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COD and ammoniacal nitrogen reduction from stabilized landfill leachate using carbon mineral composite adsorbent

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ABSTRACT

Leachate is a highly complex and polluted wastewater containing a high amount of dissolved and suspended matter produced by the introduction of percolation water through the body of the landfill. This problem can be solved by using the combination of granular activated carbon and zeolite as a filter medium. This research study is conducted to find an alternative treatment by combining low-cost adsorbent such as green mussel waste (*Perna viridis*) and ordinary adsorbent media, granular activated carbon, and zeolite. Both adsorption media were crushed and sieved to a particle size of 150 µm. Batch experiments were carried out to determine the optimal ratio of the adsorbent media. Granular activated carbon and green mussel have been classified as hydrophobic media whereas the optimal ratio was 2.5:1.5. Zeolite has no combination and is considered as hydrophilic media whereas the optimal ratio was 1.0. The best ratio for hydrophobic and hydrophilic media ratio have been selected as 7:3, according to the behavior of adsorption of organic constitutes (chemical oxygen demand (COD)) and ammoniacal nitrogen to the media. The batch experiment results indicate that the leachate concentration of COD was 310 mg/L with reduction percentage of 83% and ammonia nitrogen was 150 mg/L with reduction percentage of 63%. The optimum condition for reduction of ammonia nitrogen and COD were found with 200 rpm in shaking speed, 120 min of contact time at pH 7. The experimental result shows that both the models Langmuir and Freundlich isotherms were best fitted and favorable; that is adsorption phase reached equilibrium. According to regression coefficients (R^2), Langmuir isotherms were best fitted for COD reduction, and Freundlich isotherm was best fitted for ammoniacal nitrogen reduction. Langmuir and Freundlich isotherm adsorption capacity for COD and ammonia nitrogen were 0.9971 and 0.9914, respectively.

Keywords: Composite adsorbent; Landfill leachate treatment; Stabilized landfill leachate; Adsorption; Isotherm

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1. Introduction

Landfills is the most widely recognized techniques in many places around the world to manage waste disposal. It is the technique that is the most cost-effective and environmentally appropriate method for reducing and disposing-off municipal solid waste (MSW) and industrial solid waste [1,2]. Landfill leachate can typically contain high amount of organic matter, can be measured as organic constitutes (chemical oxygen demand (COD)), biological oxygen demand (BOD), ammonia nitrogen (NH_3–N), suspended solids (SS), heavy metals, and inorganic salt [3–5]. When a highly contaminated landfill leachate is released directly into the atmosphere or in water bodies, landfills become the main sources of contamination of the groundwater and surface water, which can percolates into soil and subsoil, seriously threatening public health, and affecting the ecosystem in aquatic life [6,7]. Most of the landfills do not have treatment facilities for leachate. Landfills from this unsuitable landfill design will contaminate our environment, particularly the groundwater and surface. Organic constitutes (COD) and ammonia nitrogen have been the two main problematic parameters in the treatment of landfill leachate. It has been seen that biological treatment of landfill leachates is very effective in the early stage reduction of organic matter [8]. When the leachate ratio of BOD_5/COD is high. With the age of the landfill, this ratio decreases [9] and over time, this process becomes less effective [10], because of the large presence of organic matter. The leachate characteristics are classified and identified as young or old leachate from the factor stated. Some methods/treatments, such as physical, chemical, and biological can be used to treat leachate. Generally, the concentration of BOD in young leachate is relatively high and typically biological treatment method is utilized for the treatment of leachate because it is effective in removing high concentration of BOD [11].

Some research works reported that the efficiency of treatment was improved with use of physico–chemical treatment alongside biological method for old leachate treatment. This techniques was widely used because of its ability to remove difficult and non-biodegradeable substances namely, PCBs, AOXs, humic acid, fulvic acid, or heavy metals from leachate [12]. In general, the young (<1 y), intermediate (1–5 y), and stabilize (>5 y) leachate ratios of BOD_5/COD were recorded as 0.5–1.0, 0.1–0.55, and <0.1, respectively [13].

