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Adsorption of Pollutants in Sanitary Landfill Leachate Using Granular Oyster Shell and Perlite

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Abstract

Landfill leachate, containing high concentrations of organic and inorganic compounds, can eutrophize aquatic systems and cause toxic effects on fauna. This study aimed to determine the optimum mix ratio for removing Chemical Osygen Demand (COD), Ammonia Nitrogen, and color using granular oyster shell and perlite as absorbent materials. The optimum dosage for Oyster Shell: Perlite was 25:15, with removal percentages of 49.67%, 64.09%, and 60.49%. The shaking speed and pH were also optimized with 58.83%, 82.76%, 76.60% and 54.72%, 50,20%, 68.97%. The outcome for shaking time was 57.90%, 70.47%, 80.43%. The adsorbent material effectively removed ±50% to 80% of removal COD, NH_3-N, and color.

1. Introduction

Sanitary landfills are a common method for disposing municipal solid waste, which can contain contaminants like heavy metals, organic substances, and nutrients. Adsorption is a promising method for removing pollutants from leachate, with natural materials like granular oyster shell and perlite being effective adsorbents (Daud et al., 2022). This study aims to investigate the use of granular oyster shell and perlite for removing pollutants from sanitary landfill leachate. The study will optimize conditions for adsorption using pH, temperature, and leachate concentration, and compare their effectiveness in removing different pollutants. Factors affecting adsorption capacity include the adsorbent's surface area and the presence of other ions in the leachate. The quality of landfill leachates is influenced by various factors, including age, precipitation, weather, waste type, and composition. As landfill age increases, leachate organics content declines and ammonia nitrogen concentration increase. This relationship can help select the best treatment method for old landfill leachates.

Table 1 *Landfill leachate classification vs age*

	Young	Medium	Old	References
Age (year)	<1	1-5	>5	Bikash & Sanjay, 2015
pH	<6.5	6.5-7.5	>7.5	
COD (g/L)	>15	3-15	<3	
BOD_5/COD	0.5-1	0.1-0.5	<0.1	
TOC/COD	<0.3	0.3-0.5	>0.5	
NH_3-N(mg/L)	<400	400	>400	
Heavy metals (mg/L)	>2	<2	<2	

Based on Table 1, the age of the landfill has a significant impact on the composition of landfill leachates. This study shows the three types of leachates have been identified with the composition. The leachate organics COD and ammoniacal nitrogen concentration increase as the landfill leachate increased. It can be concluded that the old landfill leachate is frequently heavily polluted with ammoniacal nitrogen because of the hydrolysis and fermentation of nitrogen-containing waste subtract component.

Leachate treatment technologies include chemical, physical, and biological methods. Yusoff et al. (2017) emphasize the importance of integrated treatment methods for direct release into surface water. Biological processes like activated sludge plants and aerated lagoons are widely used, but physical-chemical steps have been developed. The type of treatment should consider specific circumstances, expenses, and restrictions. This study using two types of adsorbents which is perlite and oyster shell. Zailani et., al (2018) study found that expanded perlite effectively removes metals from industrial waste leachate. It also has potential as an adsorbent for heavy metals and dyes from polluted water. The study confirms perlite's eco-friendliness and cost-effectiveness in wastewater treatment while oyster shells effectively adsorb heavy metals like lead, cadmium, and copper which is known as metals in wastewater treatment, increasing their adsorption capacity with longer contact times.

2. Methodology

This study used to determine optimal dosage for oyster shell and perlite in a Micropollutant Research Centre (MPRC) laboratory setting by optimum mix ratio. The leachate was collected from Simpang Renggam Landfill, Johor. A batch experiment was conducted to determine the optimal dosage, pH value, and contact time for removing pollutants from the leachate. The data collected was analyzed using statistical methods to determine if oyster shell and perlite together effectively work for leachate treatment. The results were used to calculate the optimum dosage, pH level, and contact time for the removal of pollutants from the leachate.

