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Adsorption of chemical oxygen demand and ammoniacal nitrogen removal from leachate using seafood waste (green mussel shell) as low-cost adsorbent

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ABSTRACT

The physical appearance of leachate when it emerges from a typical landfill site is a strongly odoured black, yellow or orange colored cloudy liquid which contains organic and inorganic pollutants which makes it unsatisfactory to be released in water bodies without any prior treatment. The goal of current study was to examine the effectiveness of waste green mussel shell (WGMS) for the removal of pollutants from stabilized leachate. Chemical oxygen demand (COD) and ammonium nitrogen (NH_3–N) was investigated as the two major contaminants in leachate. In this study, adsorption method employed for the treatment of landfill leachate. Static batch experiment was carried out with 100 mL leachate sample with pH 7, contact time 120 min, dosage 4.0 g with 200 rpm shaking speed. The best removal was achieved at 2.0 g with the removal percentage of COD and N–NH_3 were 58% and 48%, respectively. Langmuir isotherm adsorption model showed best fitted with coefficient of determination for COD with the value of R^2 = 0.9944 and NH_3–N with the value of R^2 = 0.9918 respectively as compared to Freundlich model shows the value for COD with the value of R^2 = 0.9825 and NH_3–N with the value of R^2 = 0.9508. Thus, it indicates adsorbent adsorption occurs monolayer adsorption on homogeneous surface. The WGMS provides a significantly lower cost medium for reduction of COD and NH_3–N.

Keywords: Batch adsorption; Chemical oxygen demand; Ammoniacal nitrogen (NH_3–N); Leachate treatment; Waste mussel shell

1. Introduction

Solid waste management has become a serious challenge and problematic issue due to increasing amount of waste generation across the globe. The situation is alarming as the rate of solid waste generation is exceeding the permissible limits [1]. Rapid population growth and variation in consumption pattern directly or indirectly results in generation of large amount of waste which negatively effects the environment as well as pollute water, soil and

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air [2]. Wastes are produced from industries, municipal waste, building construction waste and institutional waste, that is, (waste produced from institutions such as schools, hospitals, or prisons. These wastes include liquid, sludge, solid, and hazardous waste, but not domestic. It is stated that according to the waste sources itself, few methods of waste disposal system can be applied to control waste such as open dumping, incineration, compaction, sanitary landfilling, reduction, composting and anaerobic digestion [3,4]. Among all methods, landfilling is the most popular and longtime applied method to reduce waste disposal. However, the major cause of waste disposal technique is associated with producing leachate from the landfills [5]. Landfills have traditionally been most widely used technique to organize and managing waste-disposal also it remains in so various places across the globe. This is cost effective and globally accepted technique for disposing and reducing municipal solid waste and industry waste [6–9]. Leachate contains high substances of organic and inorganic matter, consist of chemical substances, heavy metal such as ammonia, iron, sulphur, lead, nickel, cadmium, calcium, sodium and other [10]. Basic method of leachate treatment technology is divided into two; (i) biological treatment and, (ii) chemical treatment or physical treatment [11]. Leachate treatment is very difficult, costly, and usually requires a variety of application procedures [12]. Biological treatment is effective in removing organic compounds from fresh leachate because of fresh leachate's biochemical oxygen demand (BOD)/chemical oxygen demand (COD) ratio which ranging between 0.5 and 1.0. Meanwhile, BOD/COD ratio for stabilized leachate is less than 0.1 thus making it less effective to remove ammoniacal nitrogen in stabilized leachate using biological treatment [13].