Due to cost-effectiveness, in recent years, greater attention has been focused on preparation of low-cost adsorbent as substitute or partially reducing the consumption of activated carbon. Advances in science and technology have demanded a continuous searching for an effective and affordable adsorbent. Carbon mineral composite adsorbent is considered as a new form of adsorbents of different structure and adsorption properties from each component [15,16]. Composite materials have been developed to improve the adsorption property or to produce low-cost adsorbents [17]. Mostly activated carbon is considered as the best adsorbent in removing the organic pollutants from gaseous or aqueous phases. Activated carbon adsorbent is most commonly used for purifying the air and water [15,16]. The result shows the effect of adsorption of organic matter by activated carbons above the zeolites. Zeolites may reduce or minimize peak value of ammoniacal nitrogen value of influent, whereas activated carbons gradually keep removing ammoniacal nitrogen. The combination of zeolite and activated carbon composite is effectively used for removing ammoniacal nitrogen and organic matter in micro pollutant raw water [18]. Gao et al. [19] found new zeolite–carbon (Z–C) composite materials combining great zeolite and carbons property. The surface of zeolites is a hydrophilic with a frequent alignment of molecular and cation exchange potential with micropores, making it a good adsorbent ion and catalyst [20].

Mainly, in this research, three different adsorbent materials such as activated coconut shell carbon, green mussel (*Perna viridis*) and zeolite are analyzed The reduction trend of COD and ammoniacal nitrogen on the ratio of mixed hydrophobic adsorption ACSC and GM (2.5:1.5 g) and hydrophilic adsorption ZE (1 g) are shown in Figs. 1 and 2. From the result obtained from Figs. 1 and 2, the reduction rates of COD from dissolved solids with their maximum values were 83% compared with ammoniacal nitrogen 63% respectively. The reduction of COD and ammoniacal nitrogen obtained in this study is better than the study done by Halim et al. [33] at the ratio of 2.0:2.0 g (activated carbon-rice husk carbon) and 30:10 g (zeolite-limestone) with the reduction of both parameters, respectively.

Adsorption or physicochemical technique is relatively simple and might be utilized effectively for the treatment of stabilized landfill leachates. Adsorption is basically a process of mass transferring from a substance state of liquid phase to the solid surfaces, becoming bounded physical–chemical interaction. Nowadays, increasing more focus on use of low cost material namely natural or agriculture waste, industrial process by-products, to achieve proper landfill leachate treatment, or due to local availability and environmental friendly material, conventional adsorbent is an alternative approach for the treatment of water and wastewater [13].

In this study, Shehzad et al. [34] found that the production of activated carbon from the food waste is significantly given importance from economic and environmental points of view. Therefore, the use of activated carbon that is derived from food waste must reduce the amount of food waste disposal to the landfills. The focus of this study is not only on the preparation of activated carbon, but also to investigate the reduction efficiencies and optimum adsorption capacity performance of preparing activated carbon in the elimination of contaminants such as COD, color, ammonical nitrogen, and heavy metals from the leachate wastewater. The author conclusively suggested that the food waste is potentially useful and effective tool in the area of adsorption, leading to greater pollution control, and conservation of the environment.

In a study, Abuabdou et al. [35] provides a detailed evaluation of the applicability of anaerobic membrane bioreactors treatment (AnMBR) of landfill leachate wastewater, addressing its effectiveness, disadvantages, and potential prospects. The main concerns were treatment efficiency, recovery of biogas, and membrane fouling control. Generally, AnMBR method has several advantages when compared to other systems of anaerobic and aerobic treatment. AnMBR is really a popular leachate wastewater

