



Reduction of COD and ammoniacal nitrogen from landfill leachate using granular activated carbon and green mussel adsorbent

Amir Detho^a, Zawawi Daud^{b,*}, Mohd Arif Rosli^c, Mohd Baharudin Bin Ridzuan^a, Halizah Awang^d

^aEnergy & Environment Engineering Department, Quaid-e-Awam University of Engineering, Science and Technology, Nawabshah, Sindh, Pakistan, emails: amirdetho@gmail.com (A. Detho), mdbahar@uthm.edu.my (M.B. Bin Ridzuan)

^bFaculty of Civil Engineering and Built Environment, Universiti Tun Hussein Onn Malaysia, email: zawawi@uthm.edu.my

^cFaculty of Engineering Technology, Universiti Tun Hussein Onn Malaysia, email: mohdarif@uthm.edu.my

^dFaculty of Technical and Vocational Education, Universiti Tun Hussein Onn Malaysia, email: halizah@uthm.edu.my

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ABSTRACT

Landfills leachate always has been considered the utmost problem and is an issue in the management of municipal solid waste. Generally, landfill leachate contains a high concentration of organic and inorganic matter that caused an origin of hydro-geological pollution. In this research analysis, the combination of granular activated carbon (GAC) and green mussel (GM) was determined for the reduction of organic constituents' chemical oxygen demand (COD) and ammoniacal nitrogen (NH₃-N) from stabilized landfill leachate. This study was investigated to achieve the maximum optimum conditions, speed, time, dose and pH. The characterized result revealed that leachate contains a high concentration of biochemical oxygen demand (BOD₅)/COD ratio (0.09), COD (1,829 mg/L) and ammoniacal nitrogen (406.68 mg/L), respectively. The reduction result revealed that the maximum mixture ratios of media GAC:GM for COD and NH₃-N reduction are 2.0:2.0, 200 rpm, 120 min, and 57 g. The findings from isotherm adsorption analysis, the obtained result revealed that the Langmuir adsorption data better fit than Freundlich adsorption. The mixing media provides a strong result for the treatment of leachate wastewater and potentially used as economical good and sustainable adsorbent.

Keywords: Adsorption; Granular activated carbon; Green mussel; Isotherm; Optimum parameters

1. Introduction

Nowadays, population growth is leading to contribute to increase in commercial and industrial development in most countries in the world. Some research studies concluding that the growth of economic development and improvement in living standard, the increasing food and clothing consumption has affected growth in the amount of municipal solid waste (MSW) generations. MSW is a diversity at waste, derived from various origins (i.e., residential,

agriculture, municipal services, commercial sources), where each is heterogeneous in itself [1–4].

The composition amount of solid waste increasing with each area and approximately 95% of waste is directly dumped into the landfills [5]. Landfilling method is one of the utmost frequent and prominent options for the management and disposal of MSW worldwide. Landfilling is the well-known recognized method being an alternative option for both present and within the future, generally in low-income and middle-income countries, meanwhile this technique easier and inexpensive [1]. However, the

* Corresponding author.

generation of highly polluted leachate wastewater which being as a major disadvantage of that method [6].

Leachate wastewater is generally identified as the higher contaminated and the main source of hydro-geological pollution. Leachate normally produces from the decomposition of MSW. MSW in the landfill bodies changes to landfill leachate, gases, and ash. In some cases, rainwater percolates to the landfill and mixes with the leachate. Leachate is generally characterized by an elevated concentration of dissolved organic matter (chemical oxygen demand, COD), inorganic macro constituents, halogenated hydrocarbons, ammoniacal nitrogen, xenobiotic organic substances, suspended solids and a substantial concentration of heavy metals in addition to inorganic salts and chlorinated organic [7]. Leachate wastewater treatment is exceptionally complicated, costly and usually requires various processes [5].

Earlier, over the last 20 y, various research studies on landfill leachate have been conducted around the world. Leachate treatment uses a variety of individuals and/or combines physiochemical and/or combined biological and physicochemical treatment techniques [8].

Many technologies have been used for the treatment of landfill leachate over the past recent years namely ion exchange, reverse osmosis, chemical precipitation, oxidation, membrane filtration, air stripping, and adsorption. The landfill leachate treatment by using the physiochemical method has picked up significantly greater interest due to its economic value, short use of time and not producing a large amount of sludge respectively [5].

