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STUDIES TO DETERMINE THE OPTIMAL COAGULANT DOSAGE FOR THE PRECIPITATION OF LANDFILL LEACHATE POLLUTANT

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Abstract. The treatment of landfill leachate is a critical environmental, social and economic issue. This type of wastewater poses a threat to the environment and human health, so European standards (Council Directive 1999/31/EC on landfill, Water Framework Directive) require its mandatory treatment. Landfill leachate is composed of different toxins and complex chemicals due to the presence of different kinds of pollutants and trash. These toxins and pollutants include heavy metals, organic pollutants, ammonium nitrogen, and xenobiotic compounds. Due to this complexity and variation in the chemical composition, conventional treatment methods such as bio-logical treatment and membrane separation often face various challenges. To address these challenges, the combination of treatment methods such as coagulation/flocculation as pretreatment followed by adsorption techniques has shown promising results in removing all the toxins at a much lower price using natural materials. This combination ensures the removal of suspended solids and colloidal matter trapped in the leachate using coagulation; the remaining dissolved organic and inorganic matter are removed by the adsorption stage. The uncertainties lie in the fact that it is not known exactly which coagulants, and at what doses, would optimally reduce the amount of organic matter in the landfill filtrate. Experimental studies are being conducted to select suitable coagulants. In this study, $Al_2(SO_4)_3$ and $Fe_2(SO_4)_3$ metal salt coagulants were selected for the coagulation stage. At the optimal dosages of $Al_2(SO_4)_3$, a maximum TC removal of 60% was achieved, whereas $Fe_2(SO_4)_3$ achieved a slightly higher value of about 62%. On the other hand, low TN removal efficiencies were achieved for both coagulants after the coagulation process alone, not exceeding 11%, confirming that coagulation process alone is insufficient for effective nitrogen removal. Subsequent adsorption stage significantly improved the treatment performance using ZMT, as overall TN removal exceeded 90% for both coagulants at the optimal doses, while the overall TC removal remains slightly effected.

Keywords: landfill leachate, treatment, coagulants, organic matter, adsorption.

1. Introduction

Landfill leachate is formed during the decomposition process in landfills and poses a significant environmental challenge. As water percolates through the waste accumulated in landfills, it dissolves and suspends the materials contained in the waste, forming a strong mixture called leachate (Mor & Ravindra, 2023).

The composition of the leachate can vary greatly, but it typically contains dissolved organic matter, inorganic macro-components (salts and metals), and various xenobiotic organic compounds (Kumar et al., 2025). Organic components can have harmful effects on water bodies, while inorganic macro-components (ammonia, chlorides, sulphates, and heavy metals) can cause toxicological problems (Al-Yaqout & Hamoda, 2020). The concentration of pollutants is significantly higher compared to typical domestic sewage by a significantly large margin. The uneven and high pollution content in the leachate compound requires an efficient leachate treatment method to be able to discharge and manage effluent effectively.

This effluent is mainly produced due to the overflow of the rainwater over the various layers in the solid wastes. The overall effect of microbial processes coupled with chemical and physical activities in landfills releases the contaminants present in the MSW (Municipal Solid Waste) in the percolating water, which comes out as heavily contaminated. The composition of landfill leachate can be clustered into four principal groups namely dissolved organic matter, inorganic macro-components, heavy metals, and xenobiotic organic compounds (Abdel-Shafy et al., 2024).

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Landfill leachate composition changes quite considerably depending on the age of the landfill, the types of waste, climatic conditions and operation practices (Abdel-Shafy & Mansour, 2018; Tchobanoglous et al., 2015). For example, leachates during early acid phase contain huge concentration of fatty volatile acids and other organic pollutants due to rapid breakdown. On the contrary, at the stage of methanogenic leachate, a more stable and comparatively lesser amount of leachate is recorded at this stage with low BOD/COD ratio and low heavy metal concentrations, however the concentration of ammonia does not decrease and continues to be a long-term pollutant (Abdel-Shafy et al., 2024). Landfill leachate characteristics and treatability based on its age shown in Table 1 below.