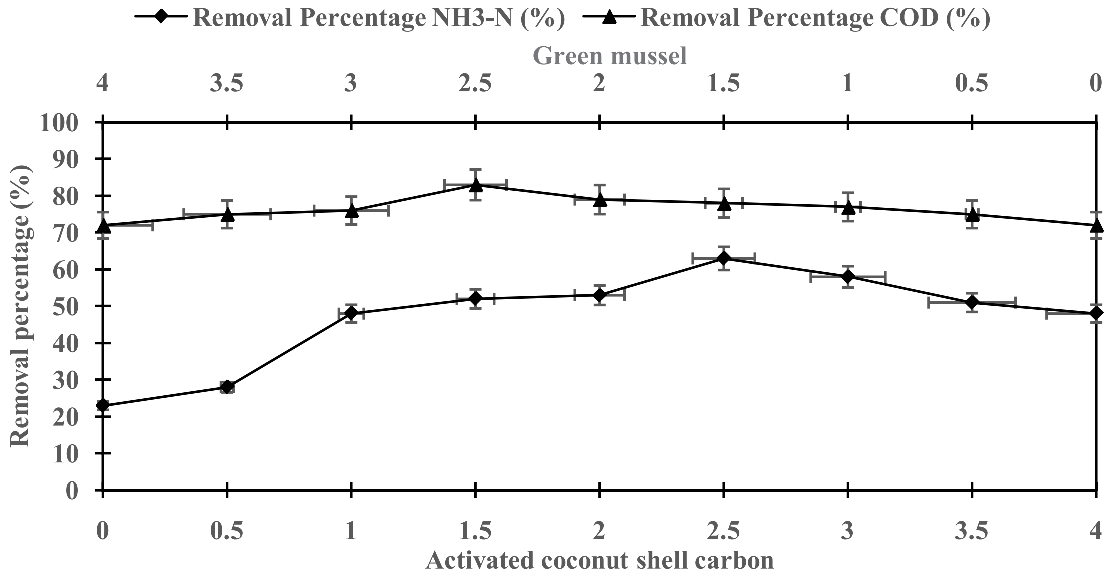

Fig. 1. Optimal ratio of hydrophobic adsorbent (activated coconut shell carbon to green mussel) based on COD and NH_3–N adsorption (100 mL of solubility/leachate, shaking speed = 200 rpm, shaking time = 120 min, particle size <150 µm, and pH = 7).

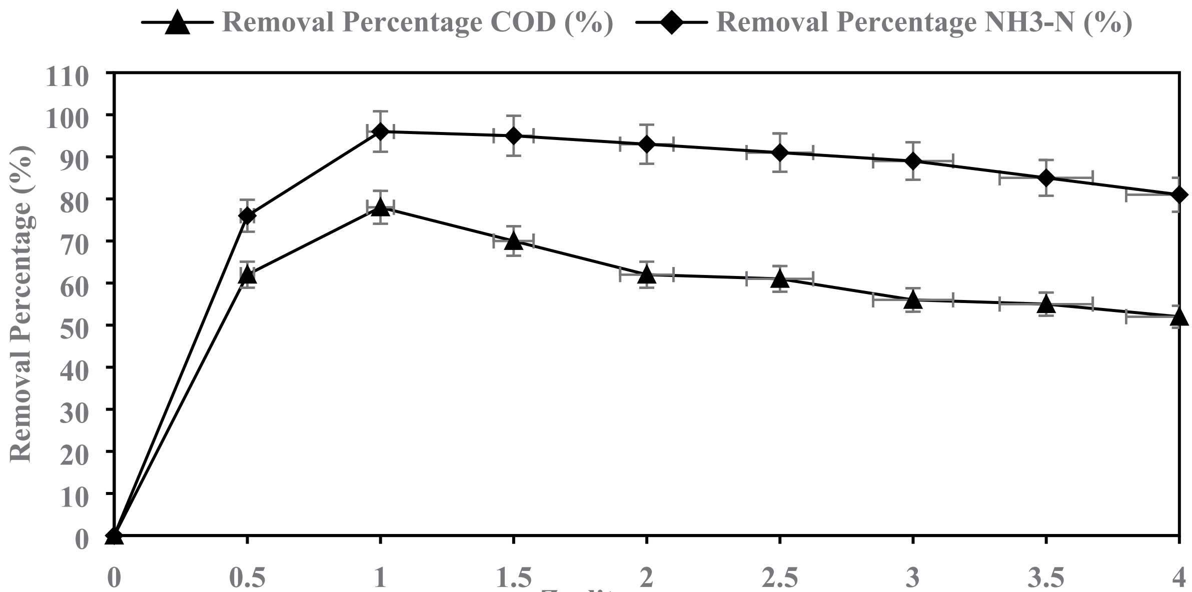

Fig. 2. Optimal ratio of hydrophilic adsorbent (zeolite) based on COD and NH_3–N adsorption (100 mL of solubility/leachate, shaking speed = 200 rpm, shaking time = 120 min, particle size <150 µm, and pH = 7).

treatment method; due to its major advantages over other conventional anaerobic and aerobic techniques. A critical analysis regarding the performance of a laboratory scale AnMBR on landfill leachate with production of biogas is also included in the above mentioned research study.