2.1 Area of Study

This research collected leachate samples from Simpang Renggam Landfill, one of Malaysia's largest landfills, located in Renggam District, Johor. The landfill, which has been in operation since the 1980s, receives waste from residential, commercial, and industrial areas. The leachate will be treated using an aerated technique at the 6-hectare landfill, which has been in operation for over 20 years and currently receives an average of 250 tonnes of solid waste daily from three different areas: Simpang Renggam, Batu Pahat, and Kluang. Figure 1 shows the location of collected leachate in the Simpang Renggam landfill site.

Fig. 1 *Simpang Renggam Landfill, Kluang, Johor*

2.2 Sample Preparation

The leachate sample from Simpang Renggam Landfill Site was stored according to the standard method of Examination of Water and Wastewater (APHA, AWWA, WEF, 2005). The raw leachate samples were obtained from Simpang Renggam Landfill Site using High-Density Polythene (HDPE) bottle. The sample of raw leachate was sent to the Universiti Tun Hussein Onn Malaysia (UTHM), Micro Pollutant Research Centre (MPRC) and kept at 4ºC in cold room for analysis.

2.3 Media Preparation

2.3.1 Perlite

Perlite was obtained from a supplier, crushed in a geotechnology laboratory, and filtered to less than 2.8mm size. It was crushed and washed with distilled water to remove small particles and stored in a dried room to prevent moisture adsorption. The adsorbent was mixed with granular oyster shell and leachate sample. Perlite was shown in the Figure 2.

Fig. 2 *Perlite after washed and used in the laboratory*

2.3.2 Oyster Shell

The laboratory experiment used granular oyster shell (GOS) collected from Malacca, cleaned to remove dirt and organic matter. The shell was soaked in a bleach and water solution for 30 minutes, then rinsed thoroughly. The oyster was dried and grinded into granular form and sieved with 2.8mm pan. The shell was sieved and stored in an airtight container to prevent moisture absorption. The shell was shown before and after grinding in Figure 3 and Figure 4.

Fig. 3 *Oyster shell after cleaning and before grinding*

Fig. 4 *Granular oyster shell after being grind*

2.4 Optimum Mix Ratio

The experiment involved separating a mixed ratio of perlite and oyster shell from a leachate sample, shaking it at 200rpm for 2 hours, and filtering it before analysis. The COD, NH_3-N, and color were determined using a HACH spectrophotometer, DR60000. The optimum dosage was determined by removing the highest percentage of NH_3-N, COD, and color from the sample. The amount of perlite and oyster shell used was shown in Table **2.**

Table 2 *Amount of oyster shell and perlite that are used to obtain optimum ratio*

No. Conical Flask	1	2	3	4	5	6	7	8	9
Perlite (g)	0.0	0.5	1.0	1.5	2.0	2.5	3.0	3.5	4.0
Oyster Shell (g)	4.0	3.5	3.0	2.5	2.0	1.5	1.0	0.5	0.0

The experiment was begun with 9 different conical flask with different ratio of Oyster Shell : Perlite. The study more focus on the finding suitable optimum ratio or dosage as a focused mark for optimum shaking time, optimum pH and optimum shaking speed.

2.5 Optimum Shaking Time

This study focuses on the removal of Chemical Oxygen Demand (COD), color, and Ammoniacal Nitrogen (NH_3-N) from leachate using an absorption process. The optimal shaking time is 150 minutes, with different time intervals of 30 minutes at 200rpm. The sample is then filtered and examined for COD, NH_3-N, and color. The average values are determined in triplets, and the percentage removal is computed using the initial reading. The experiment was conducted at room temperature and pH 7.

Table 3 *The time different for each conical flask*

No. Conical Flask	1	2	3	4	5
Hours	0.5	1.0	1.5	2.0	2.5

By referring table 3, sample of leachate were separate into 5 conical flasks with the highest removal percentage of dosage from optimum mix ratio.

2.6 Optimum Shaking Speed

This study used a 250ml conical flask with 100ml raw leachate treatment and an optimum mixing ratio with adsorbent media. The pH was set to 7, and the shaking speed varied from 50rpm to 250rpm for 120 minutes. The sample was filtered for contaminants NH_3-N, COD, and colo**r.**

Table 4 *The speed (rpm) different for determined optimum shaking speed*

No of Conical Flask	1	2	3	4	5
Speed (rpm)	50	100	150	200	250

The sample of leachate were separate into 5 conical flask as the table 4 for the determination of optimum shaking speed. This test was setup on the orbital shaker that was obtain in the MPRC laboratory.