Recently several adsorbent materials have been studied as potential and relatively cost-effective namely zeolite [9] limestone [10] and palm ash [11], in the treatment of wastewater, especially adsorbent gain high attention due to its many advantages and interest [12].

Geographical material (i.e., sandstone, sand, and mineral) are being utilized as a low-cost adsorbent for the treatment of water and wastewater because of its abundance availability in local areas and environmentally safe material. Among this mineral, green mussel waste mostly abundantly available mineral in Malaysia which is formed from seafood industrial products and are considered to be as waste material and often abandoned dumpsite to naturally deteriorate. The abandoned natural source of green mussel waste is advantageous for utilize as the base material to produced the cheapest adsorbent for the treatment of water and wastewater [13].

Activated carbon (AC) also an efficient adsorbent due to its higher porosity surface and offers a wide surface contact area on which to pollutant adsorbed. AC is also accessible as granular activated carbon (GAC) and powdered activated carbon. These characteristics made AC an efficient but expensive adsorbent. The application of adsorbents based on marine polysaccharides has developed several technologies for solving the problem of cost [14–17]. Green mussel shell is a hydrophobic surface and primarily consists of approximately equal percentage ratio of calcium oxide (CaO) about (98.37%) [18].

The combining of natural material such as green mussel (GM) with activated carbon (AC), can serve as an appropriate alternative to utilizing AC as single medium and results in sustainable economic treatment. There has been significant research has been investigated conventionally

usage of media including the combination of AC and ZE to remediate stabilized leachate. The application of green mussel as a replacement of conventional media in the treatment of landfill leachate.

This article describes the findings on the utilizing of alternative bio-media for the treatment of leachate. A combination of mixed media was experimentally examined to analyzed the ideal mixture ratio, shaken time, shaken speed, pH and dose for GAC and GM for the treatment performance reduction of COD and ammoniacal nitrogen in stabilized leachate. Experiments were carried out by partial replacement of AC with GM.

2. Materials and methods

2.1. Sampling

The sampling of leachate wastewater was manually collected from Simpang Renggam Landfill Site in Johor, Malaysia. The area of research study found at latitude 10 54' 41.64" northern and longitude 1030 22' 34.68" eastern is in Kluang District, Johor, Malaysia. The landfills have also been operated by the government for many years and receive about 250 tons of solid waste every day, which handle three regions namely Kluang, Batu Pahat and Simpang Renggam [18,19]. Raw samples of leachate were obtained according to the method mentioned by the study of Isa et al. [11]. All chemical analyses for leachate characterization were performed within the following 24 h in accordance with the Standard Methods for the Examination of Water and Wastewater [21]. All chemical reagents utilized in characterizing leachate were of analytical determination.

2.2. Media

Many studies are conducted for the treatment of leachate wastewater by using different types of media, for this research analysis the mixing ratio of GAC and GM media were utilized for the treatment of landfill leachate. The commercially available adsorbent like coconut shell activated carbon were purchased locally from Cabot Malaysia Sdn Bhd, Malaysia, at about RM400 and RM4000 per ton, respectively. The green mussel shells (GM) were obtained from Ceria Maju Restaurant situated in Parit Raja, Johor. The green mussel shell was washed several times with deionized water to removing dust and fine particles. The shells were then dried up in an oven for 24 h at 105°C. The washed, dry and clean shells were crushed with an aggregate impact value machine and retain 150 µm particle size sieve. Both the adsorbents were crushed and sieve to values between 2.00 and 3.35 mm particle size [20]. Tables 1 and 2 illustrate the general properties of the media, that is, (activated coconut shell carbon – ACSC and GM).

Experiments were conducted at a predetermined optimal shaken speed and optimal mixing ratio, particularly after each pH adjustment. The optimal pH was selected based on pH which yields a maximum reduction of COD and ammoniacal nitrogen. To determine the optimal shaken time, the 250 mL volumetric glass containing 4 mL of predetermined mixed media (GAC:GM), and pH and 100 mL leachate were shaken at the fixed shaking speed on the orbital shaking machine. The volumetric glass was retrieved

Table 1
Chemical composition of granular activated carbon

Compound	Concentration (wt.%)
CaO	3.29
SiO ₂	72.3
Al ₂ O ₃	14.7
MgO	0.84
K ₂ O	4.64
Fe ₂ O ₃	2.72
TiO ₂	0.26
P ₂ O ₅	0.11
SO ₃	0.26
CH ₂	98.55
Bulk density (g/mL)	0.619