In recent years, various technologies have been explored for treating landfill leachate including chemical precipitation, biological treatment, coagulation, ion exchange, ammoniacal stripping, reverse osmosis, and advanced oxidation process as well as natural treatment systems such as leachate recirculation and constructed wetlands have been developed [8,14]. Furthermore, all these techniques are costly and are not suitable for small scale industries as they do not give satisfactory outcome. Among all techniques, physicochemical technique by adsorption is very popular for stabilized leachate treatment as it is easy to use and cost effective if mixed with proper adsorbent and regeneration measures. Various low-cost adsorbent has been tested and used as an alternative for partially replacement of conventional material such as activated carbon and remove contaminants from leachate and aqueous solutions [15]. Ammoniacal nitrogen and chemical oxygen demand present in stabilized leachate are refractory organic compounds difficult to get rid through biological treatment [16]. Biological treatment processes becomes less effective and treatment methods like activated carbon adsorption and chemical precipitation is considered to be the ideal choice for treating soluble solids containing low organic compounds. In recent years, adsorption has gained importance removing compounds from water bodies due to low-cost, environmentally friendly, local availability, simplicity of design, ease of operation ability to adsorption techniques for wastewater treatment is widely used in many industries. Activated carbon is a renowned adsorbent for reducing of organic constituents and toxic metal. In most developing countries, the use of activated carbon keeps under control due to its high cost [17]. The utilization of basis materials in marine such as cockle shells, crab shells, green mussel shells and oyster shells possessed great potential as adsorbents in terms of its adsorption ability to reduce contaminants from contaminated water and its cost effectiveness [18].

Natural, agricultural and industrial waste are locally available material which may have potential as inexpensive sorbent. In Malaysia, mussel shell is abundantly available as a by-product from seafood industries and viewed as waste and is dumped or natural deterioration. The abundantly available natural source of waste mussel shell was used as basic materials to produced cost-effective adsorbents for treatment and eliminate of toxic from wastewater. In China, a mussel shell is used as an adsorbent for removing phosphate. They found removal percentage of phosphate was increased 29.56%–66.59% with time. These experimental outcomes have recommended the utilization of green mussel shell powder as great adsorption material for removing contamination from wastewater [19].

The selection of existing and readily available adsorbents waste green mussel shell (WGMS) in nature for adsorption treatment is a key focus to determine the adsorption capacity for the removal of organic and inorganic materials from leachate and potential in the regeneration process. In Malaysia, WGMS is abundantly available as a by-product from seafood industries and viewed as waste and for the most part left at dumpsite to naturally deteriorate. The abundantly available natural source of WGMS was used as an alternative material to conventional (activated carbon and zeolite) adsorbent to produce low-cost adsorbent for treatment and eliminate of toxic from wastewater.

In this study the use of waste green mussel shell is consider as novelty as well as to produce the low cost adsorbent for replacement of commercial material (activated carbon and zeolite). It can be concluded that waste green mussel shell (*Perna viridis*) show better removal percentages in terms of COD and NH_3–N as compared to other adsorbent material. A comparison study by previous studies and its performance has been added as shown in Table 3.

The significance of this study is to contribute new environmentally friendly technology by maintaining adsorption applications for leachate treatment and in turn can avoid the existence of pollution problem. This study is also expected to provide basic knowledge with respect to the application of composite adsorbents especially for leachate treatment. The aim of this study were determine two problematic parameters in leachate such as organic constituent (COD) and ammoniacal nitrogen (NH_3–N) and to examine latest characteristics of landfill leachate obtained from Simpang Renggam landfill site as well as the future work have been made as an emerging technique in the treatment of wastewater by utilizing local adsorbent derived from waste attributable to anthropogenic sources. Furthermore, from this study work, many scholars can utilize this data for better understanding in leachate characterization and proposed new ideas or techniques for improving efficiency of leachate treatment [20].

2. Materials and methods

2.1. Landfill leachate sampling

The leachate sample was collected from Simpang Renggam municipal landfill site in Johor, situated at latitude 10 53'41.64" N and longitude 1030 22'34.68" E Kluang District [20]. The existing area of the site was approximately 8 acres, which was insufficient to manage a large amount of waste. Every day landfill receives approximately 250 tons of waste. Therefore, the government had set up a new sanitary landfill beside the current one to cater the volume of waste discharged from the surrounding regions [21].

Raw leachate samples were collected from the influent of the detention pond in clean 20-L high density polyethylene plastic containers. The samples were transported to the wastewater research laboratory, Faculty of Civil Engineering and Built Environment, Universiti Tun Hussein Onn Malaysia (UTHM) and stored at 4°C to minimize any further changes in the properties of the leachate. According to the Standard Methods for the Examination of Water and Wastewater, all chemical analyses were performed within 24 h [22].

