Evaluation of combined thermo-chemical processes for the treatment of landfill leachate using virgin and recovered $FeCl_3$ coagulants

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Abstract

Sludge resulting from treatment of municipal waste landfill leachate contains suitable cationic substances such as Fe-based recovered coagulants which, if not recovered, can cause environmental problems. The present study aimed to maximise coagulant recoverability and investigate its potential reuse for the treatment of municipal waste landfill leachate. The study focused on establishing (i) the effect of mineral acids on leaching of Fe, (ii) the % of maximum recovery of Fe coagulant, (iii) the impact of ultrasound on recovery, and (iv) effectiveness of recovered coagulant when reused in coagulation-flocculation treatment of landfill leachate. Sulfuric acid outran hydrochloric acid in performance, with the acid leaching process leading to the recovery of 70.12% of Fe (acid concentration=3.80 M, solid-to-liquid ratio=8%, and heating time=5 h). Subsequently, a developed acid leaching process was tested, which results showed that the highest rate of Fe recovery occurred without ultrasound treatment, meaning the use of it could reduce the recovery rate due to the increase in the iron (III) oxide-hydroxide [$Fe(OH)_3$] sedimentation. Comparative experiments were undertaken with the recovered and virgin coagulants. These revealed that Fe-based recovered coagulant led to the 60.21% and 91.40% removal of COD and total suspended solid respectively, while the values of the COD and total suspended solid removal with the virgin $FeCl_3$ were 7.66% and 6.42% lower than that of Fe under optimal conditions (dosage=9.38 g/L, pH=8.94, settling time=52.9 min). The present study established that Fe recovered could be exploited as an eco-friendly coagulant to replace $FeCl_3$ in the landfill leachate treatment.

Keywords: Iron chloride, Coagulation-Flocculation, Acid leaching, Coagulant recovery, Sludge from treated landfill leachate.

1. Introduction

Remarkable quantities of coagulant are consumed in the coagulation-flocculation process involved in leachate treatment of municipal solid waste, consequently a large volume of sludge is produced. The latter is a well-known global issue arising from leachate treatment (Nayeri & Mousavi, 2022). The discharge of sludge from treated landfill leachate into the water and soil environments can thus cause color change, and then increase their turbidity and hardness. Additionally, the bio-available metals in this material can also be toxic to the life of aquatic organisms (Crittenden *et al.*, 2012; Xia *et al.*, 2018), leading to secondary pollution of the environment (Mudasir *et al.*, 2016; Ooi *et al.*, 2018). Thanks to regulatory changes and more stringent environmental regulations, other techniques, such as incineration, dewatering, landfilling, and digesting have been recently practiced around the world (Ahmad *et al.*, 2016; Huzir *et al.*, 2019; Keeley *et al.*, 2014). Nevertheless, some disadvantages of these techniques include costs and difficulties of transportation and dewatering operations (Shak & Wu, 2015; Wei *et al.*, 2017), which grow the need for lands to bury waste in landfills (Nair & Ahammed, 2014). Also, heat existing in sludge (Nair & Ahammed, 2017), or emission of greenhouse gases (GHGs) (Devi & Saroha, 2017) and other toxic compounds during biological methods and incineration have caused their less use. Against this background, managing residual sludge in an economic, sustainable, and eco-friendly manner is an important issue (Ahmad *et al.*, 2016). This has resulted in real efforts aimed at sludge reuse (Evuti & Lawal, 2011; Wei *et al.*, 2017) in applications, such as building materials (Pokhara *et al.*, 2019; Yaras, 2020), wastewater treatment (Mansour *et al.*, 2020; Singh *et al.*, 2017), and soil amendment (Elmi & AlOlayan, 2020; Öden *et al.*, 2019).

As the high concentration of metals in sludge limits its use on land (Babatunde & Zhao, 2007), the recovery of coagulants originating from the treatment of this slurry material is another sustainable area of interest that has been reported in research (Ayoub *et al.*, 2017; Chakraborty *et al.*, 2020; Salehin *et al.*, 2020). As previously mentioned, the performance and cost of recovered coagulant is comparable to the virgin one for water and wastewater treatment (Ishikawa *et al.*, 2007; Xu *et al.*, 2009b), and it can be above all compensated by reducing the costs of virgin coagulants, dewatering, and sludge disposal (Chakraborty *et al.*, 2020). In the coagulant recovery process, water-bound molecules are often transferred in the sludge mass, and problems such as the sludge volume and the cost of transporting it after recovery are greatly reduced (Huang *et al.*, 2010; Li *et al.*, 2005).