Table 2
Chemical composition of green mussel shell (GM)

Compound	Concentration (wt.%)
CaO	96.40
SiO ₂	0.44
Al ₂ O ₃	0.815
MgO	0.265
K ₂ O	0.375
Fe ₂ O ₃	0.315
TiO ₂	0.268
SO ₃	0.688
Na ₂ O	0.823
SO ₄	0.273
P ₂ O ₅	0.163
SrO	0.158
ZrO ₂	0.046
CaCO ₃	95.6
Bulk density (g/mL)	1.56

from the orbital shaking machine after each and every time interval 60, 120, 240, 300, and 360 respectively. Thereafter, the sample was filtered and analyzed by Daud et al. [20].

The optimal combination media were determined by varying the adsorbent ranges from 4 to 80 g respectively. The experiment was carried out at predetermined shaken speed, shaken time, mixing ratio, and pH. The optimal dosage was selected which yield the maximum reduction of COD and ammoniacal nitrogen. The adsorbent amount and removal (%) of COD and ammoniacal nitrogen per-unit mixing media were calculated with the following equations.

$$q_e = \frac{(C_0 - C_f)V}{m} \quad (1)$$

$$\text{Removal}(\%) = \frac{(C_0 - C_f)}{C_0} \times 100 \quad (2)$$

3. Result and discussion

3.1. Chemical analysis of leachate

Many research studies have been conducted and identified the various standard of landfill leachate collected from various landfill sites [15]. The results revealed that leachate contained a higher concentration of COD and ammoniacal nitrogen. The maximum concentration value of COD, ammoniacal nitrogen, pH, and biochemical oxygen demand (BOD₅) were 1,829, 406.68, 8.27, 163 mg/L and the ratio of (BOD₅/COD) of fresh leachate was 0.08 as shown in Table 3. The characterization result of COD and BOD₅ indicates that leachate was categorized as stabilized leachate. Generally, stabilized leachate has a higher level of COD and ammoniacal nitrogen (3,000 mg/L) and (>400 mg/L) and a lower concentration rate of BOD₅/COD ratio. The value of pH typically increasing over time, indicated decreased concentration amount of partial ionizes volatile fatty acid. Past studies work indicates that the stabilize leachate has a high concentration of pH is 7.5 [22]. The physicochemical properties of the composite adsorbent (GAC and GM) are presented in Table 4.

3.2. Comparative study of COD and ammoniacal nitrogen with other adsorbents

Mainly, in this research, the two different adsorbent materials such as GAC and GM are analyzed and compared with the other researcher. While from the analysis, the GAC and GM have shown better performance as compared to other adsorbent materials. Additionally, the overall removal percentage comparison of GAC and GM adsorbent is also included in this as listed in Table 5. From the table, it can be concluded that GAC and GM show better removal percentages in terms of COD and ammoniacal nitrogen as compared to others.

3.3. Determination of mixing media ratio

Fig. 1 shows the percentage reduction of COD and ammoniacal nitrogen within samples after combining GAC and GM in samples and realize optimal mixed ratio shaken for the reduction of COD and ammoniacal nitrogen. The most suitable condition was at 2.0:2.0 and 2.5:1.5. Both mixing ratios had COD reduction concentrations of 83% and 82%, respectively. Hence, the 2.5:1.5 mixed ratio produces a larger volume of GAC and is more costly than the 2.0:2.0 mixed ratio. The purpose of utilizing the alternative media to reduce costs and improve efficiency. Therefore, the mixed ratio 2.0:2.0 chosen was suitable as it implies less commercial mixed media ratio content. Fig. 2 exhibits a reduction of ammoniacal nitrogen at different mixed media ratios. The optimum reduction was (61%) at 2.0:2.0. The optimal condition was utilized for the furthermore experiment.

