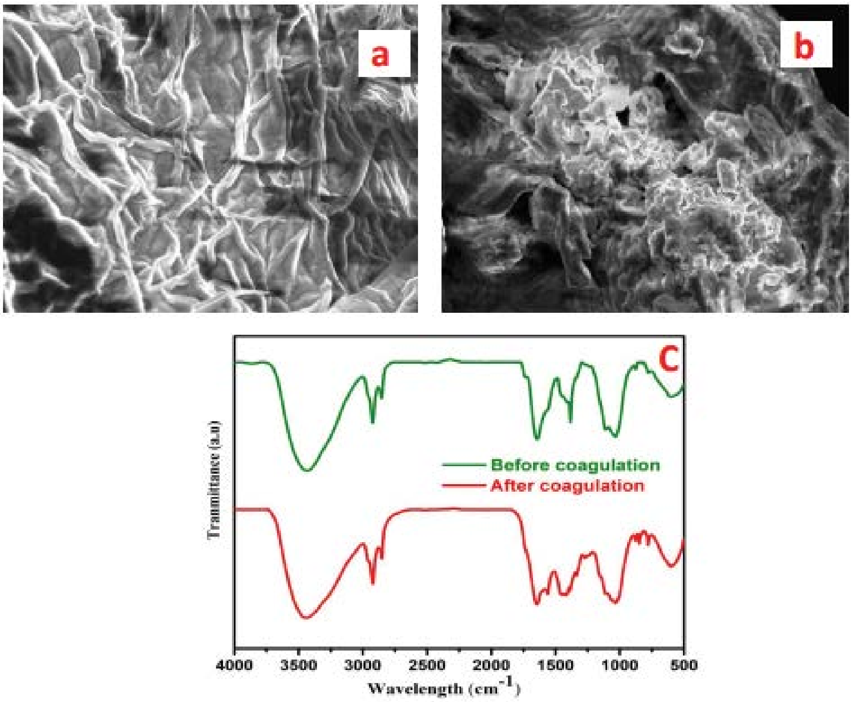


Fig. 2. Characterization study of *Musa acuminata* Colla leaves before and after the treatment (a,b) scanning electron microscopy and (c) Fourier-transform infrared spectroscopy.

Table 4
Results of experiments corresponding to L_{16} experimental plan

Run	pH (level)	Dosage (level) (g/L)	Time (level) (min)	Agitation speed (rpm)	Color (%)	COD (%)	TSS (%)	Cr (%)	Cd (%)	Cu (%)
1	1	1	1	1	15	7	12	26	24	29
2	1	2	2	2	21	10	19	28	26	31
3	1	3	3	3	17	9	15	33	29	34
4	1	4	4	4	16	6	11	35	30	36
5	2	1	2	3	42	35	35	47	47	64
6	2	2	1	4	54	39	43	49	51	67
7	2	3	4	1	49	36	41	53	55	69
8	2	4	3	2	39	33	39	57	61	74
9	3	1	3	4	78	72	76	86	73	77
10	3	2	4	3	91	77	83	89	78	87
11	3	3	1	2	84	71	84	87	78	87
12	3	4	2	1	75	70	74	85	77	87
13	4	1	4	2	65	58	61	84	71	77
14	4	2	3	1	68	64	67	85	73	79
15	4	3	2	4	63	62	65	85	74	80
16	4	4	1	3	61	59	60	87	76	80

recorded. The high S/N ratio values showed the low losses in contaminant removal. Fig. 3a–f displays the S/N graph for each control factor and response. The higher number of delta values represented a more significant factor. Less S/N ratio findings imply the coagulation process had little impact on the effectiveness of pollutant removal and little interaction with other control parameters [52]. Plotting the S/N ratio curve in accordance with the delta values acquired for each run level will result in the optimum effect for each control factor and response. As seen in Fig. 3a–f, pH significantly influenced the effectiveness of all contaminant removal when compared to other control parameters. It was also demonstrated that pH is a significantly influencing factor by the larger delta values for pH that were obtained for all of the responses, including 13.54 (color), 19.31 (COD), 15.09 (TSS), 9.27 (Cr), 9.00 (Cd), and 8.32 (Cu), when compared to the other control factors. Maximum signal to noise ratio was attained for the control factor pH. Based on maximum S/N ratio it was observed that 91% of color, 77% of COD, 83% of TSS, 89% of Cr, 78% of Cd and 87% of Cu removal was obtained at the optimum conditions of level 3 (pH = 6), dosage at level 2 (1 g/L), contact time at level 4 (60 min), and agitation speed level 3 (90 rpm).