Table 1. Leachate characteristics and treatability based on the landfill's age (source: Sossou et al., 2024)

Parameters	Young	Medium	Old
Age	0.5–5	5–10	>10
pH	<6.5	6.5–7.5	>7.5
BOD/COD	0.5–1.0	0.1–0.5	<0.1
TOC	<0.3	0.3–0.5	>0.5
Kjeldahl nitrogen (g/L)	0.1–2	N/A	N/A
Heavy metals (mg/L)	>2	<2	<2
MH3-N (mg/L)	<400	–	>400
VFA/HFA	VFA (80%)	VFA (5–30%) + HFA	HFA (80%)

Due to this complexity and diversity of pollutants within landfill leachate, it is considered a major environmental concern as it is very difficult and unpredictable in terms of composition, requiring highly efficient treatment methods. Rainwater and precipitation infiltration as well as internal biological decomposition within the landfill produce a highly toxic effluent rich in ammoniacal nitrogen, heavy metals, and traces of emerging pollutants including pharmaceuticals and microplastics (Abdel-Shafy et al., 2024; Wang & Qiao, 2024). Its characteristics and diversity in chemical composition, regulated by the landfill's age, the composition of its waste, climate conditions, and operational factors, impose the necessity of efficient, reliable, and adaptive treatment methods (Wang & Qiao, 2024).

1.1. Methods for landfill leachate treatment

Physicochemical treatment methods such as chemical oxidation, coagulation, flocculation, and adsorption are often coupled with biological treatment methods as biological treatment process is more effective in treatment of young leachate in comparison to old leachate, alternative technologies and methods are needed for more mature leachate (Yu et al., 2022).

The principle of the coagulation method is the addition of some chemical substances called coagulants, which can be combined with suspended solids and pollutants in a solution. Coagulation is a complex process with a complex mechanism. Some of the commonly used coagulants include inorganic salts such as $FeCl_2$, $FeSO_4$, $AlCl_3$ and $FeCl_3$, these are being investigated as coagulants for the treatment of organic matter in landfill leachate. According to a study conducted by (Li et al., 2010), a coagulation-flocculation- adsorption mixing process was used to treat landfill leachate. The processing parameters were further optimized using response surface methodology. In the best case, the removal of 70% of COD, 93% of suspended solids, 97% of turbidity and 74% of toxicity was achieved. The results showed that coagulation-flocculation technology is an effective technology for the pretreatment of landfill leachate.

In general, coagulation processes happen in three simple steps, Rapid mixing, slow mixing or gentle agitation, and settling. The coagulant is added into the sample and agitated at high speeds for a few minutes, then the gentle agitation process begins to allow the flocs formation, then the final stage is settling where the sample is left for sedimentation to allow for the flocs to settle down at the bottom resulting in a clear solution. As an example, a study by (Daud et al., 2023) conducted coagulation/flocculation process using jar test apparatus following the same steps (rapid mixing, slow mixing, and settling), in this study alum was used as a coagulant. Rapid mixing was treated as a variable parameter between 100 rpm and 300 rpm to determine the optimal speed, slow mixing at 30 rpm for 30 min, and settling time 24 hours. Under the optimal parameters COD removal of 73.1% was achieved under rapid mixing of 100 rpm. The study of (Daud et al., 2023) goes into detail of the effect of different parameters on the coagulation efficiency such as rapid mixing rate, settling time, and pH.

Since the components of landfill leachate are quite complex, the selection of suitable coagulants is of great significance for proper treatment of landfill leachate (Ahmed et al., 2020). Additionally, the choice of other parameters during the coagulation process is also vital to achieve excellent removal efficiency in landfill leachate treatment. (Bai et al., 2023).

Adsorption process is considered as an efficient and primary approach in landfill leachate treatment and wastewater treatment in general (Somashekara & Mulky, 2023). One of the most characteristics that makes a good adsorbent is high porosity, which consequently leads to higher surface area thus a better and quicker pollution removal (Satyam & Patra, 2024). Generally, various materials were tested as adsorbents, including activated carbon, biochar, zeolite, clays, polymer-based adsorbents, and other porous inorganic or hybrid materials (Dehghani et al., 2023; Jagadeesh & Sundaram, 2023). The principle of the adsorption method is to adsorb pollutants in water on the surface or inside of

an adsorbent to achieve the solid-liquid separation of pollutants and water (He et al., 2023). Narendra et al. (2025) used MWCNTs as an adsorbent for the treatment of mature landfill leachate collected from a landfill site in Karnataka, India. The study achieved 65.66% COD removal through adsorption alone under optimal conditions of pH 2, a temperature of 55 degrees Celsius, and 60 minutes contact time. The adsorption method has many advantages including the low cost and most importantly its effectiveness in pollutants removal. This technique is widely used to the present day in the field of landfill leachate treatment and wastewater treatment in general and in the field of pollutant removal (Hidayu & Muda, 2016).