Over recent years, some methods, such as acid digestion (Joy *et al.*, 2020; Ooi *et al.*, 2018), alkaline treatment (Atia & Spooren, 2020; Chen *et al.*, 2019), ion exchange (Meng *et al.*, 2019; Mohammadtabar *et al.*, 2019), membrane processes (Keeley *et al.*, 2014; Keeley *et al.*, 2016a), and absorption (Cho *et al.*, 2017; Xia *et al.*, 2020) have been utilized for coagulant recovery. The most common method for this purpose is acid digestion, because of its advantages; namely, high efficiency, sludge volume reduction, improved dewatering conditions, and low costs for coagulant recovery and sludge disposal, which have made it one of the most popular recovery practices (Chen *et al.*, 2012; Huang *et al.*, 2010). Additionally, this method has received much attention in most studies at the laboratory, pilot, and factory scale (Xu *et al.*, 2009b). For example, the cost of using the coagulant recovered by acid digestion has been already compared with that of virgin aluminum sulfate (alum) and polyaluminum chloride (PACl) coagulants for wastewater clarification. In this comparison, the cost of coagulant recovery had been lower than that of virgin coagulants (Ishikawa *et al.*, 2007). In addition, another study demonstrated that the initial operational costs involved in the recovery as well as the acid leaching process could be compensated by reducing the costs of virgin coagulants, dewatering, and sludge disposal (Chakraborty *et al.*, 2020). Coagulant recovery is a simple process, its efficiency is often controlled by various factors, such as sludge characteristics, coagulant type and dosage, and process operating conditions (including solution pH, mixing speed, intensity, and duration, and temperature) (Chen *et al.*, 2012; Xu *et al.*, 2009b). In line with various reports on evaluation parameters (e.g., different coagulant sources and dosages), there is a need to shed light on the effects of the quality of raw effluents, as well as coagulant type and dosage on the recovery rate in controlled environments (Nair & Ahammed, 2017). Hence, Chakraborty *et al.* (2020) reported the optimum pH of 1.5 for coagulant recovery from sludge production. In addition, the recovery rates of Al from sludge by 61-99% have been so far reported (Jung *et al.*, 2016; Nair & Ahammed, 2017). In another study, the recovery of alum from water treatment plant sludge by the acid leaching process had been further analyzed with regard to some significant factors, such as acid concentration, sludge-to-acid ratio, mixing speed, temperature, and mixing duration, affecting the amount of the Al recovery. The results had then shown that the Al recovery rate was 82.4% in optimal conditions (Fouad *et al.*, 2017a).

One of the main concerns on recycling back in use the recovered coagulant is the high amount of heavy metals and natural organic matters in the recovered materials (Chen *et al.*, 2012; Xu *et al.*, 2009b). Although such materials cannot be used for water treatment, they can be find application in treating complex wastewater where the higher concentration of metals is accepted. Therefore, recovered coagulant and sludge have been so far utilized in wastewater treatment (Chakraborty *et al.*, 2017), landfill leachate (Ishikawa *et al.*, 2007), and chemical primary treatment (Chakraborty *et al.*, 2020). In particular, the feasibility of recovering alum and Fe from sludge and their reuse in the preliminary treatment process has been investigated in depth by Xu *et al.* (2009a); Xu *et al.* (2009b). The optimum pH for the acidic digestion of alum and iron (III) chloride ($FeCl_3$) sludge has been also reported as 2.5 and 1.5, respectively. The optimal mixing duration has been further obtained as 30 and 20 min, respectively. Due to the dissolution of alum, Fe, and some other substances during the acid leaching process, the amount of sludge had significantly decreased. This was 40.3% less than original sludge volume obtained from the Fe-based coagulant at optimal pH (Xu *et al.*, 2009a; Xu *et al.*, 2009b). Chakraborty *et al.* (2017), using recovered coagulant, had further reported a reduction in total dissolved solids and chemical oxygen demand (COD) by 60-85% and 50-65%, respectively. Some researchers, such as Xu *et al.* (2009b) and Keeley *et al.* (2016b) had similarly found the removal efficiency of 53% for both COD and phosphorus (P), using recovered coagulant. Therefore, one of the main objectives in the present study was to investigate the reuse potential of Fe as a recovered coagulant in the treatment of waste landfill leachate, due to the key properties of Fe compounds to manage odors release and prevent corrosion (Gutierrez *et al.*, 2010) in water and wastewater treatment plants. In a study, coagulant recovered from wet sludge with 40 mL/L of sulfuric acid (3.5 N) showed turbidity removal of 96% with minimal cost in wastewater treatment. With this optimal condition, 76.3% recovery of aluminum and 67% volume reduction in water treatment sludge were achieved (Mora-León *et al.*, 2022). Kang *et al.* (2022) reported that under optimal conditions (initial pH 7.0 and sludge dosage containing aluminum 1588 mg/L), 87.8% of total suspended solid can be removed in a wastewater treatment process. Also, in another study, the reused treated sludge saved the hydrated lime dose applied (up to 28.4%) and reduced the sedimented sludge volume (up to 24.7%) (Madeira *et al.*, 2023). This study investigates for the first time the use of recovered Fe to achieve maximum treatment conditions in the ultrasonically enhanced leaching process (namely, acid concentration, solid to liquid ratio, and mixing duration) for waste landfill leachate. The four phased objectives under consideration were to establish (i) the effect of mineral acids on leaching of Fe, (ii) the % of maximum recovery of Fe coagulant, (iii) the impact of ultrasound on recovery, and (iv) effectiveness of recovered coagulant when reused in coagulation-flocculation treatment of landfill leachate. For this purpose, a central composite design (CCD) within the response surface methodology (RSM) was employed as an optimization tool to identify and model the effective treatment conditions of municipal waste leachate and Fe recovery from sludge of treated landfill leachate, and its reuse performance. This study also explores the chemistry of

Fe in sludge from treated landfill leachate and acid leaching solution using emission-scanning electron microscopy (FE-SEM), energy-dispersive X-ray spectroscopy (EDS), inductively coupled plasma (ICP-OES) spectroscopy, and X-ray fluorescence (XRF).