2.7 Optimum pH

The study examined the impact of pH on the removal of NH_3-N, COD, and **c**olor in an adjusted pH leachate using concentrated sulfuric acid and sodium hydroxide. The sample was shaken at an optimum time and speed of 200rpm, with one set as the control sample. The procedure was repeated triplicate for an average value.

Table 5 *The different pH value used to determine the optimum pH*

No of Conical Flask	1	2	3	4	5	6
pH value	4	5	6	7	8	9

Each of sample leachate were adjust into different pH which is in range 4-9 pH as stated in table 5. The usage of sulphuric acid (H_2SO_4) and sodium hydroxide (NaOH) were used to adjust the range of pH. The initial pH for the raw leachate from SRL is 8.

2.8 Parameter Testing Laboratory

2.8.1 Chemical Oxygen Demand

This experiment was carried out with 100ml distilled water and 1ml control of landfill sample is filled in a small bottle that contain reagents COD digester. The solution being heated at temperature of 150^{o}C for 2 hours by using COD reactor. Next, the sample was placed and cooled at room temperature before taking reading by using DR6000. The precaution for this experiment is the test is repeat for 3 times for each sample to get average value that could be obtained by Standard Methods for the Examination of Water and Wastewater (APHA, 2005).

2.8.2 Ammoniacal Nitrogen

This test was conducted by 2 samples which are distilled water as a blank and 25ml of leachate sample in a separate tube. Both samples were dropped by 3 drops of mineral stabilizer and Polyvinyl Alcohol Dispersing Agent. 1ml of Nessler reagent will be added for both sample and being shaken. Next, 10ml of the sample was poured into the 10ml test tubes for analysis using the DR6000 Spectrophotometer. The reading is recorded 3 times as a precaution method.

2.8.3 Color

USEPA Method 8025 is a standard method for determining the color of leachate by using HACH DR6000. Firstly, 10ml of distilled water known as a blank and placed in the cell holder. The zero button is pressed to zeroing the reading of color. Next, 10ml of leachate sample is transferred to sample cell and placed into the cell holder. The leachate color of the sample is measured. The reading is recorded 3 times as a precaution method.

2.8.4 Data Analysis of Adsorption Efficiency

The percentage of removal density is determined by equation 1:

$$\text{Percentage of removal} = [(C_o - Cf)/C0] \times 100 \qquad \text{(Eq. 1)}$$

Where:

C_o= Initial concentration (mg/L)
Cf= Final concentration (mg/L)

2.9 Result and Discussion

In this section, the results of the experiments have been assessed using the parameters that have been set

2.10 Density of Adsorbent

The adsorbent was added to a 100 ml measuring cylinder that had been previously filled with 50 ml of distilled water, following the weighing of 10 g of granular oyster shell and perlite. The densities of perlite and oyster shells are displayed in Table 6, with oyster shell density being higher than perlite density.

Table 6 *Density of adsorbent*

Media	Density (mg/L)
Perlite	1.00
Oyster Shell	1.67

By referring table 6, oyster shell is denser than perlite. This means it has more mass per unit volume. This difference in density could be important for the adsorption in leachate treatment because of the natural porosity for each media as an adsorbent.

2.11 Optimum Mix Ratio (Oyster Shell: Perlite)

Figure 5 shows the data given for NH_3-N, COD, and color that obtained from the experiment optimum mix ratio. The data of removal percentage for each parameter was calculated from equation 1. Figure 5 shows how each dosage affects the leachate sample's COD, NH_3-N, and color characteristics. Based on all three criteria and the following figure, the highest ratio for this experiment is 25:15 of oyster shell with 64.09% removal of NH_3-N, 49.67% removal of COD, and 60.49% removal of color. In this experiment, the shaking speed was 200 rpm, the shaking duration was 120 minutes, and the pH remained neutral at 7. Once the ideal dosage was identified, the ideal shaking time, speed, and pH were all determined using this dosage. Consequently, the elimination percentage of each parameter dependent on dosage usage is shown in Figure 5.