Recent studies indicate that adsorption and coagulation in combination enhance the overall treatment efficiency by enhancing the removal efficiencies of COD, ammoniacal nitrogen, heavy metals and colour. As an example, a pilot-scale hybrid system with adsorption as a pre-treatment step and electrocoagulation and biological treatment showed removal efficiencies of 95.5% of ammonium nitrogen, 98.8% of colour, and 85.7% of dissolved COD, which indicated the synergistic effect of adsorption and coagulation-based processes (Genethliou et al., 2023). The use of zeolite as an adsorbent in the adsorption step led to a dramatic increase in the removal of ammonium, which proves the effectiveness of natural adsorbents in combination systems. This combination processes methodology has been further advanced by innovative modified coagulants and adsorbents. A modified coagulant, polyaluminum chloride sulfate (PACS), was also shown to be very effective in removing suspended solids and color in membrane bioreactor effluent of landfill leachate, with 79.3% COD and 91.3% color removal during coagulation, which was further enhanced to 98.9% COD removal after sequential electrooxidation (Bayat et al., 2023). This underscores the benefits of using tailored coagulants in combination with adsorption and oxidation processes to achieve stringent discharge standards.

The nature and the dose of the coagulant play a significant role in achieving an efficient pollutant and color removal during the coagulation processes. According to a study by Narendra et al. (2025) using $FeCl_3$ as a primary coagulant, the dose of the coagulant used effects the removal efficiency of both COD and color. This is due to the enhanced charge neutralization and sweep floc formation while avoiding excessive restabilization of colloids. In this study, two metal salt coagulants, $Fe_2(SO_4)_3$ and $Al_2(SO_4)_3$ were tested on leachate collected form a single landfill located in Lithuania. The collected leachate was pretreated using electro-coagulation which reduced its COD to 5 g/L. This landfill leachate will be analyzed to determine its characteristics with focus on ammoniacal nitrogen (NH_4-N) concentrations before coagulation, after coagulation, and after adsorption. Although coagulation is not expected to reduce NH_4-N concentrations, adsorption is expected to reduce it significantly. The coagulation process in this study will be a preliminary visual test to determine approximate optimal coagulant doses before starting jar test apparatus and conducting further experiments by adjusting several key factors such as pH and the mixing conditions. All the samples studied with different coagulant dose will also be subject to adsorption process using an adsorbent consisting mainly of zeolite mining tailings (ZMT) grains to determine the adsorption effect on reducing NH_4-N concentrations. The preliminary coagulation process is expected to find the optimal coagulants dose and reduce color and turbidity of the leachate, while adsorption is expected to reduce the NH_4-N concentrations.

2. Methodology

2.1. Characteristics of the landfill leachate under investigation

The leachate under study was collected from a landfill located in Lithuania which was in use for 40 years and collected more than a million tons of waste during those years. The landfill is now closed and adapted for biogas production. The leachate from this old landfill is still a problem, as its contamination is higher than the regulated values for discharge into wastewater networks or into nature.

2.2. Coagulation experiments

Two coagulants were selected for preliminary studies for the determination of the optimal coagulants dose for the treatment of landfill leachate under study, $Fe_2(SO_4)_3$ and $Al_2(SO_4)_3$. According to scientific literature, these coagulants are metal salts which achieve between 50 and 70% of COD and color removal (Chen et al., 2019).

The goal of this visual test is to find coagulant doses achieving the best visual results, such as in color removal and low sludge production. Three different doses of each of the two coagulants were tested, resulting in six samples in total. Six samples of leachate were collected in measuring cylinders of 1 L, each containing 500 ml of leachate to conduct preliminary coagulation experiments. To each of these samples, a different coagulant dose was added.