The aim of this research study was focused on the use of new composite material for the treatment of leachate. The leachate sampling was collected from Simpang Renggam landfills site (SRLS) according to the procedure described in Standard Method water and wastewater examination [21]. Previous studies indicate that the concentration of organic constitutes (COD) and ammonia nitrogen (NH_3–N) was upto 2,739.06 and 1,765.34 mg/L and the ratio of BOD/COD was as low as 0.06–0.12, respectively [13]. In this study beside the combination of activated carbon and zeolite, the low-cost adsorbent namely green mussel (*P. viridis*) has been used to partially replace activated carbon and zeolite. The purpose of this study is to produce activated carbon-zeolite, single composite adsorbent and low-cost adsorbent material such as green mussel. In order to achieve this objective, the optimal mixture for the above adsorption material must be determined based on the reduction of organic constitutes (COD) and ammoniacal nitrogen reduction.

2. Materials and methods

2.1. Adsorbent materials

For this analysis green mussel shell (*P. viridis*) was obtained from Ceria Maju Restaurant in Parit Raja, Johor, Malaysia. The commercially available adsorbents like coconut shell activated carbon and zeolite were purchased from Cabot Malaysia Sdn Bhd. and PT. Anugerah Alam Sdn. Bhd at the rate of about RM4000 and RM400 per ton, respectively. The media density has been measured, that is, weight of media/volume of media. Each of the media was grounded by 150 μm particle size using the ceramic ball mill for powdering the media. Table 1 categorized the adsorption material as low-cost adsorbent as well as indicated the type of material property as hydrophobic–hydrophilic.

Raw leachate sample used in this study was taken from the SRLS; located within the Klaung district, at latitude 10 53′41.64 ″22′34.68 N and 10 30″ E in Johor, Malaysia. The SRLS area covers about 6 ha, and aerated lagoon is used as method for handling landfill leachate. It is well equipped with a collection pond for leachate, but without any other treatment. SRLS is now over 12 y old and the SRLS collects about 250 tons of total solid waste per day, covering three regions that are Batu Pahat, Simpang Renggam, and Kluang [22].

Table 2 presents sample collection and characterization of landfill leachate wastewater collected from (SRL) site. Leachate wastewater contains a high concentration of COD, ammonical nitrogen as shown in Table 2. The collected samples are put in a HDPE ("high density polyethylene") plastic container, transported to the lab, and stored at 4°C. The experimental analysis and procedures were performed according to the American Public Health Association standard methods (APHA 2008) [23].

Ordinary Portland cement (OPC) is selected as a binder for binding all the adsorbents as a single medium. The amount of OPC was determined by means of attrition measurement; procedure modification [24]. Generally, five grams of composite media of particles ranges from 1.18 to 2.36 mm which were placed in 100 mL of raw leachate. The maximum reduction condition will be determined at 200 rpm with 120 min time interval, respectively. The mixture was filtered through a 1.18 mm size sieve and washed with distilled water. The particles which did not pass the respective sieve will be transferred to a pre-weighed glass watch. In this study, the quantity of OPC as binder had to be adequate to produce strong enough media that could not be broken during the batch analysis, particularly in the particle size effect experiment. The aim of this experiment was to get the minimum OPC amount that gives minimum attrition percentage.

2.2. Optimum ratio

The determination of optimal ratios between hydrophobic–hydrophilic mixing media, were determine the adsorption property toward COD and ammoniacal nitrogen, the main crucial problematic parameter in landfill leachate treatment. Batch experiment analysis was performed to determine the optimum adsorption ratio condition with 200 rpm in shaking speed, 120 min of contact time at pH7. The ratio that gave the optimal reduction of COD and ammonia nitrogen contaminant as shown in Tables 3a–c is considered to be the optimal ratio [36,37].