3.4. Determination of optimum shaking speed

The obtained mixed ratio of media (2.0:2.0 g) was used to applied optimal shaken speed determination, which plays an important role in the reduction of parameters. Figs. 3 and 4 illustrate that the reduction of COD and ammoniacal nitrogen decreases with increasing shaken

Table 3
Simpang Renggam Landfill Site leachate characteristic

Parameter	Leachate concentration	Soluble dissolved solids from the landfills by previous researchers			Malaysia leachate discharge standard, mg/L (Malaysia Environmental Quality Act, 1974)
		[32] ^a	[33] ^b	[34] ^c	
Reference	This study				
pH	8.27	8.26	7.60	8.29	6.0–9.0
SS (mg/L)	367	671	407	165	50
NH ₃ -N (mg/L)	406.68	541	868	1,890	5
COD (mg/L)	1,829	1,844	828	2,338	400
BOD ₅ (mg/L)	163	270	146	227	20
Color (Pt-Co)	4,788	4,721	3,199	4,527	100
BOD ₅ /COD	0.08	0.15	0.18	0.09	–
Fe (mg/L)	–	–	3.10	5.41	5.0

^aSimpang Renggam Landfill Site;

^bMatang Landfill Site, Perak, Malaysia;

^cPulau Burung Landfill Site, Palau Pinang, Malaysia.

Table 4
Physicochemical characteristics of composite adsorption media

Physiochemical properties of composite media	
Specific gravity (g/cm ³)	2.45
BET surface area (m ² /g)	95.95
Porosity (%)	57.40
Water absorption (%)	52.62
Methylene blue number (mg/g)	7.97
Iodine number (mg/g)	21.93
Cation exchange capacity (meq/g)	0.80

BET: Brunauer–Emmett–Teller.

speed rate within the ranges of 50–200 rpm respectively. These findings results can be attributed diffusion rate to the adsorbent in fluid bulk to surface of the adsorbent, it increases within the rate of shaken speed. In addition, the reason might be due to a reaction that may occur on the green mussel (GM) surface as a result of the attraction force between the ion presented in the fluid and the green mussel (GM) and the surface adsorbate of activated carbon. For further increasing the shaken speed more than 200 rpm, the reduction percentage of parameters decreases. This result may be due to decreasing adsorption vacant site and decreasing in the concentration gradient in the fluid between adsorbate and adsorbent. The optimal shaken speed was followed at 200 rpm for further experiments and the reduction percentage of COD and ammoniacal nitrogen was obtained (75%) and (65%), respectively.

3.5. Determination of optimum pH

The aqueous solutions of pH is a key function in the process of adsorption and influence the surface charging of ion surface group of adsorbent surface and adsorbate particle. The determination of the pH effect on the process of adsorption was examined, varying the value of pH solution ranges from 4 to 12 respectively. Figs. 5 and

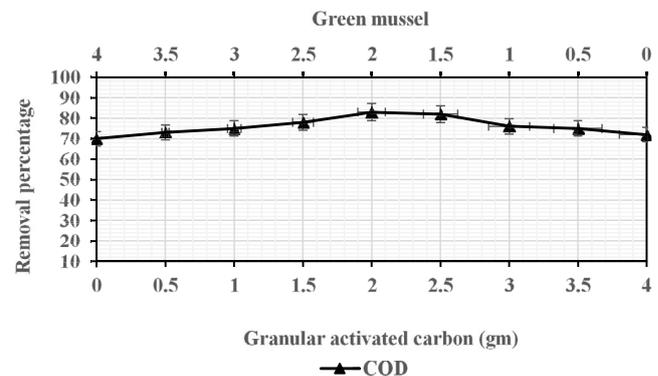


Fig. 1. Mixing ratio between ACSC and GM in COD reduction.

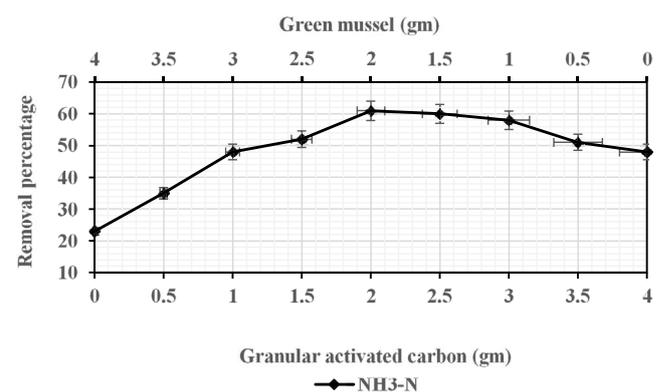


Fig. 2. Mixing ratio between ACSC and GM NH₃-N reduction.