2. Materials and methods

2.1. Landfill leachate

The leachate samples were collected from the Municipal Solid Waste Landfill No. 1 in the city of Mashhad, affiliated to the Waste Management Organization of Razavi Khorasan Province, from Neyshabur-Mashhad Road, Iran. The samples collected were then transported to the laboratory in high-density polyethylene containers, and kept at a temperature of 4° C (APHA, 2005), until use in order to minimize biodegradation. Before the sludge treatment took place, sample characteristics were also determined, as detailed in Section 3.1, via the analytical methods reported in Section 2.3

2.2. Experimental set-up

2.2.1. Sludge preparation by coagulation-flocculation

Coagulation-flocculation experiments were performed in a jar test apparatus, where 500 mL of leachate sample was injected into each of the glass jars and the effects of pH, coagulant dosage, and settling time were investigated. The optimized values of the study were 9.38 g/L and 8.98 for $FeCl_3$ dosage and pH, respectively, with rapid mixing speed (200 rpm) and slow mixing (40 rpm) having 3 and 30 min time, respectively (Yusoff *et al.*, 2018). Iron chloride ($FeCl_3$) as a commercial virgin coagulant, hydrochloric acid, sulfuric acid, and sodium hydroxide were purchased from Merck & Co. Then, the groups were allowed to remain for 52.9 min according to the settling time. Figure S1 shows the flow diagram of experimental study. The sludge settled at the bottom of the beakers was also collected, and subsequently placed in an oven at a temperature of 105 °C to remove the remaining water content for 24 h (Nair & Ahammed, 2017). Finally, it was crushed using a mortar, and the produced powder was then passed through a sieve with 30 mesh (500 µm). The chemical composition of the sludge from treated landfill leachate is reported in Section 3.2.

2.2.2. Fe coagulant recovery

Acid digestion is one of the most popular recovery practices (high efficiency, sludge volume reduction, improved dewatering conditions, and low costs for coagulant recovery) (Chen *et al.*, 2012; Huang *et al.*, 2010). Following preparation described in section 2.2.1, the sludge from treated landfill leachate was acidified (Phase 1 in Figure S1) using a mineral acid (H_2SO_4 and HCl in different concentrations were compared in effectiveness at equal molarity) which established H_2SO_4 was more efficient and safer. Then, 50 mL of H_2SO_4 was added to a 250-mL filter flask containing sludge samples from treated landfill leachate. As acid concentration and solid-to-liquid ratio (Ahmad *et al.*, 2021), as well as mixing duration (Nair & Ahammed, 2014), could thus affect coagulant recovery, a solid-to-liquid ratio of 1% was tested. The latter was increased gradually until a significant decrease in the response (Fe recovery) of the acid leaching process was observed. Since Fe recovery was very low in the solid-to-liquid ratio (less than 4%), the range of the study was fixed at 4.59-21.40% for the solid-to-liquid ratio. The range of other parameters is presented in Section 2.4.

The speed and mixing duration were further adjusted with an automatic controller. Table 1 shows a summary of the important parameters of the experimental conditions in researchers' studies.

Table 1. Range of operational parameters obtained from literature (Acid leaching process)

Operating parameter	Range	Reference
Mixing speed (rpm)	80-300	(Ahmad *et al.*, 2021; Cheng *et al.*, 2015; Parsons & Daniels, 1999)
Mixing duration (h)	0.1-8	(Nair & Ahammed, 2015; Ooi *et al.*, 2018)
Operating temperature (°C)	25-130	(Keeley *et al.*, 2016a; Meng *et al.*, 2016; Ooi *et al.*, 2018)
Settling time (min)	15-60	(Nair & Ahammed, 2017; Parsons & Daniels, 1999)
Centrifuge speed (rpm)	3700-5000	(Chakraborty *et al.*, 2020; Ooi *et al.*, 2018)

Based on the studies reported in Table 1, the solutions had been continuously stirred at a constant speed of 100 rpm (Nair & Ahammed, 2017) for different periods (Phase 2 in Figure S1). The reaction temperature was kept constant at 90 °C, using a hot plate and a Liebig condenser (Meng *et al.*, 2016). Then, it was allowed to settle for 20 min. Therefore, mixing speed, temperature, and settling time were considered as constant factors.

Following settling, the study focused on clarifying the effect of adding an ultrasound step (Phase 3 in Figure S1) on the optimal Fe recovery values obtained by acid leaching. For this purpose, ultrasonic generator with a frequency of 20 Hz and power of 100 W was used. Three scenarios were analysed: (i) without ultrasound, (ii) ultrasound with 20-min settling time (Deng *et al.*, 2009), and (iii) 5-min ultrasound per 1/2 h (Abdolhosseinzadeh *et al.*, 2015) of the acid leaching process with 20-min settling in the ultrasonic, were tested. After finishing the acid leaching process, the mixture was cooled and centrifuged for 10 min at 3700 rpm (Chakraborty *et al.*, 2020), which led to the separation of the solids from the solution. Then, the leaching solutions containing coagulants were analyzed in terms of the Fe content and Fe recovery was calculated using Equation 1 (Nair & Ahammed, 2014).

$$Fe\ recovery\ (\%) = \left[\frac{Fe_{LC}}{Fe_{WTS}}\right] \times 100 \qquad (1)$$

where Fe_{LC} and Fe_{WTS} are the amounts of Fe in the leaching solution and sludge from treated landfill leachate, respectively.

By achieving optimal values of coagulation-flocculation using commercial virgin $FeCl_3$ (coagulation-flocculation I in Figure S1), the recovered Fe was then used in place of the virgin commercial $FeCl_3$ in a jar test apparatus to compare these two coagulants in terms of COD and total suspended solid removal (coagulation-flocculation II in Figure S1). The recovered Fe used was obtained during the optimal conditions of the acid leaching process (Phase 1, 2 and 3 in Figure S1). After completion of coagulation-flocculation I and II, a sample of supernatant was extracted from approximately 3 cm under the surface of the sample and filtered for further measurement and analysis of metal pollutants. The percentage of each pollutant removal was calculated using Equation 2 (Daud *et al.*, 2018).

$$Removal\ (\%) = \left[1 - \frac{C_f}{C_i}\right] \times 100 \qquad (2)$$

where C_i and C_f are the initial and final concentrations of municipal solid waste leachate, respectively.