2.3. Batch adsorption experiment

The optimal batch experiments were conducted to determine the optimal condition using 5 g of media and 100 mL of leachate or 50 g/L of media concentration. To

Table 1
Adsorption materials

Type of adsorbent	Main adsorbent	Low cost adsorbent
Hydrophobic	Activated coconut shell carbon	Green mussel (*Perna viridis*)
Hydrophilic	Zeolite	–

Table 2
Simpang Renggam landfill site leachate characteristic

Parameter	Initial leachate concentration	Malaysian leachate discharge standard, mg/L (Malaysia Environmental Quality Act, 1974)
pH	8.27	6.0–9.0
SS (mg/L)	367	50
NH_3–N (mg/L)	406.68	5
COD (mg/L)	1,829	400
BOD_5 (mg/L)	163	20
Color (Pt-Co)	4,788	100
BOD_5/COD	0.08	–

Table 3a
Hydrophobic, activated coconut shell carbon – green mussel (*Perna viridis*)

ACSC (g)	GM (g)
0.0	4.0
0.5	3.5
1.0	3.0
1.5	2.5
2.0	2.0
2.5	1.5
3.0	1.0
3.5	0.5
4.0	0.0

ACSC: Activated coconut shell carbon; GM: Green mussel (*Perna viridis*)

Table 3b
Hydrophilic, zeolite

ZE (g)
0.0
0.5
1.0
1.5
2.0
2.5
3.0
3.5
4.0

ZE: Zeolite

Table 3c
Combination hydrophobic–hydrophilic

Hydrophobic (g)	Hydrophilic (g)
0	8
1	7
2	6
3	5
4	4
5	3
6	2
7	1
8	0

optimize ammoniacal nitrogen and aggregate organic (COD) percentage reduction by adsorbent. The static batch experiment was carried out to obtain the optimal conditions for all relevant factors like pH, contact time, shaking speed, dosage, and particle size. Adsorption isotherm model experiments were also carried out using distilled water with varying concentrations of leachate in a reaction mixture consisting of 50 g/L of adsorbent and 100 mL of leachate solution.

2.4. Analytical method

The concentration of COD was determined by using closed reflux and colorimetric (5220-D) method where the ammonia nitrogen concentration was determined by using Nessler reagent (Method-8038) method by using UV-vis spectrophotometer DR6000. The treated samples were first filtered before each measurement by using 0.45 μm filtered paper. All the procedures were carried out according to the Standard Water and wastewater examination method [25].

3. Result and discussion

3.1. Comparative study of COD and ammoniacal nitrogen with other adsorbent

Mainly, in this research, the three different adsorbent materials, that is, activated coconut shell carbon, green mussel (*P. viridis*), and zeolite were analyzed and compared with previously reported work. From the results, it was observed that ACSC, GM, and ZE have shown better performance as compared to other adsorbent materials. Additionally, the overall reduction percentage comparison of ACSC, GM, and ZE adsorbent is also included as listed in Table 4. From Table 4, it can be concluded that ACSC, GM, and ZE show better reduction percentage in terms of COD and ammoniacal nitrogen as compared to others.

3.2. Adsorbent preparation

A novel and inexpensive adsorbent is prepared from abundant waste materials for the reduction of COD and ammoniacal nitrogen as an organic and inorganic pollutant from leachate wastewater solutions. Coconut shell was collected at industrial site, washed, crushed, and rinsed with sulfuric acid of known concentration and kept for 24 h into the oven at 150°C for the reduction of volatile impurities and moisture content. The pyrolyzed sample was crushed to predetermined particle sizes of <150 μm