3.4. ANOVA analysis

The error variance and multi response properties are described using analysis of variation. The correlation between expected values, experimental findings, and the effectiveness of the design model is provided by the ANOVA result. ANOVA allows one to note the percentage contribution of each factor [53,54]. As seen in Table 5, this model is significant because the p values for color, COD, TSS, and all three heavy metals (Cr, Cd, and Cu) are less than 0.05 and the *F* values are greater than the *p*-values. When the value of *F* exceeds the value of *F* in the table, the null hypothesis is rejected [55]. Table 5 displayed that there were 15 degree of freedom values in total, which was more than each degree of freedom values. The higher percentage of contribution from the control factors indicates that they have a significant impact on the responses. The high percentage impact of pH made to each response in this experimental design is listed in Table 6. For all of the responses, the percent confidence level was greater than 90, as shown in Table 5. The study's lowest *S*-values support the design's best fit and the model's improved responsiveness, as shown in Table 5. Observed critical f value was 3.39. In this design, every *F*-value was higher than the crucial *F*-value. Design was therefore significant. Cr, Cd, and Cu had R^2 values of 99.92, 99.98, and 99.80, respectively, and adjacent Cr, Cd, and Cu had R^2 values of 99.60, 99.90, and 99.00, respectively. This demonstrated that responses and control variables had a strong correlation Table 6. R^2 and adjacent R^2 values also more than 99.5%, it confirms the adequacy of design model. For all heavy metal removal, the percentage of predicted values and confidence limit values were greater than 90. The operational parameters for the elimination of color, COD, TSS, Cr, Cd, and Cu were chosen in accordance with the optimization design, and they were dosage level 2 (1 g/L), contact time level 4 (60 min), and agitation speed level 3 (90 rpm). Table 7 shows the very less deviation between observed and predicted values; it confirms the excellent adequacy of Taguchi statistical experimental optimization design for treatment of landfill leachate.

3.5. 2D contour plot and probability plot

Interaction effect and removal efficiency of each single response (color, COD, TSS, Cr, Cd, and Cu) was analyzed with all the four control factors using 2D contour plot and probability plot. Two independent factors can be marked in the x and y axes of this 2D contour plot to analyze their

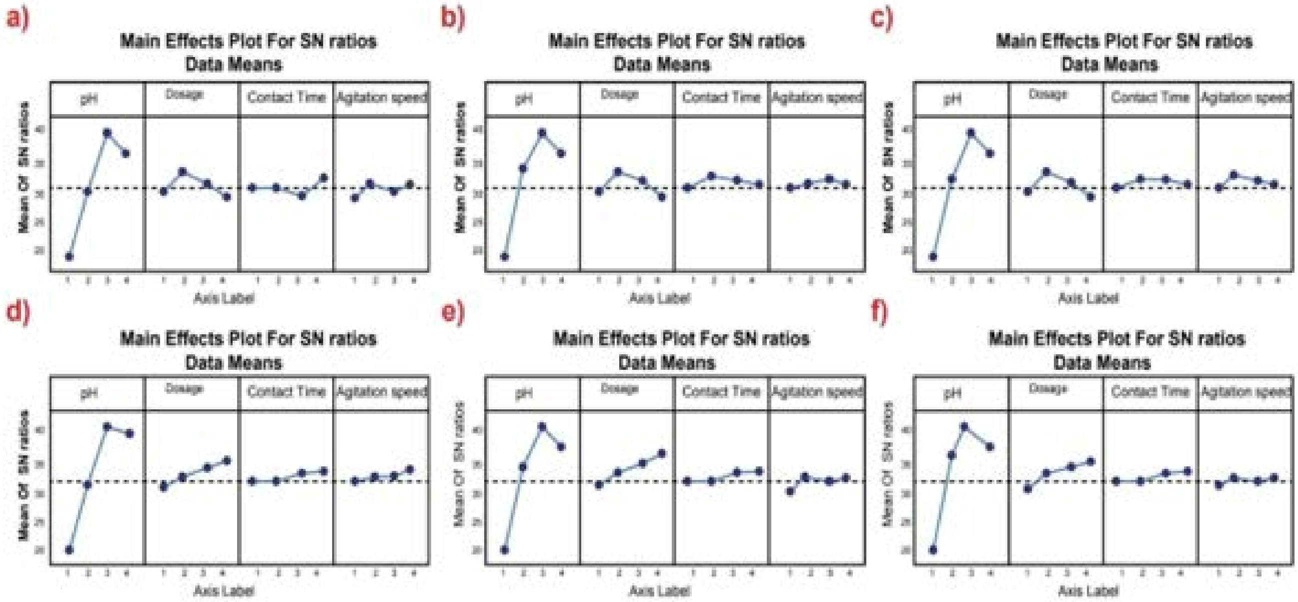


Fig. 3. Mean effect curve for S/N ratio for (a) color, (b) chemical oxygen demand, (c) total suspended solids, (d) Cr, (e) Cd, and (f) Cu.

Table 5
Analysis of variance for color removal, COD, total suspended solids

Source	DF	Seq. SS	Adj. SS	Adj. MS	*F*	*p*
Analysis of variance for S/N ratios of colour removal						
pH	3	426.057	426.057	142.019	400.09	0.000
Dosage	3	8.749	8.749	2.916	8.2	0.015
Time	3	0.642	0.642	0.214	4.60	0.437
Residual error	6	2.130	2.130	0.355		
Total	15	437.578				
Analysis of variance for S/N ratios of COD						
pH	3	935.506	935.506	311.835	460.71	0.000
Dosage	3	7.704	7.704	2.568	3.79	0.077
Time	3	2.831	2.831	0.944	4.39	0.333
Residual error	6	4.061	4.061	0.677		
Total	15	950.102				
Analysis of variance for S/N ratios of total suspended solids						
pH	3	545.799	545.799	181.933	207.25	0.000
Dosage	3	9.459	9.459	3.153	3.59	0.086
Time	3	1.701	1.701	0.567	4.65	0.413
Residual error	6	5.267	5.267	0.878		
Total	15	562.227				

Table 6
Analysis of variance for heavy metals (Cr, Cd and Cu)