2.3. Analytical methods

The input and output parameters of the coagulation-flocculation (COD and total suspended solid) and acid leaching (Fe concentrations) processes were additionally reviewed and repeated. The Fe recovery data were next determined as the average. As well, the sludge sample from treated landfill leachate was analyzed using a detailed analysis. A TESCAN MIRA III FE-SEM (Czech), equipped with an EDS was used to analyze the floc morphology and major components. The XRF analysis for the elemental identification of the sludge from treated landfill leachate, including Fe compounds was then performed (PHILIPS Co., model PW1410, the Netherlands). The pH value was also determined with the General Purpose pH Meter, EDT DirectION, and other total suspended solid (TSS), COD and five-day biochemical oxygen demand (BOD_5) parameters based on the Association of Schools and Programs of Public Health (APHA) standards (APHA, 2005). The concentration of the heavy metals was further measured by inductively coupled plasma (ICP-OES) spectroscopy.

2.4. Statistical analysis

The statistical evaluation, modeling and optimization of the coagulation-flocculation and acid leaching experiments were further performed using the Design-Expert 11.0.0 software package. The central composite design (CCD) of the second phase in this study consisted of

20 experiments with five levels and three variables. The amount of rotatability also depended on the number of points in the design, as mentioned in Equation 2. The range and levels of the independent variables in the acid leaching process (namely, acid concentration, solid-to-liquid ratio, and heating time) for model development are given in Table 2.

$$\propto = [N_f]^{\frac{1}{4}} \quad (3)$$

where N_f is the number of points in the cube portion of the design ($N_f = 2^k$, k is the number of factors). Therefore, α is equal to $(2^3)^{1/4} = 1.682$ according to Eq .(3)

Table 2. Experimental range and coded levels of the independent variables for CCD design

Independent Factors	Symbols		Coded levels				
	Actual	Coded	-1.68	-1	0	+1	+1.68
Acid concentration (M)	X_1	x_1	1.32	2.00	3.00	4.00	4.68
Solid-to-liquid ratio (%)	X_2	x_2	4.60	8.00	13.00	18.00	21.41
Heating time (h)	X_3	x_3	0.98	2.00	3.50	5.00	6.02

To determine the adequacy of the model constructed and the relationship between the factors and responses, analysis of variance (ANOVA) and three-dimensional (3D) contour charts were used for evaluation. The quality of the appropriate polynomial model was also then expressed by the coefficient of determination (R^2) and the lack of fit (LOF). The Fe recovery of the quadratic model for predicting the optimal conditions can be expressed according to Equation 4, which is expanded according to the number of variables in the linear models, namely, the model with mutual effects, the quadratic model, and the reduced quadratic model.

$$Y = \beta_0 + \sum_{i=1}^{m} \beta_i X_i + \sum_{i=1}^{m} \beta_{ii} X_i{}^2 + \sum_{i=1}^{m} \sum_{j=2}^{i} \beta_{ij} X_i X_j + \varepsilon \quad (4)$$

where Y is the predicted response (Fe recovery), m is the number of factors, i is the linear coefficient, β_0 the constant coefficient, β_i the linear coefficients, β_{ii} the quadratic coefficients, β_{ij} the interaction coefficients, ε is the random error and Xi, Xj is the response of the variables.

3. Results and discussion

3.1. Landfill leachate

The leachate collected from the Municipal Waste Landfill No. 1 in the city of Mashhad, was used for all experiments. The physical and chemical properties of the samples were then measured. The average COD, five-day biochemical oxygen demand (BOD_5), and total suspended solid were 13800, 2100, and 2840 mg/L, respectively. Its pH was also equal to 7.8, which was observed as a dark brown color. Other leachate characteristics are reported in

Table S1. Of note, the changes in the landfill leachate characteristics of this study, as well as the difference with the leachate of other studies, including Zou *et al.* (2023), Cheng *et al.* (2020) and Lim *et al.* (2012), can be attributed to consumer nutrition in the months of the year and the lack of separation from the origin of waste (Pashaki *et al.*, 2021). As well, 71% of urban solid waste by weight in Iran is made up of perishable materials, of which only 5% is recycled on average (Rajaeifar *et al.*, 2015).

3.2. Coagulation-flocculation and sludge characteristics from treated landfill leachate

At first, a series of coagulation-flocculation experiments were performed under different operating conditions (namely, initial pH, virgin $FeCl_3$ dosage, and settling time) on the municipal waste landfill leachate. The removal efficiency of COD and total suspended solid was 52.55% and 84.98%, respectively, under optimal independent variables ($FeCl_3$ dosage=9.38 g/L, pH=8.94, settling time=52.9 min). The pollutant removal rate could be further related to hydrolyzed ferric ion (Fe^{3+}) species, reacting with hydroxide (OH), and then forming ferric oxyhydroxide [$Fe(OH)_3$] or ferrous hydroxide [$Fe(OH)_4^-$] (Ching *et al.*, 1994).