Table 4
Performance comparison with various other adsorbent

Adsorbent	COD reduction	Ammoniacal nitrogen reduction	References
AC-Cockle shell	31.98%	27.82%	[28]
Zeolite–Feldspar	49%	45%	[29]
Green mussel	45.5%	–	[30]
Sugarcane bagasse activated carbon	77.8%	41.05%	[31]
Rice husk carbon composite	27.61%	51.0%	[32]

by using ceramic ball mill [29]. The preparation of mussel shells required washing with distilled water several times to remove dust and fine particles. Then, the shells were dried in the oven at 105°C for 24 h. The cleaned and dried shells were then crushed with aggregate impact value machine and retained at 0.3–0.425 mm sieve. Then, the sieved waste mussel shell was calcinated at 750°C for 3 h [30]. A low cost natural zeolite is abundant in nature and was used as single media and as a component of the composite media. The samples were grounded to crush the particle size of <150 μm by using ceramic ball mill [13].

The adsorption media composition was determined based on the COD and ammonia nitrogen adsorption behavior. The optimal compositions of hydrophobic designed adsorbent material such as activated coconut shell carbon and green mussel (*P. viridis*) has been determine by varying the ratio of the both adsorbent material. The ideal ratio for activated coconut shell carbon and green mussel (*P. viridis*) was 2.5:1.5 g, the optimal reduction percentage of COD and ammonia nitrogen were 83% and 63% based on the operating conditions of 100 mL of leachate, 200 rpm shaking speed, 120 min contact time, particle size of <150 μm, and pH7 as shown in Fig. 1. The ideal ratio of hydrophilic adsorbent material zeolite was 1.0 g, the reduction percentage of COD and ammonia nitrogen were 78% and 96% based on the operating conditions 100 mL of leachate, 200 rpm shaking speed, 120 min contact time; and particle size <150 μm and pH 7 as shown in Fig. 2. The optimal ratio for adsorbent hydrophobic and hydrophilic material was 5:3. The reduction percentage of COD and ammonia nitrogen were 72% and 67% based on the operating conditions 100 mL of leachate, 200 rpm shaking speed, 120 min contact time; and particle size <150 μm and pH 7 as shown in Fig. 3. Ammonia has been selected for testing the hydrophilic media because of its very soluble properties in aqueous solution, either in the forms of ammonia or ammonium. In this research study, the zeolite was a significant material for hydrophilic media an ion-exchange sorbent having a high affinity for ammonium-ions. Activated coconut shell carbon adsorbent was found effective for adsorption of hydrophilic compound. It is effectively used for removing organic contaminant from the wastewater [26].

Prior to the experiment, chemical composition of activated coconut shell carbon, green mussel, and zeolite were determined by X-ray fluorescence spectrometry (XRF) instrument. Table 5 shows the properties of ACSC, GM, and ZE respectively.

Table 2 describe the characterization of raw leachate sample are taken from SRLS sited in Kluang district in Johor, Malaysia.

3.3. Adsorption isotherm analysis

To optimize sorption experiments it is important to determine the adsorption isotherm. For this study, the two well-known isotherm models of adsorption (Langmuir and Freundlich) were used. The isothermic Langmuir model was used for analyzing the formation of monolayers by adsorption on the adsorbent surface. On the other hand, the Freundlich isothermic model describes the process at a heterogeneous level (according to the surface adsorptions) in which the adsorbate surface concentration on the adsorbent increases with the elevation in the initial concentration of the solution. The correlation coefficient R^2 demonstrates or differentiates suitability of each equation.

For both the Langmuir and Freundlich isotherm model equations. The correlation coefficient R^2 value for COD indicates the value of R^2 in Langmuir was better fitted as compared to the Freundlich. Similarly, the correlation coefficient R^2 value for NH_3–N indicates the value of

Table 5
Chemical properties of ACSC, GM and ZE

Formula	Activated coconut shell carbon (wt.%)	Formula	Green mussel (*Perna viridis*) (wt.%)	Zeolite (wt.%)
Al	0.109	CaO	82.48	2.88
Ca	0.2171	SiO_2	0.44	61.5
CH_2	9,852.00	Al_2O_3	0.815	12.26
CI	4.07	MgO	0.265	0.64
Cu	0.21	K_2O	0.375	3.90
Fe	8.18	Fe_2O_3	0.315	1.48
K	30.28	TiO_2	0.26	0.21
Mg	3.61	SO_3	0.688	0.32
Mn	0.18	Na_2O	0.028	0.29
Mo	0.16	SO_4	0.11	–
P	7.74	P_2O_5	0.163	0.15
Re	0.50	SrO	0.158	0.13
S	4.09	ZrO_2	0.046	–
Si	0.52	$CaCO_3$	95.6	–
Zn	0.0043	C		7.55
Zr	0.07707			