Two coagulant stock solutions were prepared, one for each of the coagulants used. The stock solutions were prepared by adding 1.25 g of each coagulant to two separate volumetric flasks, then to each flask 0.25 L of distilled water was added to dissolve the coagulants, resulting in two stock solutions, each containing a dose of coagulant of 5 g/L. To start the test, different volumes of stock solutions were added to each sample, resulting in different coagulant doses in each sample. The doses used in this experiment, the volume of added coagulant stock solution, the coagulant dose in the sample, and the dosage ratio of coagulant to g of COD are shown in Table 2.

Table 2. $Al_2(SO_4)_3$ and $FeSO_4$ coagulants doses used in the preliminary coagulation experiment

Coagulant	Cylinder	Sample volume (ml)	Added Coagulant V(ml)	Coagulant dose in sample (g/L)
$Al_2(SO_4)_3$	1	500	50	0.5
	2	500	25	0.25
	3	500	100	1
$Fe_2(SO_4)_3$	4	500	50	0.5
	5	500	25	0.25
	6	500	100	1

Doses achieving the best results will be considered in future studies to determine the effect of other parameters such as pH in a coagulation test using a jar test apparatus.

Three different samples were collected for each treatment stage (raw leachate, samples after coagulation, and samples after adsorption), pH and NH_4-N were measured for all samples, and the reported results represent the average of the measured values. The COD of the leachate was previously measured and found to be 5 g/L.

After the addition of the coagulant stock solution, the mix was left to settle for 24 hours before starting the measurement of the parameters (NH_4-N). The samples were collected from the six cylinders using a plastic syringe 2 cm below the surface. All measurements were performed in 3 replicates, and their average was reported in Table 3 in the results part.

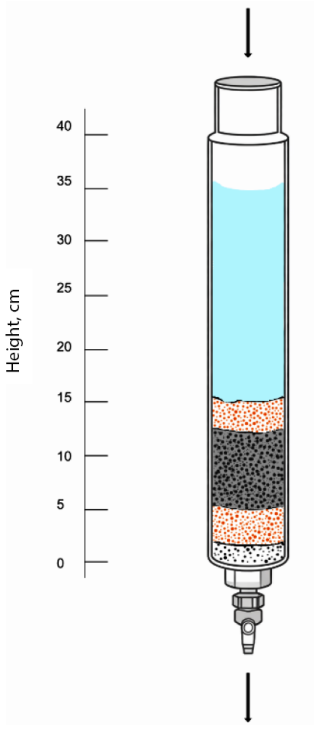


Figure 1. Filtration column. From bottom to top: pebble layer, quartz sand layer, ZMT layer, top quartz sand layer, layer of filtered liquid above the filler

2.3. Adsorption experiments

The adsorption experiment was performed by filtering diluted LL (landfill leachate) samples through a column (Figure 2). The supporting layer of the column consisted of small granite pebbles and a 3 cm high layer of quartz sand poured over them. Figure 1 illustrates the filtration column used during the adsorption process.

The main adsorbent layer of the column packing consisted of zeolite mining tailings (ZMT) grains (0.6–1.0 mm). ZMT was obtained from a Ukrainian Sokyrnytsya deposit and crushed by mechanical grinding, the fraction obtained by sieving the zeolite grains through sieves. The mass of this layer was 200 g. Two ZMT samples (100 g each) were used, the characteristics of which are given in the results section. These samples were selected after they had previously been tested for the removal of ammonium nitrogen from artificial wastewater. The NH_4-N removal efficiency then reached 85%, when the initial ammonium nitrogen concentration was 36 mg/L.

The ZMT layer was pressed from above with a 3 cm high layer of quartz sand. Filtration was carried out from top to bottom at a rate of 15 mL/min. The filtration rate was selected based on the authors' previous studies (Mažeikienė et al., 2021).

2.4. Analytical methods

The composition of the chemical elements in ZMT samples was determined using the X-ray fluorescence (WD-XRF) method. For this purpose, a ZSX Primus IV spectrometer (Rigaku Corp., Japan) equipped with an Rh tube with an anode voltage of 4 kV was used. For XRF analysis, samples were pressed into 37-mm diameter tablets (Januševičius et al., 2024).

FTIR – „Bruker Invenio R FT-IR" spectrometer + ATR diamond crystal + Nitrogen coling detectors.