The sludge from treated landfill leachate obtained from the coagulation-flocculation process was used for the acid leaching process. The produced sludge from treated landfill leachate had pH, and total solid of 7.30 and 7.52% respectively. The sludge pH of this study was almost similar to Chen *et al.* (2012) and Ooi *et al.* (2018), who reported the pH of the collected sludge from treated landfill leachate as 7.3-8.8 and 7.4, respectively. Also, the solids content of sludge is reported as 4.2% in other studies (Ahmad *et al.*, 2016; Ahmad *et al.*, 2021). The Fe dosage found following the sludge from treated landfill leachate was 6.07% by weight, which could be due to the presence of Fe in the raw leachate samples and $FeCl_3$ coagulant used in the coagulation-flocculation process. Ahmad *et al.* (2021) and (Li *et al.*, 2010) reported the Fe dosage in sludge as 7.44% and 1.14%, respectively. This difference (solids content and Fe dosage in sludge from treated landfill leachate) with other studies can be attributed to the type and amount of coagulant used, the source of sludge collection, and the type of treated effluent. The elements in sludge from treated landfill leachate, using EDS, are given in Table S2 and Figure 2b.

The EDX elemental analysis also showed that carbon (C), oxygen (O), sodium (Na), silicon (Si), sulfur (S), chloride (Cl), potassium (K), calcium (Ca), P, and Fe were present in the sludge from treated landfill leachate. On the other hand, the elements, such as calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+), potassium (K^+), ammonium (NH_4^+), iron (Fe^{2+}), manganese (Mn^{2+}), chloride (Cl^-), sulfate (SO_4^{2-}), and hydrogen carbonate (HCO_3^-) ions could usually occur in leachate (Kjeldsen *et al.*, 2002). Therefore, it is concluded that most elements in the sludge production from treated landfill leachate are obtained from the original raw leachate. Similarly, the coagulation-flocculation process removes not only organic matter, but also other waste landfill leachate pollutants (such as metal ions), due to the absorption or sweep flocculation of precipitates (Shin *et al.*, 2008). The difference in the content of heavy metals in other studies could be consequently due to the variation in the quality of waste leachate from various landfill sites and the type of coagulant materials. The Fe recovery by the acid leaching process accordingly helps the coagulation-flocculation process. Fe_2O_3 also acts like the Fe-based coagulants, and then increases the COD and total suspended solid removal rates. Figure 2a shows the FE-SEM images of the floc containing $FeCl_3$ with 500-nm magnification. Here, the particles were observed in the form of irregular and uneven surfaces, which were strongly accumulated. These observations can be attributed to the accumulation of leachate pollutants (Teh *et al.*, 2014). Thus, it is established that such a phenomenon is achieved by two mechanisms, namely 1) the adsorption by bridging between particles and 2) charge neutralization as shown by Table S2 and Figure 2.

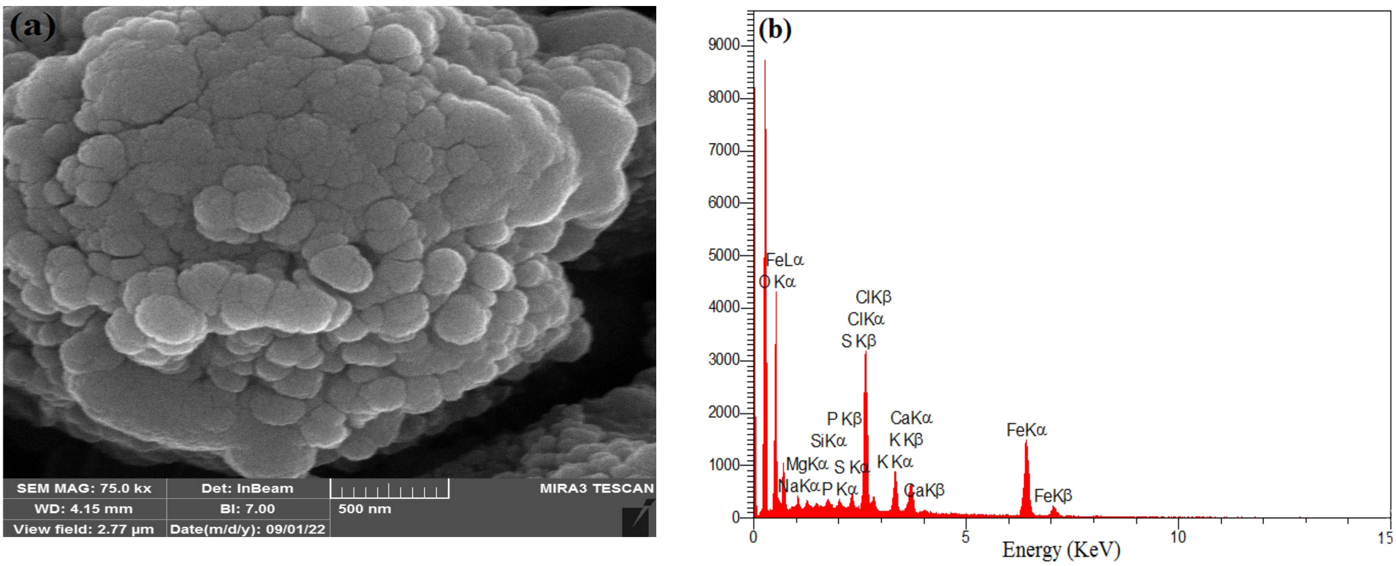

Fig. 2. FESEM images (a) and EDS analysis (b) of the precipitates of sludge from treated landfill leachate.

3.3. Assessment of experimental results and model fitting

The effect of two acids, H_2SO_4 and HCl, with different concentrations (i.e., 2, 3, and 4 M) on the acid leaching process of production sludge from treated landfill leachate was investigated. The results showed that HCl at a concentration of 4 M had greater Fe recovery than H_2SO_4. However, the amount of HCl required reaching 4 M concentration was 2 times higher than the amount required for 2 M concentration. Finally, H_2SO_4 was selected for the experiments in Phase 2 with respect to its marginal cost benefit and safer management than HCl (Chakraborty *et al.*, 2020).