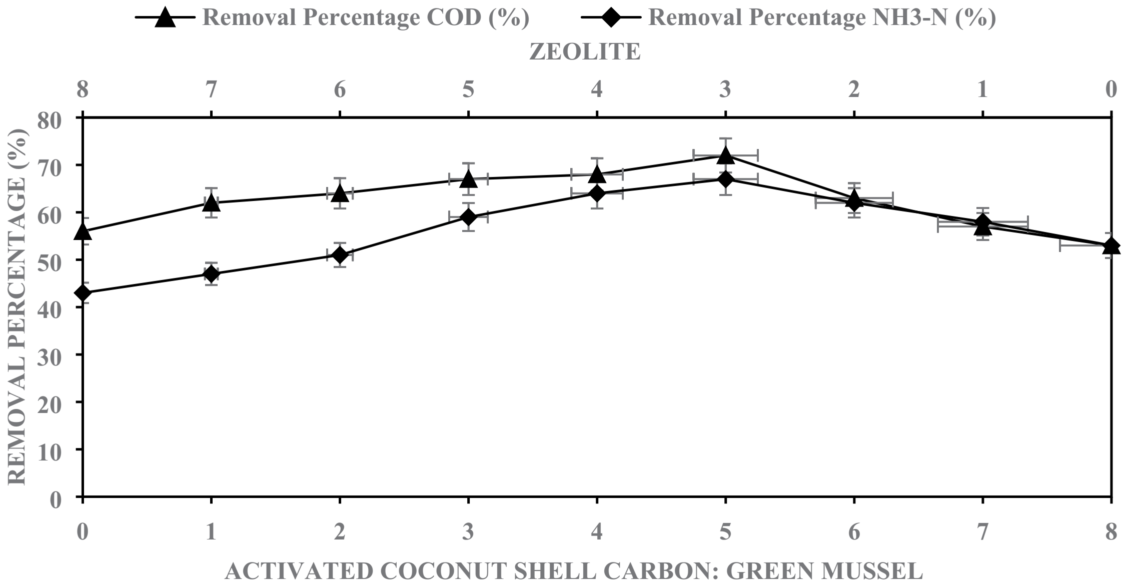

Fig. 3. Optimal ratio of hydrophobic–hydrophilic absorbent materials based on COD and NH_3–N adsorption (100 mL of solvent, shaking speed = 200 rpm, contact time = 120 min, particle size <150 µm, and pH = 7).