Total carbon (TC) and Total nitrogen (TN) were also measured… Total carbon and nitrogen in 40 ml liquid samples were quantified using a SHIMADZU TOC-VC-SN system with TNM-1 and ASI-V modules. Nitrogen was converted to NO through high-temperature combustion (720 °C) and detected via ozone-induced luminescence, while carbon was oxidized at 680 °C and measured as CO_2 by infrared detection (SHIMADZU, n.d.)

All measurements were performed in 3 replicates.

3. Results and discussion

3.1. Results of coagulation experiments

The pH of the leachate was calculated in all stages of treatment and was found to be around a value of 8. After letting the mixture of all six cylinders settle for 24 hours, it can be observed that both coagulants ($Fe_2(SO_4)_3$ and $Al_2(SO_4)_3$) exhibited the same results in sludge production. The higher the volume of added stock

coagulant solution, the higher the amount of sludge produced. Regarding colour removal, it is observed that $Al_2(SO_4)_3$ achieved better results than $Fe_2(SO_4)_3$, and it is also observed that the flocs in the samples where the later coagulant was added were broken and incoherent, whereas the samples where $Al_2(SO_4)_3$ was used exhibited a lighter colour and a much more coherent flocs that sedimented at the bottom of the cylinder.

These results prove that $Al_2(SO_4)_3$ is suitable for further experiments using the same studies doses. Further studies reducing the pH are required as a pH value of 8 is on the higher side. Scientific literature proves that coagulation is efficient at slightly acidic pH values between 4–6. NH_4-N concentrations after the coagulation process are shown in Table 3 in the next section.

3.2. Results of adsorption experiments

NH_4-N concentrations were calculated before and after all the treatment stages (coagulation and adsorption). Table 3 below shows the recorded values.

Table 3. NH_4-N concentrations across all the landfill leachate treatment stages

Samples	RL	After coagulation	After adsorption
Avg (1)	1686.533	1716.187	22.240
Avg (2)	1686.533	2702.160	35.213
Avg (3)	1686.533	2068.320	38.920
Avg (4)	1686.533	1619.813	53.747
Avg (5)	1686.533	1816.267	259.467
Avg (6)	1686.533	1549.387	44.480

It is observed that the coagulation process did not reduce NH_4-N concentrations, in contrast, it is seen to increase in some samples. However, further treatment of the samples through the adsorption process significantly decreased NH_4-N concentrations, proving that the adsorption process is highly efficient for NH_4-N removal. Figure 4 shows a graph that compares the concentration of NH_4-N before coagulation, after coagulation, and after the adsorption processes.

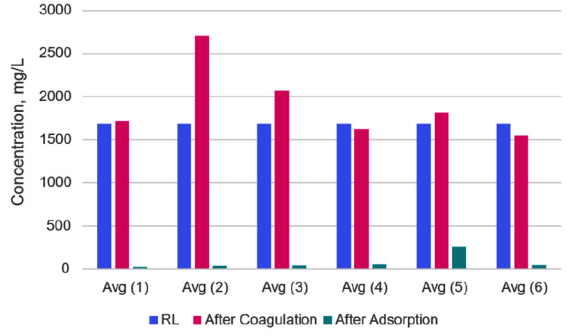


Figure 2. Concentrations of NH_4-N across all the treatment stages

Results of obtained Total carbon (TC) and Total nitrogen (TN) concentrations are shown in Table 4.

Table 4. Total carbon (TC) and Total nitrogen (TN) concentrations across all the treatment stages