The acid leaching process in Phase 2 was investigated during 20 experimental tests (including central and pivotal points in the experiment designed by central composite design) using the sludge from treated landfill leachate as a function of acid concentration (X_1), solid-to-liquid ratio (X_2), and heating time (X_3) for the Fe recovery (Y). The observed experimental response data (namely, the Fe recovery) in relation to the process variables of acid concentration, solid-to-liquid ratio and heating time are presented in Table S3. Based on the experimental values, regression models were developed using the quadratic model, which seems suitable for predicting the recovery of Fe from the sludge of treated landfill leachate (Equation 5).

$$Y \text{ (Fe recovery)} = 52.62 + 5.57\mathrm{X}_1 - 7.33\mathrm{X}_2 + 4.23\mathrm{X}_3 + 0.1816X_1X_2 + 0.568 - 3.70X_2X_3 - 3.55\mathrm{X}_1^2 + 1.86\mathrm{X}_2^2 - 1.20\mathrm{X}_3^2 \quad (5)$$

In Table S4, the ANOVA results for the model (Equation 5) are illustrated. The F-test showed that the Fe recovery model was significant at the 99% confidence interval. The LOF obtained in this study was not significant (P-value>0.05), which confirmed good fit data on the selected model (Muhamad *et al.*, 2013). The coefficient of variation, R^2 and adequate precision in this study was equal to 3.75%, 0.98 and 28.13, respectively. These indicated values the good repeatability, very high stability and confirmed that the model could be used to predict the Fe recovery. The significant condition of the model factors was then considered in order to achieve a proper fit. The significant factors of the Fe recovery included X_1, X_2, X_3, X_2X_3, X_1^2, X_2^2 and X_3^2 , but other factors were not significant. Three input factors of the process also had a significant effect on the output variable response. The positive sign in the equations implied the synergy between mutual interactions or individual effects, while the negative sign revealed the opposite effects (Kakoi *et al.*, 2017).

The plot of normality of the residuals and predicted versus actual values of the Fe recovery process also support the satisfactory level of the model (Figure 3). In particular, the probability that the residuals are normal near a line is shown in Figure 3a, whilst the distribution of the residuals also did not have a specific trend, as compared with the fitted values. In addition, the distribution of the residuals did not show any trends, such as sinusoidal changes in relation to the order of the experiments. Therefore, it was concluded that the residuals were normally distributed (Ahmad *et al.*, 2021; Nair & Ahammed, 2014), and the hypothesis of the normality of the Fe recovery data was accepted. The plot of the predicted values to the actual ones is depicted in Figure 3b, wherein the actual values are distributed next to the straight line, proving that the models are in good agreement with the measured values (Nair & Ahammed, 2015). These plots showed sufficient agreement between the real data and those extracted from the models. This adaptation is presented by the higher values of R^2 and adequate precision more than 4 (Table S4). According to Figure 3b, the quadratic model is statistically significant, and can be thus used to predict and optimize the independent variables of the central composite design.

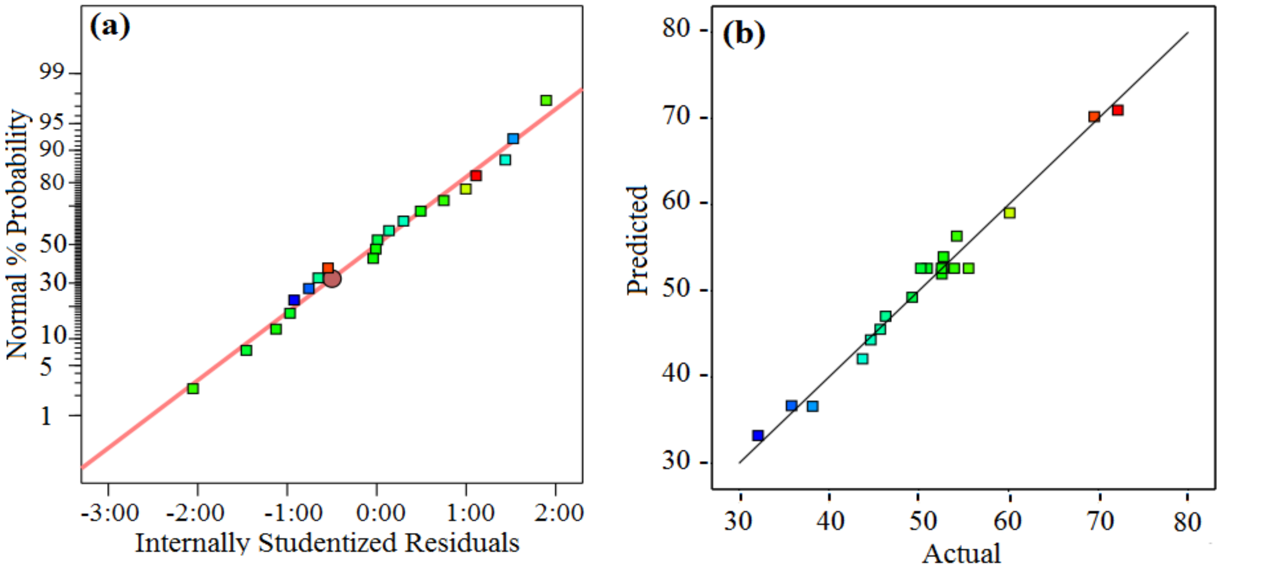

Fig. 3. Design-expert plot; a) Normal probability of studentized residuals and b) predicted vs. actual values plots for Fe recovery