6 illustrate that the reduction percentage rate of COD and ammoniacal nitrogen concentration at varying pH ranges. The adsorption efficiency rate clearly improved continuously with pH increasing until it reached 7, and decreases gradually until the pH value reached 12. Accordingly, the reduction percentage of COD decreased until the state of

Table 5
Performance comparison with various other adsorbent

Adsorbent	COD removal	Ammoniacal nitrogen removal	References
AC-cockle shell	31.98%	27.82%	[28]
Zeolite-feldspar	49%	45%	[29]
Green mussel	45.5%	–	[14]
Sugarcane bagasse activated carbon	83.61%	46.65%	[30]
Rice husk carbon composite	27.61%	51.0%	[31]
GAC-GM	83%	63%	This study

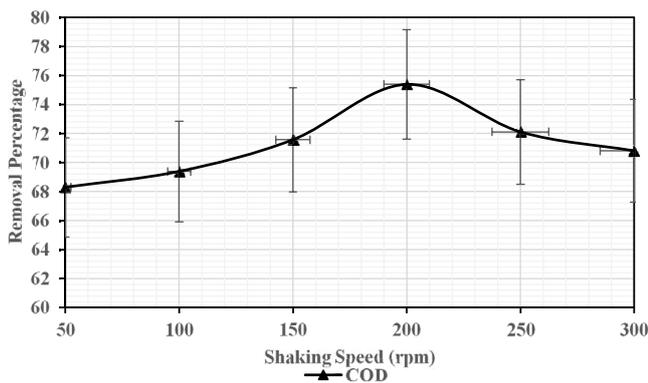


Fig. 3. Optimum shaking speed for ACSC and GM in COD reduction.

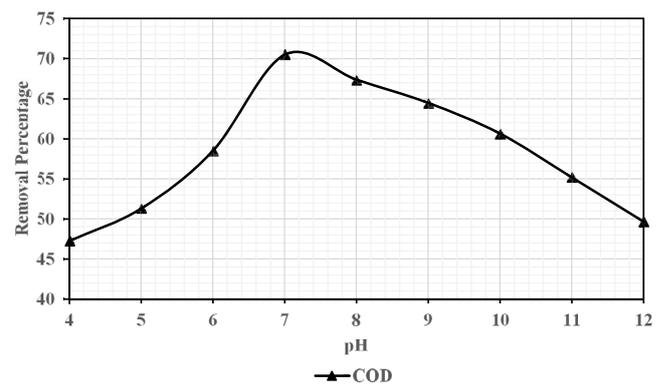


Fig. 5. Optimum pH for ACSC and GM in COD reduction.

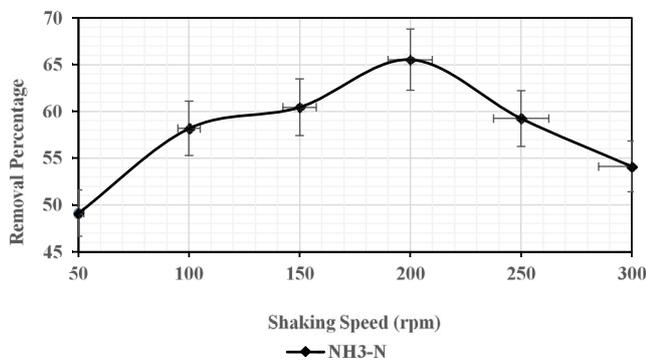


Fig. 4. Optimum shaking speed for ACSC and GM in NH₃-N reduction.

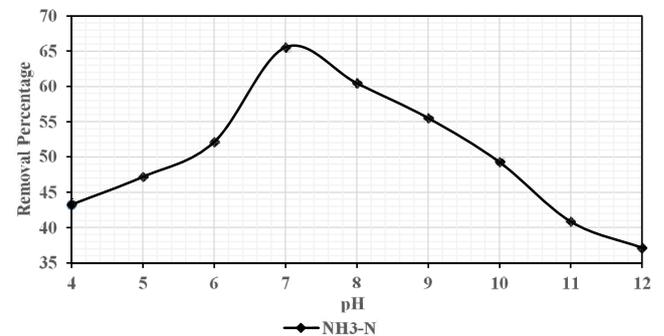


Fig. 6. Optimum pH for ACSC and GM in NH₃-N reduction.

equilibrium at pH 7. The results revealed that the optimal reduction rate of COD and ammoniacal nitrogen at pH 7 are 70% and 65%, respectively. The optimal pH solution was followed at pH7 for further experiment.