Source	DF	Seq. SS	Adj. SS	Adj. MS	*F*	*p*
Analysis of variance for S/N ratios of Cr						
pH	3	238.724	238.724	79.5745	834.96	0.000
Dosage	3	4.135	4.135	1.3783	14.46	0.004
Time	3	1.773	1.773	0.5909	6.20	0.029
Residual error	6	0.572	0.572	0.0953		
Total	15	245.203				
Analysis of variance for S/N ratios of Cd						
pH	3	207.833	207.833	69.2777	1,303.2	0.000
Dosage	3	3.877	3.877	1.29242	4.31	0.001
Time	3	1.368	1.368	0.4559	8.58	0.014
Residual error	6	0.319	0.319	0.0532		
Total	15	213.397				
Analysis of variance for S/N ratios of Cu						
pH	3	176.536	176.536	58.8453	708.87	0.000
Dosage	3	2.821	2.821	0.9403	11.33	0.007
Time	3	0.524	0.524	0.1747	2.10	0.201
Residual error	6	0.498	0.498	0.0830		
Total	15	180.379				

impact on each individual response. The simultaneous effects of pH and dose on color, COD, and TSS elimination are shown in Fig. 4a. Maximum of 91% color removal showed at pH 6 and 1 g/L MACL coagulant dosage, the removal percentage was increased upto pH 6 after that color removal percentage was decreased. The coagulant had relatively low removal effectiveness in alkali pH, due to a reduction in positive hydrogen ions subareas in coagulation process. Coagulant dosage concentration also played a crucial role in the removal of COD, color, TSS and heavy metals. At 1 g/L of coagulant dose, a maximum of 77% COD and 83% TSS elimination were achieved. Fig. 4a shows that the removal percentage efficiency increases with increasing coagulant dosage up to 1 g/L, but decreases as MACLC dosage concentration increases. Since there was a reduction

Table 7
Model summary of optimization of experimental condition and contribution of each factor

Source	pH	Dosage (mg/L)	Time (min)	Agitation speed (rpm)	Observed (%)	Predicted (%)	Confidence limit (%)	*S*	*R*-sq. (%)	*R*-sq. Adj. (%)
	Value	Value	Value	Value						
	Level	Level	Level	Level						
Color	6	1	60	60	91	95	90–95	0.7652	99.60	97.99
	3	2	4	2						
COD	6	1	60	60	77	80	95–100	0.9854	99.69	98.47
	3	2	4	2						
TSS	6	1	60	60	83	85	95–100	0.9140	99.55	97.75
	3	2	4	2						
Cr	6	1	60	60	89	91	95–100	0.2511	99.92	99.60
	3	2	4	2						
Cd	6	1	60	60	78	85	90–95	0.1196	99.98	99.90
	3	2	4	2						
Cu	6	1	60	60	87	90	95–100	0.3464	99.80	99.00
	3	2	4	2						

Table 8
Summary of the coagulants used for the treatment landfill leachate

S. No.	Coagulant	Percentage of removal parameters	References
1.	Sago trunk	Colour (94.7%), SS (99.2%)	[25]
2.	Psyllium husk	COD (58%), colour (79%), TSS (78%)	[26]
3.	Jackfruit seed starch	COD (26.85%)	[28]
4.	*Nephelium lappaceum* seed	COD (64.4%), colour (77.8%)	[30]
5.	*Ocimum basilicum* L. with alum	COD (65.7%), colour (81.8%),	[62]
6.	Lateritic soil		[63]
7.	*Moringa oleifera* seed	Cu (98%), Cd (98%)	[64]
8.	*Tamarindus indica* seeds	SS (99.5%), colour (97.4%), COD (73.6%)	[65]
9.	*Salvia hispanica* seeds	COD (39.76%)	[66]
10.	$SnCl_4$ as a coagulant and *Jatropha curcas* as flocculant	Colour (98.53%), COD (74.29%), SS (99.78%)	[67]
11.	Guar gum	COD (22.568%)	[68]
12.	Tapioca starch flour	SS (98%), colour (96%), COD (60%)	[69]
13.	Ferric chloride	COD (69.0%), BOD_5 (60.0%), colour (88.0%)	[70]
14.	Ferric chloride	COD (71.0%), turbidity (95.0%)	[71]
15.	*Musa acuminata* Colla leaves	Colour (91%), COD (77%), TSS (83%), Cr (89%), Cd (78%), Cu (87%)	Present study

or saturation in coagulant bridge sites, higher coagulant dose concentration shown repulsion during the process. Charge neutralization is slowed down and sludge formation is accelerated at a less coagulant bridge site. Before and after the treatment of the landfill leachate, the AAS instrument was used to measure the percentage of heavy metal removal. The initial concentration of chromium ion was found in leachate sample was 1.198 mg/L, after the coagulation treatment with MACLC the concentration of chromium ion was found as 0.132 mg/L in treated sample. The concentration of Cu and Cd ions was measured both before and after the treatment process using the same methodology. Using MACLC, this coagulation method was capable of removing 78% of Cd and 87% of Cu. The effect of pH and coagulant dosage as controller parameters on the removal of the metals Cr, Cd, and Cu is shown in Fig. 4b. At pH 6 and 1 g/L of MACL coagulant dose, the maximum removal of 89% Cr, 78% Cd, and 87% Cu was accomplished. Agro-based bio waste has more amino and hydroxyl groups than other types of biowaste, and it also has more proteins and polysaccharides on its surface, which makes it more likely to bond with heavy metal ions [56]. The agricultural-based coagulant MACLC may react with metal cations during this coagulation process and produce colloid-polymers during the charge neutralization process, leading to a significant amount of heavy metal removal. The removal of