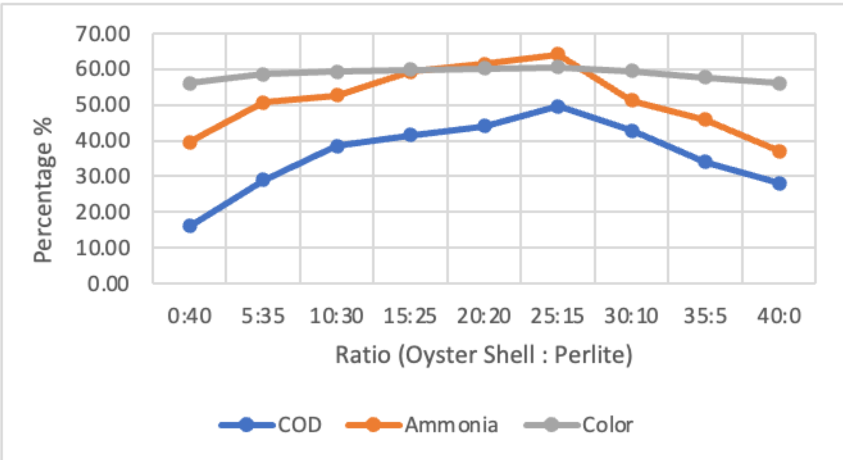

Fig. 5 *The graph with ratio and all parameters removal percentage*

2.12 Optimum Shaking Time

Figure 6 shows the data and the removal percentage for the highlight parameters which is COD, Ammoniacal Nitrogen and color. As the figure 5, the calculation of removal percentage was obtain from the equation 1. Figure 6 shows the line graph that highlights how shaking affects the removal percentage for every parameter that was investigated in leachate that was shaken for a duration of 30 to 120 minutes while treated with oyster shell and perlite. The figure will gradually stabilise after a while, and an increase in contact time does not necessarily translate into a rise in the removal percentage. The root of this is that adsorbate has been deposited on the adsorbent medium's accessible adsorption sites.

Consequently, Figure 6 shows that at 120 minutes, the optimum removal percentages for NH_3-N, COD, and color were 70.47%, 57.90%, and 80.43%, respectively. Between 30 and 90 minutes, the removal percentage growing dramatically; however, between 90 and 120 minutes, the percentages barely changed, suggesting that the removal percentage had reached equilibrium. Meanwhile, the total shaking time parameter with the highest potential for removal has been determined to be 120 minutes. Therefore, the results of this study, which included a 120-minute shaking period, might be applied to the next research into how to deal with leachate in media.