3.4. Effect of operating parameters on coagulant recovery

The 3D contour plot (the model developed by the central composite design) to explain the fixed and interaction effects of the variables are shown in Figure S2. Each diagram displays the interaction effects of two independent variables, while the third independent variable is kept constant. In Figure S2a and b, the Fe recovery increases with the growth in the acid concentration from 2 to 4 M. By adding more acid, more Fe is then separated from the sludge from treated landfill leachate, and enters the solution, because the $Fe(OH)_4^-$ deposits are the dominant species in the sludge from treated landfill leachate. Therefore, the Fe recovery occurs more in higher acid concentrations. These results have been already observed in the reports by Li *et al.* (2005) and Nair and Ahammed (2014). Figure S2c and b show that the Fe recovery increases as the heating time rises. This ramping trend in the heating time boosts the Fe recovery in all acid concentrations and solid-to-liquid ratio. One of the reasons for the upward trend in the Fe recovery is the higher activity and reaction rate when the heating time is elevated. Our findings also comply with Fouad *et al.* (2017a) and Fouad *et al.* (2017b) who reported increased metals leached from sludge including Fe by increasing heating time. However, the effect of the heating time on the Fe recovery was stronger in higher acid concentrations than in the lower ones.

In Figure S2a and c, the Fe recovery has a downward trend following the growth in the solid-to-liquid ratio, where similarly to heating time, the effect on Fe recovery was again more prominent at higher acid concentration. Therefore, the Fe concentration in its solution increases, while the Fe recovery decreases with a higher solid-to-liquid ratio. Therefore, a lower solid to liquid ratio is achieved if the acid leaching process is to find the highest Fe recovery rate. Otherwise, it is suggested to use a higher solid to liquid ratio to gain a thick coagulant solution. This agrees with the literature reported that the recovery rate from sludge of treated landfill leachate increases by increasing acid concentration and the solid-to-liquid ratio (Fan *et al.*, 2013; Fouad *et al.*, 2017b). According to the elliptic shape of the contour lines with the long axis along the axis of the heating time in Figure S2a and c, the least and most effective variable in the Fe recovery is related to the heating time and the solid to liquid ratio. Our findings are also consistent with Nair and Ahammed (2014), who reported solid-to-liquid ratio as the most effective variable in recovery rate. Moreover, the perturbation plot in Figure S2d shows how the Fe recovery is affected by the input variables acid concentration, solid to liquid ratio and heating time. All three factor input factors affect the response following a quadratic behavior. Therefore, it is concluded that the effective variables are solid to liquid ratio, acid concentration, and heating time, respectively (in Table S4, F-value of solid to liquid ratio was higher than acid concentration and heating time in Fe recovery).

3.5. Fe recovery optimisation

First, a numerical optimization was applied. Then, with multiple responses provided by Design-Expert, the "sweet spot" for the variables was met. Additional experiments were performed to confirm the agreement between the model and the experimental results (Table 4). In general, the efficiency of the acid leaching process for the Fe recovery was relatively high and satisfactory. According to Table 4, the results of the response parameter in both experimental and predicted models were close to each other (error=0.92). The experimental value of the Fe recovery from the sludge of treated landfill leachate under optimal conditions was 70.12%, when the independent variables of acid concentration, solid-to-liquid ratio, and heating time were equal to 3.80 M, 8%, and 5 h, respectively. Therefore, the correctness of the operating conditions was established by calculating the error of the response variable (namely, the Fe recovery). The recovery rates of 53-97.5% of the sludge containing different coagulants in the acid leaching process have been so far reported (Jung *et al.*, 2016; Yazdani *et al.*, 2019). For example, Yazdani *et al.* (2019) found 53% recovery rate of Fe from the sludge of treated landfill leachate as a coagulant, which was much lower, compared with this study. However, the coagulant recovery rate here was slightly higher than the results in Ooi *et al.* (2018). On the other hand, Jung *et al.* (2016) recovered Al by 97.5% from water treatment plant sludge. This difference could be due to the discrepancy in the compositional characteristics of the sludge, prepared from various sources and using different methods. In this study, sludge was obtained from the waste landfill leachate treatment process during a series of coagulation-flocculation experiments. Chen *et al.* (2012) further recognized that factors, such as total suspended solid characteristics along with coagulant type and concentration could affect the recovery rate. The remarkable recovery in this study showed that the acid leaching process could effectively extract the metals in the sludge from treated landfill leachate.

Table 4. Observed Fe recovery at optimum conditions using sludge from treated landfill leachate.

Run	Conditions		Fe recovery (%)
1	Acid concentration (M) = 3.80	Experimental values	70.12
	Solid-to-liquid (%) = 8	Model response	71.04
	Heating time (h) = 5	Error	0.92

3.6. Impacts of ultrasound treatment time and performance of recovered coagulant

After the Fe recovery experiments, the optimal conditions of the acid leaching process were investigated with respect to the effects of ultrasound, as described in section 2.2.2. In general, the surface of the sludge solids from treated landfill leachate is exploded by liquid cavities, caused by ultrasound. Ultrasound also induce temperature and short-term pressures on the surface, create defects, and change the shape of the sludge surface. As a result, the material enters the solution from the solid surfaces of the sludge (Kim, 2016). Therefore, it increases the movement of ions from the surface of the oxide compounds to the liquid phase. Following this action, metal leaching from sludge of treated landfill leachate to solution augments during the acid leaching process. The possible mechanism may mainly contribute to the synergistic effects of ultrasound caused by the cavitation phenomenon, which is related to the presence of sludge particles that provide additional nuclei and thus the number of cavitation events (Yang *et al.*, 2013). At the same time, ultrasound can redouble the $Fe(OH)_3$ sedimentation (Xie *et al.*, 2009), which was observed in the experiments here. Comparing the Fe recovery values from the leachate treatment sludge in this study showed that the highest efficiency in the Fe recovery was assigned to Scenario 1 (without ultrasonic) (70.12%).