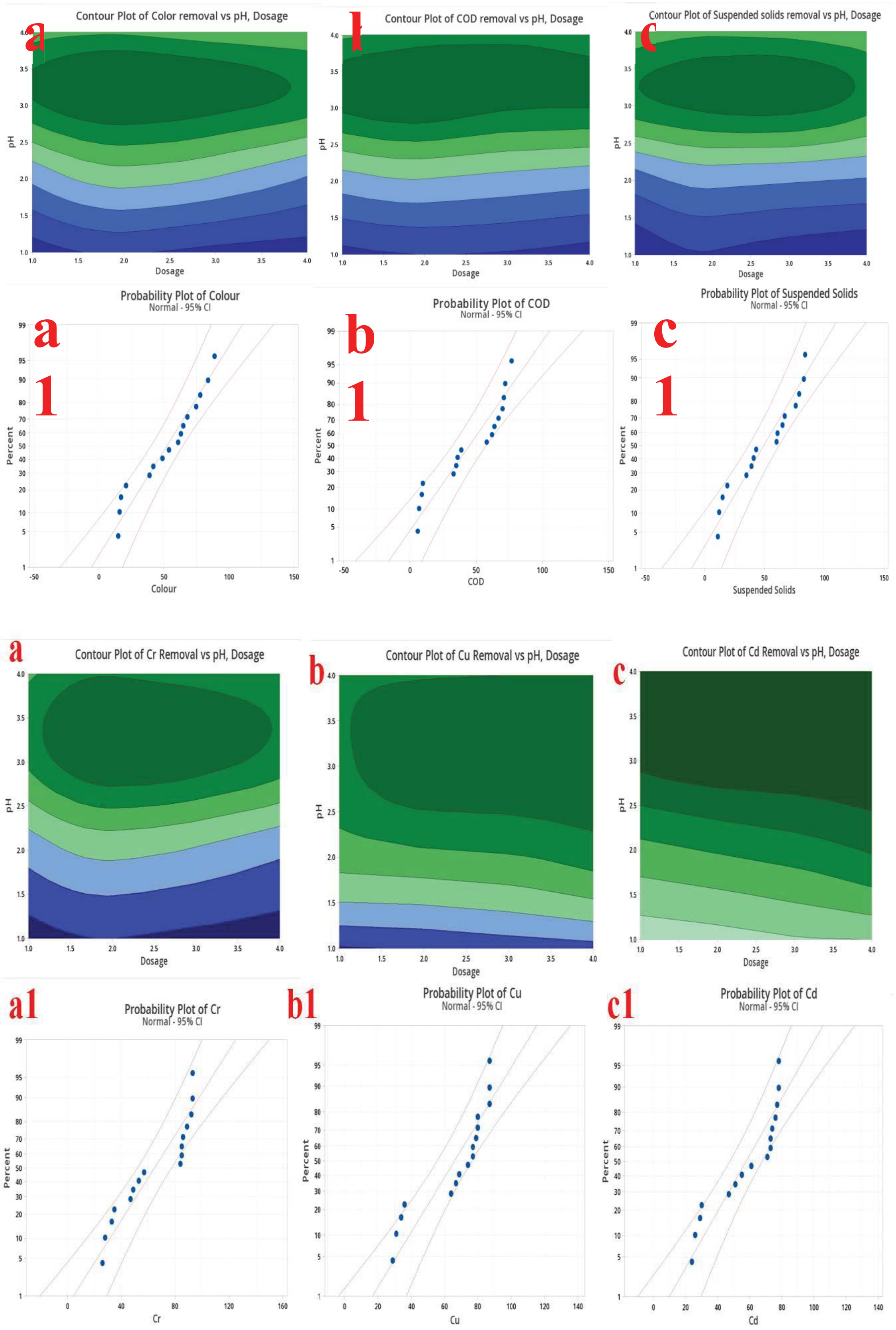


Fig. 4. 2D contour plot (a, b, c) and probability plot (a1, b1, c1) for removal of (a) color, chemical oxygen demand, and total suspended solids and (b) heavy metals Cr, Cu, and Cd.

heavy metal ions from wastewater is naturally enhanced by the amino groups and protein functional groups present in natural coagulant. More positive charge in the wastewater improves the creation of more flocs and the sweeping mechanism, which accelerates the degradation and settlement of pollutants [57]. Similarly, banana waste applied for the removal of heavy metals Cr and Cd by various researchers and reported that it has efficient capacity in the heavy metal removal. A maximum of 91.94% of Cr(VI) ion was removed by banana peel as adsorbent [58], banana blossom peel adsorbent has good potential on removal of chromium(VI) ion [59] and banana peel adsorbent reported as a good alternative for removal of heavy metal ions cadmium (93.2%) and lead (83.78%) in *Oryza sativa* rice. Exchange of cations like sodium, magnesium, calcium, and potassium, and fixation of proton in wastewater sample and bio-coagulant may increases the removal of metal ions [60]. Higher levels of organic components in wastewater considerably enhance the coagulation process; however, the main drawback in this study is the production of sludge. Fig. 4a (a1, b1, c1) and b (a1, b1, c1), which depict the likelihood probability plots for all the residuals, respectively. It indicated the good interaction effects [61] between the pH and coagulant dose for the removal of color, COD, TSS, Cr, Cd, and Cu. In the pollutant degradation in landfill leachate, contact period and agitation speed also played significant roles. After initially declining at 10–20 min of reaction time, the removal effectiveness of MACLC gradually increased up to 60 min. However, the pollutant removal percentage starts to fall after 60 min due to the coagulant dosage having less available bridging space. With increasing and significantly reducing agitation speed, the interaction impact between the control and response variables is also diminished. The agitation speed that was most effective for this investigation was 90 rpm. MACLC has demonstrated significant pollutant degrading efficiency with minimal sludge formation, making it a valuable alternative to chemical coagulants since it is a corrosive-free coagulant and also readily available agricultural waste.