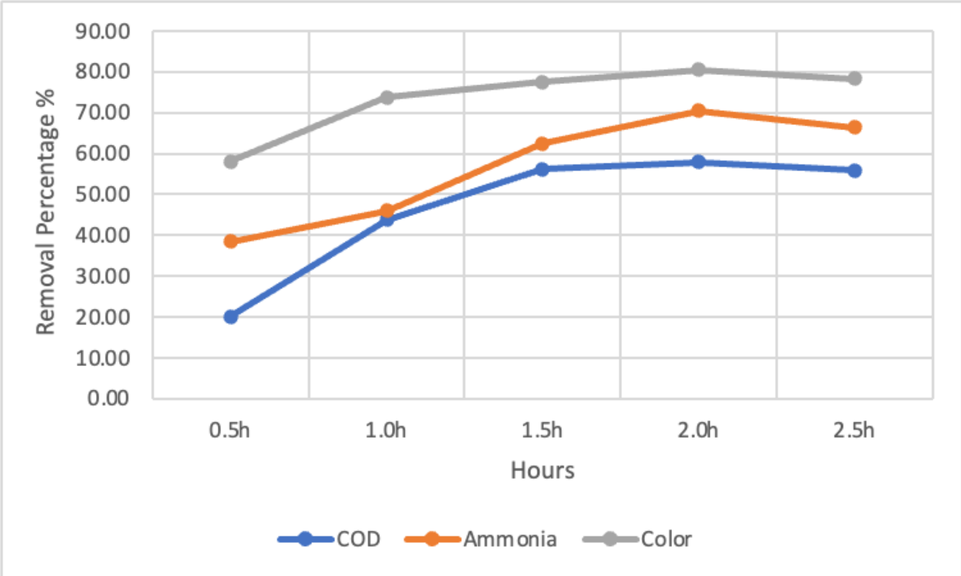

Fig. 6 *The graph for hours with all parameters removal percentage*

2.13 Optimum Shaking Speed

Figure 7 shows the COD, NH_3-N, and color removal percentages at different mixing speeds. As the speed increases, the removal percentage initially increases, reaching a maximum at 200 rpm. However, the removal percentage decreases slightly at 250rpm. Figure **7** shows the combination graph in removal percentage for COD, NH_3-N, and color in the leachate treatment. The 2**5**:**15** dosage utilised in this experiment was extracted from a prior study with a pH of 7.0 and a 120-minute shaking period. The shaking speed was available in four different speed ranges, from 50 rpm to 120 rpm. The highest removal of COD, color, and NH_3-N occurred at 200 rpm, with removal percentages of 82.76%, 58.83%, and 76.60%, respectively, as Figure **7** shows. Thus, the three factors that were assessed **which is** pH 7.0, 120 minutes of shaking time, and 200 rpm of shaking speed have the highest potential for removal.

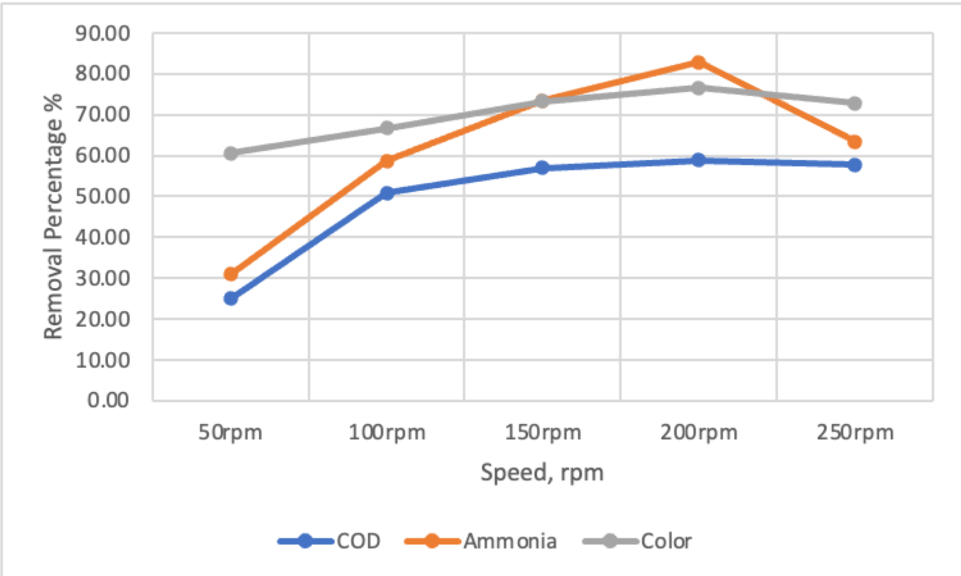

Fig. 7 *The graph relation between speed and percentage for all parameters*

2.14 Optimum pH

As the figure 8 given, it is shows that the removal percentage for each parameter increase until pH 7. It is possible that the higher pH levels are affecting the solubility of the pollutants, making them more difficult to remove from the wastewater. The high pH levels could be affecting the activity of the bacteria that are responsible for biodegradation of pollutants. However, the removal percentage for pH8 and pH 9 was slightly decrease as shown in the figure 8. Figure 8 displays the line graph showing the impact of the pH-adjusted removal percentage for all parameters examined in leachate treated with oyster shell and perlite (25:15) and agitated for 120 minutes. This information was gathered from an earlier study project. This investigation's pH was adjusted to range from 4.0 to 9.0, including acidic, neutral, and alkaline settings, in order to stabilise the leachate. Xing (2013) states that changing the pH will have the impact of making the nanoparticles more alkaline for the adsorption process. The hydroxyl radical and hydroxide precipitate that were created filled the active site in the nanoparticles and prevented additional activity. According to Figure 8 for this investigation, pH 7.0 resulted in the highest removal percentages for NH_3-N, COD, and color, which were 50.20%, 54.72%, and 75.01%, respectively. At pH 8.0 and 9.0, removal percentages dropped, supporting the earlier researcher's hypothesis that an alkaline atmosphere would inhibit adsorption activity.