While the recovery rates for Scenarios 2 and 3 were 3.28% and 8.02% lower than Scenario 1, respectively. One of the reasons for no effect of ultrasound on the Fe recovery was the increase in $Fe(OH)_3$ dosage in the acid leaching process, which decreased the recovery rate (Barrera-Godinez *et al.*, 1992; Xie *et al.*, 2009). Therefore, ultrasound reduce the efficiency of the Fe washing from the sludge of treated landfill leachate to the solution.

The detailed analysis of heavy metals in the recovered solution under optimal conditions shows that metals such as Al (163.2 mg/L), Mn (46.7 mg/L), Mg (783.5 mg/L), and Si (2498.5 mg/L) in addition to Fe (15454.3 mg/L) are present in significant amounts. In other studies, the recovered solution content had been further reported to be different (Nair & Ahammed, 2014; Yazdani *et al.*, 2019). Yazdani *et al.* (2019), reported the Fe and Al amount in acid leaching solution as 8723 mg/L and 488 mg/L, respectively, which is lower than the values of this study (the sludge from treated landfill leachate used contained $FeCl_3$ coagulant). In another study, the amount of recovered iron from PACl and alum sludge was 99.3 mg/L and 187.0 mg/L, respectively. Also, 782.9 mg/L and 685.1 mg/L of Al were measured in acid leaching solution (Nair & Ahammed, 2014). This variance can be attributed to some factors, such as the type and amount of operating parameters in the acid leaching process and even the type and amount of coagulant materials in raw sludge. The presence of four measured metals would be thus useful because these elements can act as coagulants. However, high amounts of these elements indicate that recovered coagulant is unsuitable for being used in drinking water treatment, but for reuse in wastewater treatment (Chakraborty *et al.*, 2020; Ishikawa *et al.*, 2007). Accordingly, the recovered coagulant was employed to compare it with virgin $FeCl_3$ in landfill leachate treatment.

The recovered Fe could remove 60.21% and 91.40% of COD and total suspended solid during the coagulation-flocculation process, respectively. While these values for virgin $FeCl_3$ under optimal conditions (dosage = 9.38 g/L, initial pH = 8.94, settling time = 52.9 min) were equal to 52.55% (COD) and 84.98% (total suspended solids). This increase in removal efficiency could be due to the presence of Mn^{2+}, Mg^{2+}, Si^{2+}, and Al^{3+} in recovered coagulant, which together with Fe^{3+} could play a role in the coagulation-flocculation process (Xu *et al.*, 2009a), while only Fe could contribute to the coagulation-flocculation process in the virgin $FeCl_3$. Nair and Ahammed (2014) were also able to remove 71% of COD from the effluent of the up-flow anaerobic sludge blanket (UASB) reactor with the recovered coagulant. Similarly, Ayoub and Abdelfattah (2016) reported the recovered coagulant use, and the sludge volume reduction parameter for three different treatment plants was observed as 47-90%, which had a significant effect on minimizing sludge management costs. Therefore, the recycling back coagulants following recovery from leachate treatment sludge can significantly lower the amount of virgin coagulants for the wastewater treatment sector. As a result, the consumption of natural resources and fossil fuels to produce chemicals declines by reducing the costs related to the purchase of virgin coagulants (Nair & Ahammed, 2014).

4. Conclusions

This study established that the coagulant recovered from the sludge of treated landfill leachate could be effectively used as a replacement material of virgin equivalent coagulants in the coagulation-flocculation process by understanding the complex process of acid leaching. The following results were thus obtained using the central composite design:

- The coagulation-flocculation process with $FeCl_3$ as a virgin coagulant under optimal conditions (dosage=9.38 g/L, pH=8.94, settling time=52.9 min) could remove 55.52% of COD and 84.98% of total suspended solid from the municipal waste landfill leachate.
- The Fe recovery equal to 70.12% under optimal conditions showed that the correctness of the design of the experiments and the equations was confirmed, and the experimental results corresponded with the prediction (acid concentration=3.80 M, solid-to-liquid ratio=8%, and heating time=5 h) with an error of 0.92%. Among the variables, the solid-to-liquid ratio was the most effective in the Fe recovery by the acid leaching process.
- The developed experiments of the acid leaching process with ultrasound in different time intervals denoted that the highest amount in the Fe recovery was related to Scenario 1 (without ultrasonic) (70.12%). However, the recovery rates for Scenario 2 (ultrasound with 20-min settling time) and Scenario 3 (5-min ultrasound per 1/2 h of the acid leaching process with 20-min settling in the ultrasonic) were 3.28% and 8.02%, less than Scenario 1, respectively.
- The coagulation-flocculation experiments aimed at coagulant reuse indicated that the recovered Fe could remove 60.21% and 91.40% of COD and total suspended solid, respectively. Therefore, the quality of the treated leachate was comparable with the recovered coagulant and virgin $FeCl_3$.
- The continuous coagulant recovery and its reuse in waste landfill leachate treatment is a cost-effective and eco-friendly technique for sludge disposal from treated landfill leachate. In future studies, an additional quantitative cost-benefit analysis of

recovered coagulants versus virgin coagulants is needed. Plus, experimental trials testing different combinations of virgin and recovered coagulant must be undertaken to ascertain which split (virgin: recovered ratio) guaranties equal or comparable absorbing performance at each recycling iteration.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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