The characteristics of the utilized coagulants have a significant impact on the performance of the coagulation process, which can either increase or decrease the effectiveness of the treatment. Natural coagulants are always readily available and are known for being free of toxins, particularly those made from plant extracts. The coagulation process for the treatment of landfill leachate in earlier studies showed good removal efficiency using natural coagulants. The removal effectiveness of various natural coagulants on the landfill leachate is listed in Table 8. Table 8 indicates that natural coagulants were both highly competitive with chemical coagulants and demonstrated good removal efficiency of the pollutant characteristics of the landfill leachate. Although the coagulation process is an efficient and inexpensive way to treat leachate wastewater, it has some drawbacks such as production of sludge. After dewatering, the sludge waste can be utilized for agricultural purposes and can be used in manufacturing of cement and brick as coarse aggregate or fine aggregate.

4. Conclusion

The potential of MACLC for removing color, COD, TSS, Cr, Cd, and Cu from leachate is demonstrated in the current study. In acidic pH conditions, MACL coagulant has shown higher pollutant removal efficiency. The elimination of color, COD, TSS, Cr, Cd, and Cu was greatly aided by pH. Maximum 91% of color, 77% of COD, 83% of TSS, 89% of Cr, 78% of Cd, and 87% of Cu at 6 pH, 1 g/L coagulant dosage, contact time 60 min, and agitation speed 90 rpm. Taguchi experiment design (OA L_{16}) was efficient design method for the pollutant removal of leachate. More than 97% of the pollutant removal was achieved using R^2 values and adjacent R^2 values, which speaks to the effectiveness of this design. Least S, p (<0.05) and high F-value also confirms the adequacy of this model. The probability plot effectively illustrates how the responses and parameters interact. The results of this experiment indicate that MACLC is the best coagulant since it has the highest percentage of pollutant elimination, is simpler to prepare, is more widely available is simple to dispose of, and is also not harmful to the environment. In order to analyse the interactions between different parameters, the Taguchi experiment design method decreases the number of tests, chemical usage, expense, and time. A viable method for analysing the optimization of process variables for the pollutant reduction in leachate is this robust design.

Conflicts of interest

The authors do not have any competing interest.

Authors contributions

M. Mohan contributed in the collection of leachate samples, carried out optimization study; find the inference of Taguchi optimization study and drafting the original manuscript.

P. Arulmathi contributed in analysis the results and evaluates the adequacy of Taguchi optimization design, drafting and review the manuscript.

References

[1] T.A. Kurniawan, W.-h. Lo, G.Y.S. Chan, Radicals-catalyzed oxidation reactions for degradation of recalcitrant compounds from landfill leachate, Chem. Eng. J., 125 (2006) 35–57.

[2] N. Mekjinda, R.J. Ritchie, Breakdown of food waste by anaerobic fermentation and non-oxygen producing photosynthesis using a photosynthetic bacterium, Waste Manage., 35 (2015) 199–206.

[3] J.E. Silveira, J.A. Zazo, G. Pliego, E.D. Bidóia, P.B. Moraes, Electrochemical oxidation of landfill leachate in a flow reactor: optimization using response surface methodology, Environ. Sci. Pollut. Res., 22 (2015) 5831–5841.

[4] T.A. Kurniawan, W.-h. Lo, Removal of refractory compounds from stabilized landfill leachate using an integrated H_2O_2 oxidation and granular activated carbon (GAC) adsorption treatment, Water Res., 43 (2009) 4079–4091.

[5] S.M. Raghab, A.M. Abd El Meguid, H.A. Hegazi, Treatment of leachate from municipal solid waste landfill, HBRC J., 9 (2013) 187–192.

[6] A.A. Tatsi, A.I. Zouboulis, K.A. Matis, P. Samaras, Coagulation–flocculation pretreatment of sanitary landfill leachates, Chemosphere, 53 (2003) 737–744.

[7] N. Paxeus, Organic compounds in municipal landfill leachates, Water Sci. Technol., 42 (2000) 323–333.

[8] T. Griessler Bulc, M. Zupančič Justin, Chapter 4 – Sustainable Solution for Landfill Leachate with a Use of Phytoremediation, A.A. Velinni, Ed., Landfill Research Trends, Nova Science Publishers, Inc., 2007, pp. 103–139.

[9] A. Mojiri, Phytoremediation of heavy metals from municipal wastewater by *Typhadomingensis*, Afr. J. Microbiol. Res., 6 (2012) 643–647.

[10] S. Ghafari, H.A. Aziz, M.H. Isa, A.A. Zinatizadeh, Application of response surface methodology (RSM) to optimize coagulation–flocculation treatment of leachate using poly-aluminum chloride (PAC) and alum, J. Hazard. Mater., 163 (2009) 650–656.