3.6. Determination of optimum contact time

The concentration amount of adsorbed species in bulk liquid and contact time interaction with the adsorbent surface and adsorbate has a vital role to play in minimization the toxins from the effluent. The time required to achieve optimum reduction efficiency when equilibrium achieved by sorption method. Figs. 7 and 8 illustrate that the effect of contact time with the adsorbent on the

reduction of COD and ammoniacal nitrogen. The reduction percentage of COD and ammoniacal nitrogen was increased with respect to time and attain equilibrium at about 120 min. The results revealed that the optimal reduction rate of COD and ammoniacal nitrogen at 120 min are 72% and 63%, respectively. The optimal contact time was followed at 120 min for further experiment.

3.7. Determination of optimum dosage

The determination of optimum adsorbent dosage of mixed media was investigated to attain economically saving and greater efficiency. The concentration effect of adsorbent on COD and ammoniacal nitrogen adsorption were determined by dose variations ranges from 3–80 g, at fixed optimum parameter (mixing media 2.0:2.0 g,

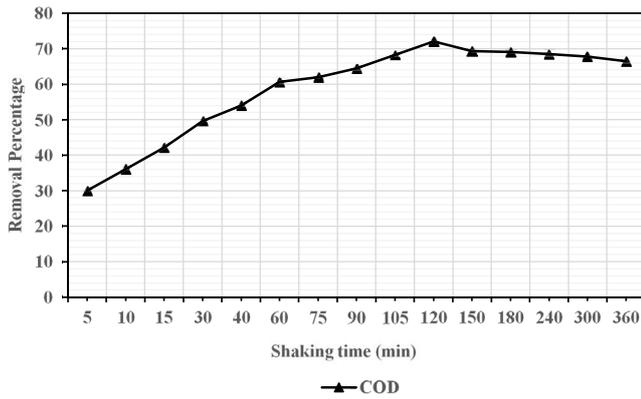


Fig. 7. Optimum contact time for ACSC and GM in COD reduction.

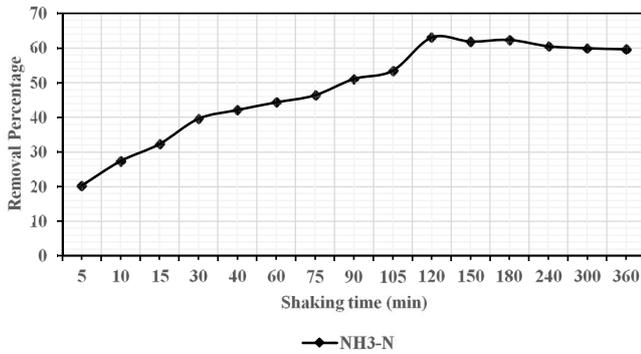


Fig. 8. Optimum contact time for ACSC and GM in NH₃-N reduction.

200 rpm shaken speed, 120 min shaken time, and pH 7). The results revealed that the reduction percentage increase with increasing the adsorbent dose till it attains the optimal mass of 57 g (55%); the reduction percentage then begins to decrease with further increasing the dosage of adsorbent (see Fig.). The reduction percentage rate of COD and ammoniacal nitrogen is about (88% and 82%), respectively. After that, the reduction percentage decreases as the dosage of adsorbent further increases as shown in Figs. 9 and 10.

3.8. Adsorption isotherm analysis

Isotherm adsorption evaluation is important for optimizing the sorption experiment. In this research analysis, two well-known adsorption isotherm models are utilized (Langmuir and Freundlich). The formation of the Langmuir adsorption isotherm model describes monolayers by adsorption on the surface of the adsorbent. This Langmuir isotherm model assumes that a finite (homogeneous) adsorption site exists on the surface of the adsorbent to which no additional adsorption will occur while that surface is completely occupied.

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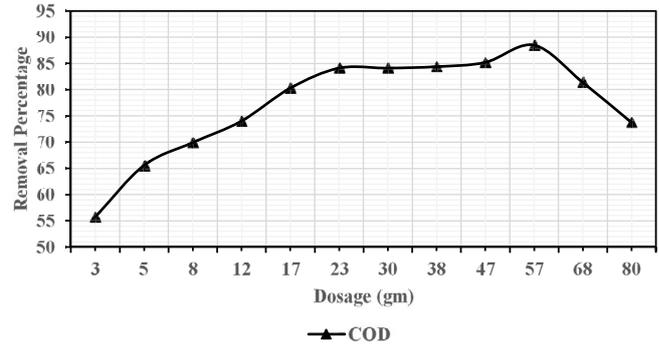


Fig. 9. Optimum dosage for ACSC and GM in COD reduction.