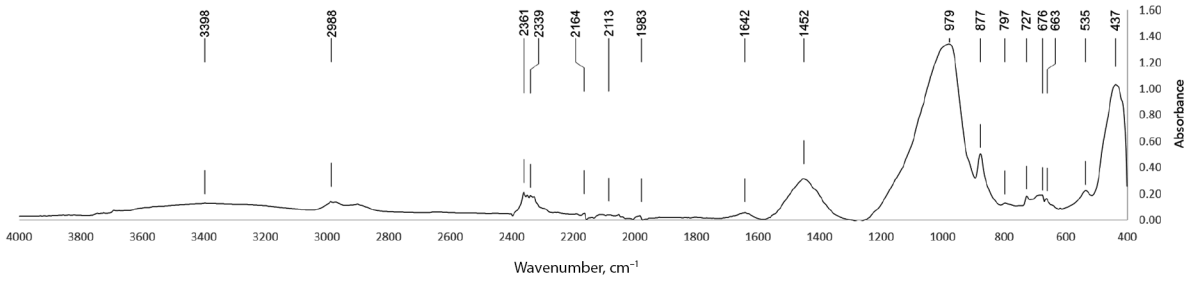
Treatment process	Sample	TC, mg/L	TN, mg/L
Untreated	RL1	905	1800
	RL2	896	1790
Coagulation	1	358	1607
	2	367	1650
	3	404	1565
	4	391	1571
	5	342	1640
	6	356	1598
Adsorption	ADS 1	420	154
	ADS 2	432	136
	ADS 3	398	131
	ADS 4	450	111
	ADS 5	411	146
	ADS 6	435	138

TC has decreased by 60% after coagulation process, and increased after the adsorption process, indicating carbon leaching or adsorbent contribution. TN decreased by 10.6% after the coagulation process, and inccreased by a significant percentage of 91.5% after the adsorption process, indicating an excellent TN removal. The hybrid process of coagulation-adsorption overall achieved a TC removal of 52.9% and a TN removal of 92.4%, indicating an efficient treatment method.

It is observed that coagulation and adsorption processes worked in synergistic effect. Coagulation process reduced color and suspended solids, whereas adsorption reduced dissolved chemicals (NH_4-N) proving that further treatment of landfill leachate through adsorption after coagulation process as pretreatment stage.

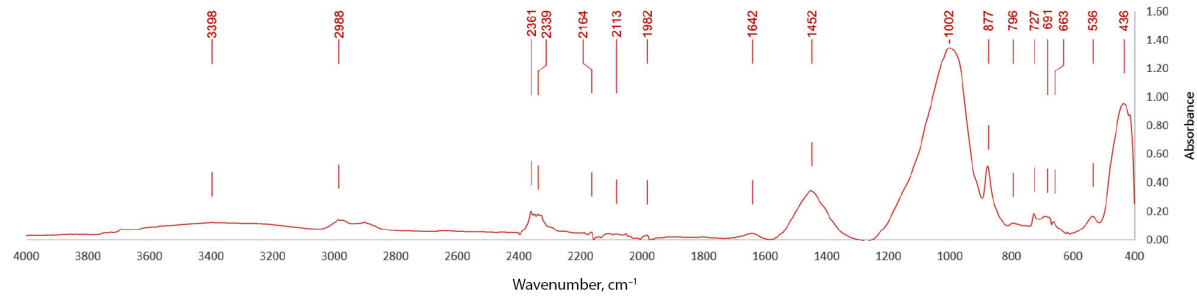
As for the analysis of the ZMT sample used as the adsorbent using the X-ray fluorescence (WD-XRF) method, two FTIR spectra were obtained. FTIR spectra data of ZMT and their main peak positions is given in Figures 3 and 4.

The results in Figures 5 and 6 show that the chemical structure of both ZMT samples is very similar. The FTIR spectra reveal the surface functional groups of the two ZMT adsorbents. The absorption band at ~3400 cm^{-1} is associated with stretching vibrations of –OH groups and adsorbed water, hydroxyl groups and adsorbed water are characteristic of clays and oxides. The band at ~1640 cm^{-1} is associated with H–O–H bending vibrations, which confirms the adsorbed water. The intense band around 1000–1030 cm^{-1} is assigned to Si–O–Si and Si–O–Al bonds, which are characteristic of clay minerals. In the lower wavenumber region (800–500 cm^{-1}),



Note: the x-axis is the wavenumber (cm^{-1}), and the y-axis is the absorption intensity.

Figure 3. Peak data of ZMT sample No. 1



Note: the x-axis is the wavenumber (cm^{-1}), and the y-axis is the absorption intensity.

Figure 4. Peak data of ZMT sample No. 2

metal–oxygen (Al–O, Mg–O, Fe–O) vibrations are observed, which are characteristic of oxides and silicates. The observed slight differences in intensity between the two ZMT samples can be attributed to different moisture content, unequal surface activity, and different degrees of adsorption.