[11] A. Maleki, M.A. Zazouli, H. Izanloo, R. Rezaee, Composting plant leachate treatment by coagulation–flocculation process, Am.-Eurasian J. Agric. Environ. Sci., 5 (2009) 638–643.

[12] L.M. Rui, Z. Daud, A.A.A. Latiff, A. Aziz, Treatment of leachate by coagulation–flocculation using different coagulants and polymer: a review, Int. J. Adv. Sci. Eng. Inf. Technol., 2 (2012) 1–4.

[13] D.Y. Ha, S.H. Cho, Y.K. Kim, S.W. Leung, A new approach to characterize biodegradation of organics by molecular mass distribution in landfill leachate, Water Environ. Res., 80 (2008) 748–756.

[14] E.M. Rocha, V.J. Vilar, A. Fonseca, I. Saraiva, R.A. Boaventura, Landfill leachate treatment by solar-driven AOPs, Sol. Energy, 85 (2011) 46–56.

[15] D.L. Baun, T.H. Christensen, Speciation of heavy metals in landfill leachate: a review, Waste Manage. Res., 22 (2004) 3–23.

[16] V. Chanudet, M. Filella, Size and composition of inorganic colloids in a peri-alpine, glacial flour-rich lake, Geochim. Cosmochim. Acta, 72 (2008) 1466–1479.

[17] R.I. Méndez-Novelo, E.R. Castillo-Borges, M.R. Sauri-Riancho, C.A. Quintal-Franco, G. Giacomán-Vallejos, B. Jiménez-Cisneros, Physico-chemical treatment of Merida landfill leachate for chemical oxygen demand reduction by coagulation, Waste Manage. Res., 23 (2005) 560–564.

[18] S.F. Ramli, H.A. Aziz, F.M. Omar, M.S. Yusoff, H. Halim, M.A. Kamaruddin, K.S. Ariffin, Y.-T. Hung, Reduction of COD and highly coloured mature landfill leachate by tin tetrachloride with rubber seed and polyacrylamide, Water, 13 (2021) 3062, doi: 10.3390/w13213062.

[19] M. Samadi, M. Saghi, A. Rahmani, J. Hasanvand, S. Rahimi, M.S. Syboney Hamadan, Landfill leachate treatment by coagulation–flocculation process, J. Environ. Health Sci. Eng., 7 (2010) 253–258.

[20] J.F. Perez, J. Llanos, C. Saez, C. Lopez, P. Canizares, M.A. Rodrigo, Treatment of real effluents from the pharmaceutical industry: a comparison between Fenton oxidation and conductive-diamond electro-oxidation, J. Environ. Manage., 195 (2017) 216–223.

[21] M. Mathuram, R. Meera, G. Vijayaraghavan, Application of locally sourced plants as natural coagulants for dye removal from wastewater: a review, J. Mater. Environ. Sci., 2508 (2018) 2058–2070.

[22] R. Kant, Textile dyeing industry an environmental hazard, Nat. Sci., 4 (2012) 1–5.

[23] N.A. Awang, H.A. Aziz, *Hibiscus rosa-sinensis* leaf extract as coagulant aid in leachate treatment, Appl. Water Sci., 2 (2012) 293–298.

[24] F.P. Camacho, V.S. Sousa, R. Bergamasco, M.R. Teixeira, The use of *Moringa oleifera* as a natural coagulant in surface water treatment, Chem. Eng. J., 313 (2017) 226–237.

[25] H.A. Aziz, N.I.M. Sobri, Extraction and application of starch-based coagulants from sago trunk for semi-aerobic landfill leachate treatment, Environ. Sci. Pollut. Res., 22 (2015) 16943–16950.

[26] Y.A.J. Al-Hamadani, M.S. Yusoff, M. Umar, M.J.K. Bashir, M.N. Adlan, Application of psyllium husk as coagulant and coagulant aid in semi-aerobic landfill leachate treatment, J. Hazard. Mater., 190 (2011) 582–587.

[27] S. Omar, N.S.M. Yusoff, M.S. Muhamad, N. Hamidon, N.H.A. Hamid, H. Harun, N.M. Sunar, R. Ali, The characterization of banana blossom peel and floret as an adsorbent for the removal of manganese in groundwater, Mater. Sci. Forum, 981 (2020) 362–368.

[28] M.S. Yusoff, N.A. Mohamad Zuki, Optimum of treatment condition for *Artocarpus heterophyllus* seeds starch as natural coagulant aid in landfill leachate treatment by RSM, Appl. Mech. Mater., 802 (2015) 484–489.

[29] M. Nithya, M. Abirami, The leachate treatment by using natural coagulants (pine bark and chitosan), Int. Res. J. Eng. Technol., 5 (2018) 2711–2714.

[30] H.A. Aziz, N.S. Rahmat, M.Y.D. Alazaiza, The potential use of *Nephelium lappaceum* seed as coagulant–coagulant aid in the treatment of semi-aerobic landfill leachate, Int. J. Environ. Res. Public Health, 19 (2021) 420, doi: 10.3390/ijerph19010420.

[31] N.S.M. Zin, Z.A. Zulkapli, Application of dual coagulant (alum + barley) in removing colour from leachate, MATEC Web Conf., 103 (2017) 06002, doi: 10.1051/matecconf/201710306002.