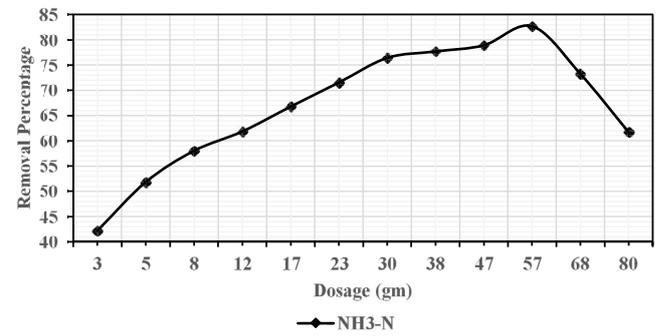


Fig. 10. Optimum dosage for ACSC and GM in NH₃-N reduction.

adsorption isotherm model describes monolayers by adsorption on the surface of the adsorbent. This Langmuir isotherm model assumes that a finite (homogeneous) adsorption site exists on the surface of the adsorbent to which no additional adsorption will occur while that surface is completely occupied.

On the other hand, Freundlich explains heterogeneity adsorption surface which surfaces adsorbent concentration increases within the initial solution concentration [23,24].

The Langmuir isotherm is represented by Eq. (3).

$$\frac{1}{q_e} = q_m K_L \frac{C_e}{1 + K_L C_e} \quad (3)$$

The linearized Langmuir equation is expressed as follows:

$$\frac{1}{q_e} = \frac{1}{q_m} + \left(\frac{1}{q_m K_L} \right) \left(\frac{1}{C_e} \right) \quad (4)$$

The maximum adsorbate amount (q_m) obtained within the given system can be determined from the isotherm. The linear Langmuir form of isotherm is reported as ($1/q_e$) vs. ($1/C_e$). The slope = $1/q_m K_L$ then slope = intercept/ K_L , then K_L = intercept/slope.

The Freundlich represented by Eq. (5).

$$\log q_e = \log K_f + \frac{1}{n} \log C_e \quad (5)$$

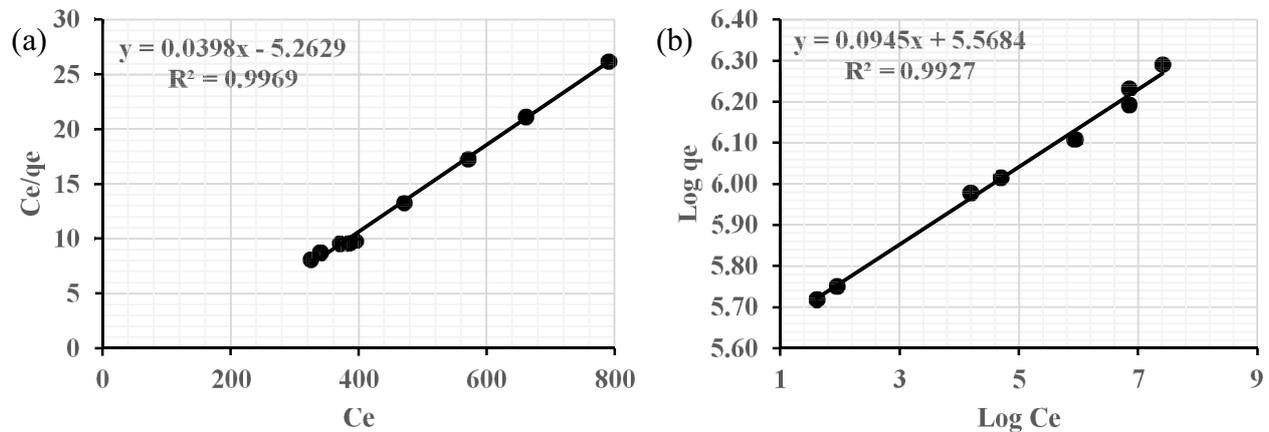


Fig. 11. Isotherm models for COD removal (a) Langmuir and (b) Freundlich.