X-ray fluorescence (XRF) is given in Figures 5 and 6.

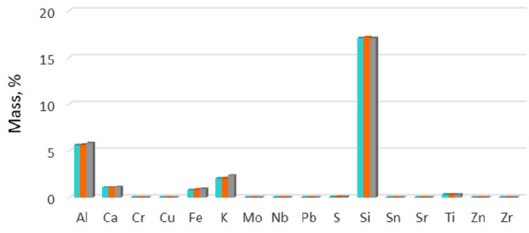


Figure 5. XRF data of ZMT sample No. 1

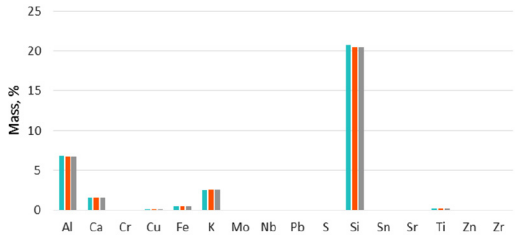


Figure 6. XRF data of ZMT sample No. 2

FTIR analysis reveals chemical bonds and functional groups, while XRF confirms the elemental composition. Both methods complement each other, proving that ZMT is suitable for use as an adsorbent. As can be seen from Figures 5 and 6, the elemental composition was dominated by Si, Al, K, Ca, Fe. Both samples also contained small amounts of Ti (0.24–0.3%), Cu (0.01–0.17%), and other elements (each of them accounted for less than 0.01%). Zeolite mining wastes, which contain copper (Cu), titanium (Ti) and iron (Fe), are a valuable resource for the development of functional materials. Zeolite wastes can be recycled into modified zeolites suitable for heavy metal removal and catalytic applications. The ion exchange properties of zeolites are exploited, and the adsorption is improved by the incorporation of metals such as Cu. Ti incorporation can be applied to achieve photocatalysis (Izidoro et al., 2019; de Carvalho Izidoro et al., 2024). The literature review shows that the mining tailings can be used as raw material to the synthesis of modified zeolites, which can be used for wastewater treatment.

4. Conclusions

This study has found preliminary optimal doses that can be further studied at different pH values. The study also found $Al_2(SO_4)_3$ to be more reliable than $Fe_2(SO_4)_3$ as the first exhibits better color removal and lower sludge production. These dosages of $Al_2(SO_4)_3$ can be further studied under different pH values using jar test apparatus. Other parameters such as mixing conditions and settling time can also be altered to find the optimal value of each parameter that is suitable for the landfill leachate under study.

Coagulation using $Al_2(SO_4)_3$ achieved better results than that using $Fe_2(SO_4)_3$ as the $Al_2(SO_4)_3$ achieved a better color removal, more coherent, and better-settling flocs at the tested doses, making it suitable for further optimization. The study also confirmed that coagulation alone was not enough to reduce NH_4-N concentrations, and in some cases even led to higher concentrations in some samples, achieving negative removal efficiencies of –1.76%, –60.22%, –22.64%, 3.96%, –7.69%, 8.13 for samples 1 to 6 respectively, whereas the subsequent adsorption process using ZMT adsorbent significantly decreased NH_4-N across all samples, achieving excellent removal efficiencies of 98.68%, 97.91%, 97.69%, 96.81%, 84.62%, 97.36% for samples 1 to 6 respectively. The hybrid treatment process achieved an overall TC removal of 52.9%, and an excellent TN removal of 92.4%, indicating an efficient treatment approach. These findings suggest that coagulation should be used as a pretreatment process for the removal of suspended and colloidal matter and improve the leachate's condition for the adsorption process that follows. The use of ZMT proved to be an effective adsorbent for the removal of NH_4-N, giving it a second life and demonstrating a potential reuse as a low-cost filtration medium.

Overall, the study provided an overview of the optimal doses of $Fe_2(SO_4)_3$ and $Al_2(SO_4)_3$ as coagulants for the treatment of the investigated landfill leachate and showed that the combination of coagulation and adsorption processes can be a promising option for its treatment. Future work will focus on optimization of pH and mixing conditions using jar test apparatus as well as the evaluation of long-term performance of ZMT adsorbent as a low-cost and natural adsorbent for landfill leachate treatment.

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