[32] T.J.H. Banch, M.M. Hanafiah, A.F.M. Alkarkhi, S.S. Abu Amr, Factorial design and optimization of landfill leachate treatment using tannin-based natural coagulant, Polymers, 11 (2019) 1349, doi: 10.3390/polym11081349.

[33] M.B. Silva, L.M. Carneiro, J.P.A. Silva, I. dos Santos Oliveira, H.J. Izário Filho, C.R. de Oliveira Almeida, An application of the Taguchi method (robust design) to environmental engineering: evaluating advanced oxidative processes in polyester-resin wastewater treatment, Am. J. Anal. Chem., 5 (2014) 828–837.

[34] Syafalni, H.K. Lim, N. Ismail, I. Abustan, M.F. Murshed, A. Ahmad, Treatment of landfill leachate by using lateritic soil as a natural coagulant, J. Environ. Manage., 112 (2012) 353–359.

[35] A. Martínez-Cruz, M.N.R. Valencia, J.A. Araiza-Aguilar, H.A. Nájera-Aguilar, R.F. Gutiérrez-Hernández, Leachate treatment: comparison of a bio-coagulant (*Opuntia ficus* mucilage) and conventional coagulants using multi-criteria decision analysis, Heliyon, 7 (2021) e07510, doi: 10.1016/j.heliyon.2021.e07510.

[36] H. Alwi, J. Idris, M. Musa, K.H. Ku Hamid, A preliminary study of banana stem juice as a plant-based coagulant for treatment of spent coolant wastewater, J. Chem., 2013 (2013) 165057, doi: 10.1155/2013/165057.

[37] R. Premkumar, S. Rajesh, M. Prasanna venkadesh, Feasibility study on application of natural coagulants, J. Phys. Conf. Ser., 2070 (2021) 012186, doi: 10.1088/1742-6596/2070/1/012186.

[38] N.A. Akbar, S. Sabri, A.A. Abu Bakar, N.S. Azizan, Removal of colour using banana stem adsorbent in textile wastewater, J. Phys. Conf. Ser., 1349 (2019) 012091, doi: 10.1088/1742-6596/1349/1/012091.

[39] V. Priya, P. Shrama, A. Kumar, Phytoremediation of banana leaf - functionalization of mercaptan-cellulose and its utilization for the removal of heavy metals in aqueous environment, Asian J. Adv. Basic Sci., 8 (2020) 26–38.

[40] A. Ali, K. Saeed, F. Mabood, Removal of chromium(VI) from aqueous medium using chemically modified banana peels as efficient low-cost adsorbent, Alexandria Eng. J., 55 (2016) 2933–2942.

[41] K.-H. Chong, P.L. Kiew, Potential of banana peels as bio-flocculant for water clarification, Prog. Energy Environ., 1 (2017) 47–56.

[42] S. Omar, N.S.M. Yusoff, M.S. Muhamad, N. Hamidon, N.H.A. Hamid, H. Harun, N.M. Sunar, R. Ali, The characterization of banana blossom peel and floret as an adsorbent for the removal of manganese in groundwater, Mater. Sci. Forum, 981 (2020) 362–368.

[43] Z. Beril Gönder, S. Arayici, H. Barlas, Advanced treatment of pulp and paper mill wastewater by nanofiltration process: effects of operating conditions on membrane fouling, Sep. Purif. Technol., 76 (2011) 292–302.

[44] B. Basak, B. Bhunia, S. Mukherjee, A. Dey, Optimization of physicochemical parameters for phenol biodegradation

by *Candida tropicalis* PHB5 using Taguchi methodology, Desal. Water Treat., 51 (2013) 6846–6862.

[45] A. Reyhani, H. Mashhadi Meighani, Optimal operating conditions of micro- and ultra-filtration systems for produced-water purification: Taguchi method and economic investigation, Desal. Water Treat., 57 (2016) 19642–19654.

[46] Z.Z. Abidin, N. Ismail, R. Yunus, I.S. Ahamad, A. Idris, A preliminary study on *Jatropha curcas* as coagulant in wastewater treatment, Environ. Technol., 32 (2011) 971–977.

[47] B.D. Cobb, J.M. CIarkson, A simple procedure for optimising the polymerase chain reaction (PCR) using modified Taguchi methods, Nucleic Acids Res., 22 (1994) 3801–3805.

[48] S.M. Phadke, Quality Engineering Using Robust Design and Analysis, Prentice Hall Englewood Cliffs, NJ Google Scholar, 1989.

[49] A.H. Jawad, R.A. Rashid, M.A.M. Ishak, K. Ismail, Adsorptive removal of methylene blue by chemically treated cellulosic waste banana (*Musa sapientum*) peels, J. Taibah Univ. Sci., 12 (2018) 809–819.

[50] A. Jeyaseelan, N. Viswanathan, Design of amino-functionalized benzene-1,4-dicarboxylic acid-fabricated lanthanum-based metal–organic frameworks for defluoridation of water, J. Chem. Eng. Data, 65 (2020) 5328–5340.

[51] K. Lal, A. Garg, Physico-chemical treatment of pulping effluent: characterization of flocs and sludge generated after treatment, Sep. Sci. Technol., 52 (2017) 1583–1593.

[52] I.A.W. Tan, A.L. Ahmad, B.H. Hameed, Adsorption isotherms, kinetics, thermodynamics and desorption studies of 2,4,6-trichlorophenol on oil palm empty fruit bunch-based activated carbon, J. Hazard. Mater., 164 (2009) 473–482.