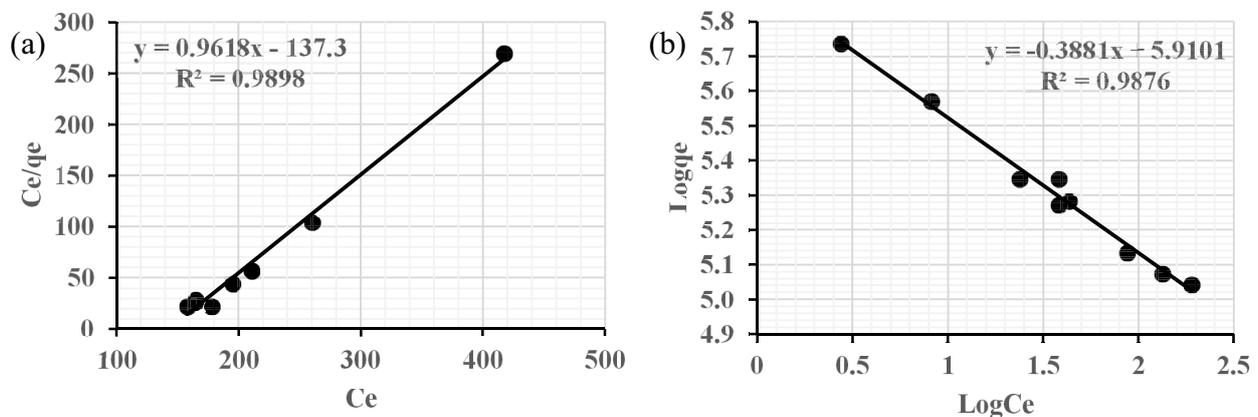


Fig. 12. Isotherm models for $\text{NH}_3\text{-N}$ removal (a) Langmuir and (b) Freundlich.

The linear Freundlich form of isotherm is reported as $\log C_e$ vs $\log q_e$. The slope and intercept are reported as $1/n$ and $\log K_f$.

Adsorption isotherm defines the adsorbent potential of adsorbent material and thus important research for the treatment of wastewater. Figs. 11 and 12 illustrate a reduction of COD for isotherms (Langmuir model and Freundlich model). Figs. 11 and 12 represent a percentage removal of ammoniacal nitrogen in both isotherms (Langmuir and Freundlich) using the optimal degree of media of adsorption parameter obtained. Table 4 shows the coefficient of correlation and constants from Eqs. (3)–(5). The exponent test $1/n$ shows the adsorption favorability.

The $(1/n)$ value approaches 1 shows adsorption is favorable, whereas the value of $(1/n)$ greater than 1 signify adsorption is unfavorable. The constants, (K_L) and (K_f) are related to adsorption energy magnitude and capacity of adsorption [25–27].

The coefficient of correlation (R^2) differentiates equation suitability to observe some results with other parameters. The value of correlation for COD obtained from the (Langmuir and Freundlich) equation illustrated that (R^2) value better fitted to Langmuir model as compared to Freundlich model. Whereas, the (R^2) value of ammoniacal nitrogen remained higher. The findings results

revealed that, contrary to the analysis by the study [26], the Langmuir model is better suited over the Freundlich model for determining equilibrium adsorption for COD and ammoniacal nitrogen using uptake by the mixing media.

4. Conclusions

For this research analysis, the potential of the GAC and GM adsorbent mixture was used to investigate the reduction percentage of organic constituents (COD) and ammoniacal nitrogen from stabilized leachate. The functional parameter which includes optimum mixed ratio, pH, shaken time, shaken speed, and dosage influence the reduction efficiency of adsorption mixing media. The optimum parameters were found as shown in mixed media ratio 2.0:2.0, shaken speed 200 rpm, shaken time 120 min, pH 7, and amount of dosage 32 g respectively. The isotherm study of adsorption shows the Langmuir model yields a better fitting experimental result when compared with the Freundlich model. The granular mixed adsorbent media material is being used as a better and cost-effective adsorbent for the treatment of stabilized leachate. The correlation value (R^2) was investigated in this present research work, which is <1 . The recent findings results contribute to add the existing literature growing interested

in alternatives to conventional media for the treatment of leachate. In addition, the research work is underway to evaluate other factors, including the desorption and disposal of exhaust adsorbent which affects the field of the research study.

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