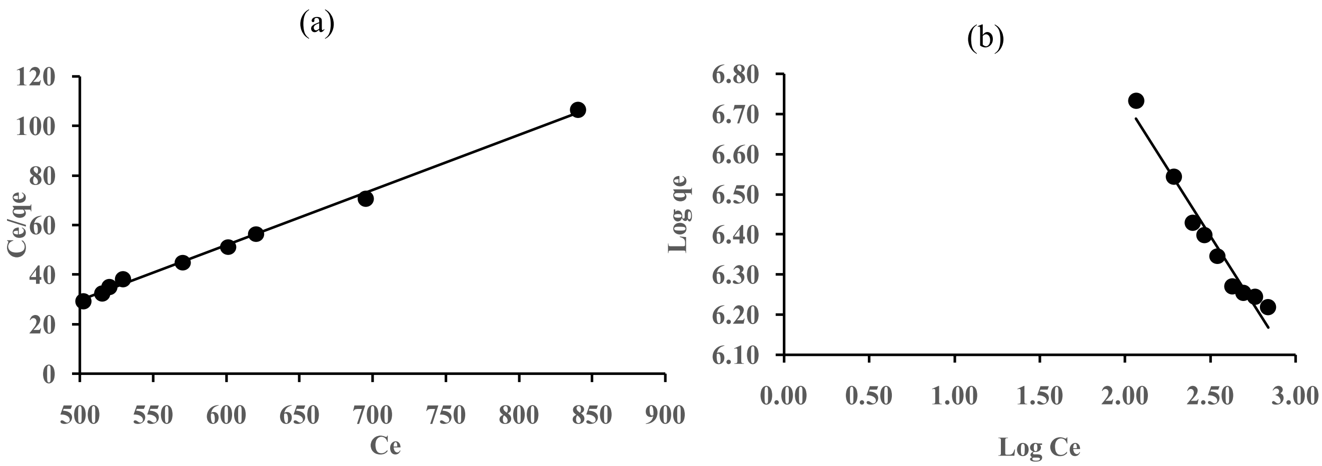

Fig. 4. Isotherm models for COD reduction (a) Langmuir and (b) Freundlich.

R^2 in Langmuir which was better fitted as compared to the Freundlich model (Figs. 4 and 5). The finding results suggest that Langmuir isotherm model is best fitted with strong correlation coefficient R^2 = 0.9971 for the reduction of COD and R^2 = 0.9914 for the reduction of Ammonia nitrogen, respectively. It implies the formation of monolayer while Langmuir isotherm model is favorable in COD, which also indicates the homogeneous surface [27,28].

4. Conclusions

The composite adsorbent material media was prepared using activated coconut shell carbon and zeolite and relatively low-cost adsorbent namely green mussel (*P. viridis*). The optimal ratio for activated coconut shell carbon and green mussel (ACSC:GM) was 2.5:1.5 whereas for zeolite (ZE) was 1.0. The overall ratio of hydrophobic and hydrophilic such as (ACSC:GM):(ZE) was 5:3. The optimal condition for static batch experiment was determined at 200 rpm and the shaking time was 120 min at pH 7. This research study indicates that both the isotherm models Langmuir and Freundlich show that this media is the best adsorption for organic constitutes (COD) and for an ammoniacal reduction in leachate wastewater treatment. According to the correlation coefficient or regression (R^2) value, COD adsorption is best fitted to Langmuir and Freundlich isotherm models, respectively.

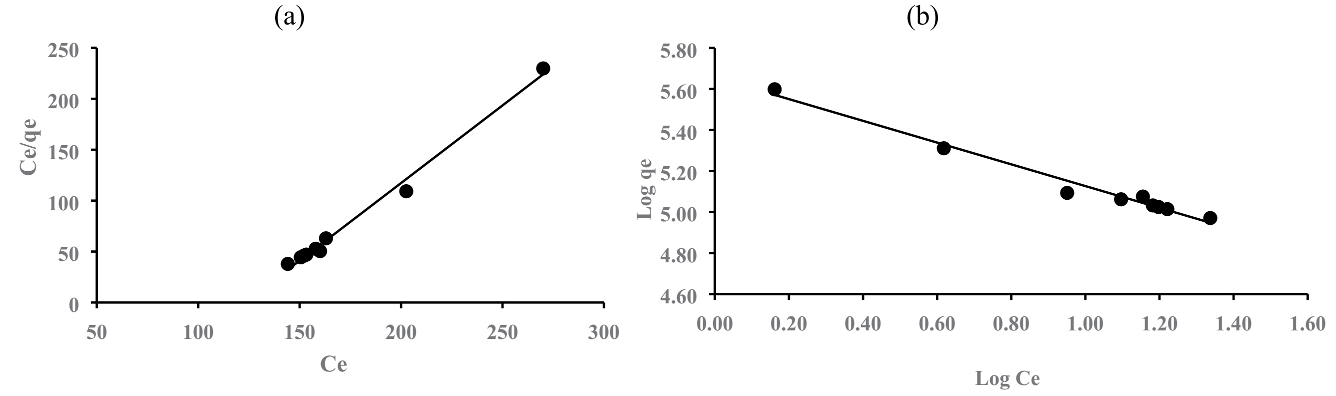

Fig. 5. Isotherm models for NH_3–N reduction (a) Langmuir and (b) Freundlich.

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