As an outcome, after 120 minutes of shaking at pH 7.0, the parameter has the greatest total potential for removal. The findings of this study, which used a pH of 7.0 and a 120-minute shaking time, might be used in the following research to calculate the shaking speed in revolutions per minute (rpm). Figure 8 shows the percentage of each parameter that has been removed based on the optimal pH adjustment.

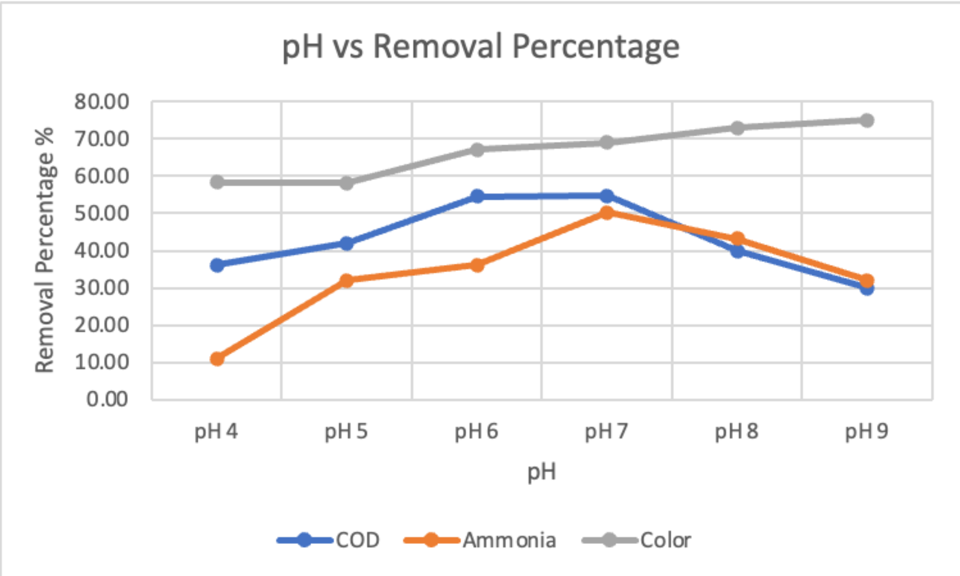

Fig. 8 *The graph pH relation with removal percentage for all parameters*

3. Conclusion

Leachate, a toxic liquid found at landfills, is composed of various chemical components obtained from soluble materials and byproducts of chemical and biological processes. This study aimed to determine the optimum dosage the selected adsorbent using activated carbon and oyster shell, focusing on parameters like chemical oxygen demand (COD), ammonia nitrogen, color, and pH value. The ideal ratio for removing high percentage parameters was found to be 25:15 (perlite: oyster shell), with the highest effective removal percentage at 200 rpm and an effective shaking time of 2.0 hours. The optimal pH was pH 7. The capacity of media to remove parameters can be increased by combining oyster shell with perlite as an adsorbent. The optimal proportion of oyster shell to perlite was 25:15, removing 49.67% of COD, 64.09% of ammonia nitrogen, and 60.49% of color. The greatest removal parameter was color, with an 80.43% removal percentage at the ideal shaking time of two hours. The optimal shaking speed was 200 rpm, resulting in maximum removal and removal percentages exceeding 50%.

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Conflict of Interest

The authors declare that there is no conflict of interests regarding the publication of the paper.

Author Contribution

The authors confirm contribution to the paper as follows: ***study conception and design:*** *Silahudin and Daud;* ***data collection:*** *Silahudin;* ***analysis and interpretation of results:*** *Silahudin and Daud;* ***draft manuscript preparation:*** *Silahudin and Daud. All authors reviewed the results and approved the final version of the manuscript.*

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