[53] A. Asghar, A.A. Abdul Raman, W.M.A.W. Daud, A comparison of central composite design and Taguchi method for optimizing Fenton process, Sci. World J., 2014 (2014) 869120, doi: 10.1155/2014/869120.

[54] S.S. Madan, K.L. Wasewar, Optimization for benzene acetic acid removal from aqueous solution using CaO_2 nanoparticles based on Taguchi method, J. Appl. Res. Technol., 15 (2017) 332–339.

[55] D.C. Montgomery, G.C. Runger, Applied Statistics and Probability for Engineers (with CD), John Wiley & Sons, 2007.

[56] M. Abdulsalam, A. Al-Arbeed, Brown coal fly ash concrete mixture for industrial waste containment, Adv. Mater. Res., 831 (2014) 73–77.

[57] N.K. Mondal, A. Samanta, S. Chakraborty, W.A. Shaikh, Enhanced chromium(VI) removal using banana peel dust: isotherms, kinetics and thermodynamics study, Sustainable Water Resour. Manage., 4 (2018) 489–497.

[58] M.A. Selimin, A.F.A. Latif, M.S. Muhamad, H. Basri, T.C. Lee, Adsorption efficiency of banana blossom peels (*Musa acuminata* Colla) adsorbent for chromium(VI) removal, Mater. Today Proc., 57 (2022) 1262–1268.

[59] M. Motaghi, P. Ziarati, Adsorptive removal of cadmium and lead from *Oryza sativa* rice by banana peel as bio-sorbent, Biomed. Pharmacol. J., 9 (2016) 739–749.

[60] N. Meunier, J. Laroulandie, J.F. Blais, R.D. Tyagi, Cocoa shells for heavy metal removal from acidic solutions, Bioresour. Technol., 90 (2003) 255–263.

[61] N. Sh. El-Gendy, S.S. Abu Amr, R.A. El-Salamony, H.N. Nassar, Factorial design analysis for biosorption of Reactive Red-84 dye using fermentation spent waste biomass, biosorbent regeneration and desorbed dye photo-degradation using TiO_2 nanoparticles, Desal. Water Treat., 56 (2015) 3403–3413.

[62] M.A. Rasool, B. Tavakoli, N. Chaibakhsh, A.R. Pendashteh, A.S. Mirroshandel, Use of a plant-based coagulant in coagulation–ozonation combined treatment of leachate from a waste dumping site, Ecol. Eng., 90 (2016) 431–437.

[63] Syafalni, H.K. Lim, N. Ismail, I. Abustan, M.F. Murshed, A. Ahmad, Treatment of landfill leachate by using lateritic soil as a natural coagulant, J. Environ. Manage., 112 (2012) 353–359.

[64] T.C. Shan, M.A. Matar, E.A. Makky, E.N. Ali, The use of *Moringa oleifera* seed as a natural coagulant for wastewater treatment and heavy metals removal, Appl. Water Sci., 7 (2017) 1369–1376.

[65] H.A. Aziz, Y.C. Yii, S.F.F. Syed Zainal, S.F. Ramli, C.O. Akinbile, Effects of using *Tamarindus indica* seeds as a natural coagulant aid in landfill leachate treatment, Global Nest J., 20 (2018) 373–380.

[66] B. Tawakkoly, A. Alizadehdakhel, F. Dorosti, Evaluation of COD and turbidity removal from compost leachate wastewater using *Salvia hispanica* as a natural coagulant, Ind. Crops Prod., 137 (2019) 323–331.

[67] S.F.F.S. Zainal, H.A. Aziz, F.M. Omar, M.Y.D. Alazaiza, Influence of *Jatropha curcas* seeds as a natural flocculant on reducing tin(IV) tetrachloride in the treatment of concentrated stabilised landfill leachate, Chemosphere, 285 (2021) 131484, doi: 10.1016/j.chemosphere.2021.131484.

[68] S.Y. Cheng, P.-L. Show, J.C. Juan, T.C. Ling, B.F. Lau, S.H. Lai, E.P. Ng, Sustainable landfill leachate treatment: optimize use of guar gum as natural coagulant and floc characterization, Environ. Res., 188 (2020) 109737, doi: 10.1016/j.envres.2020.109737.

[69] N.S.M. Zin, H.A. Aziz, M.N. Adlan, A. Ariffin, M.S. Yusoff, I. Dahlan, Treatability study of partially stabilized leachate by composite coagulant (prehydrolyzed iron and tapioca flour), Int. J. Sci. Res. Knowl., 2 (2014) 313, doi: 10.12983/ijsrk-2014-p0313-0319.

[70] I. El Mrabet, M. Nawdali, S. Rafqah, H. Valdés, M. Benzina, H. Zaitan, Low-cost biomass for the treatment of landfill leachate from Fez City: application of a combined coagulation–adsorption process, Euro-Mediterr. J. Environ. Integr., 5 (2020) 63, doi: 10.1007/s41207-020-00201-y.

[71] H. Bellouk, I. El Mrabet, K. Tanji, M. Nawdali, M. Benzina, M. Eloussaief, H. Zaitan, Performance of coagulation-flocculation followed by ultra-violet/ultrasound activated persulfate/hydrogen peroxide for landfill leachate treatment, Sci. Afr., 17 (2022) e01312, doi: 10.1016/j.sciaf.2022.e01312.