2.2. Preparation of waste mussel shell powder

The waste mussel shell samples were procured from Ceria Maju Restaurant situated in Parit Raja, Johore Malaysia. Mussel shell surface were completely cleaned with a brush and washed with deionized water several times to evacuate all the sand and fine particle, then oven-dried at 105°C for 24 h. Clean and dry shell samples were then crushed and grinded in a ceramic ball mill and retained around 75–150 μm sieved size. Powdered shell was then heated to a high temperature (calcinated) of 750°C for 3 h. This calcination process took three stages to complete; (i) temperature 640°C to 750°C where the powder is pure $CaCO_3$, (ii) temperature 640°C to 670°C where the real calcination actually begins and, (iii) temperature 670°C to 750°C where composition of $CaCO_3$ and CaO finally presents [23].

2.3. Batch adsorption experiments

The potential static batch experiment was carried out to determine the waste mussel shell powder in removing COD and NH_3–N from leachate. The adsorption experiment was conducted in a predetermined dosage of waste mussel shell powder filled with 100 mL of fresh leachate samples in a 100 mL Erlenmeyer flask at pH 7, 200 rpm, and 120 min contact time [24–28]. A sample tube glass beaker was placed on an incubator shaker. The optimum removal mixing ratio determined the effect of dosage on the batch adsorption, various dosage amounts of waste mussel shell powder (WMSP) ranged from 0.5, 1.0, 1.5, 2.0, 2.5, 3.0, 3.5 and 4.0 g was design. The glass was shaken for 2 h to make sure that the removal pollutant reached equilibrium, then, the samples were removed and analyzed for the residue concentration of COD and NH_3–N. The adsorption equilibrium capacity was calculated by the following equation:

$$q_e = \frac{V\left(C_o - C_e\right)}{M} \tag{1}$$

where q_e is the equilibrium adsorption capacity in (mg/g), C_o and C_e are the initial and equilibrium NH_3–N and COD concentrations in the leachate (mg/L). V is the volume of leachate solution in (L) and M is the weight of the adsorbent waste mussel shell in (g). The removal percentage efficiency of NH_3–N and COD in the solution calculated by using following equation:

$$E\left(\%\right) = \frac{\left(C_o - C_e\right)}{C_o} \times 100 \tag{2}$$

2.4. Analytical method

The ammoniacal nitrogen (NH_3–N) was determined by using Nessler's method, using HACH spectrophotometer, DR6000 and chemical oxygen demand (COD) was determined by using standard method of APHA as described [22] with the use of HACH spectrophotometer, DR6000, respectively.

2.5. Isotherm analysis

Langmuir and Freundlich isotherm models are widely used in an aqueous solution between the sorbate and the sorbent molecules to illustrate the processes of adsorption. Isotherm models such as isotherm and Freundlich have been used in this study to test the sorption process. According to the Langmuir model [29], all accessible sorption sites are homogenous and morphologically uniform. The Langmuir model's equation is given by;

$$\frac{1}{q_e} = \frac{q_m K_l C_e}{1 + K_l C_e} \tag{3}$$

The concept of Freundlich describes adsorption as a multilayer reversible process over heterogeneous surfaces [30]. The equation of the model is expressed by:

$$q_e = K_f C_e^{1/n} \tag{4}$$

3. Results and discussions

3.1. Characteristics of Simpang Renggam leachate

The leachate characterization is essential for determining the degree of leachate stability and most significant to decide appropriate effective method applicable for leachate treatment. The characteristics of leachate was examined and the results are shown in Table 1. The average value of COD and BOD_5 in leachate sample was 1,829 and l63 mg/L, respectively. The ratio of BOD_5/COD was less than 0.1, revealed that the leachate was stabilized. Thus, physico-chemical technique like adsorption is best fit for reduction of contaminants from the leachate [31].

3.2. Chemical composition of waste mussel shell

The chemical composition of waste green mussel shell used in this study was performed using X-ray diffraction

Table 1
Characteristics of raw leachate at Simpang Renggam landfill leachate site

Parameter	Minimum	Maximum	Average	Malaysia Leachate Standard, mg/L
pH	7.65	8.27	–	6.0–9.0
SS (mg/L)	316	367	341.5	50
NH_3–N (mg/L)	404.07	406.68	405.375	5
COD (mg/L)	1,595	1,829	1,712	400
BOD_5 (mg/L)	140	163	138.66	20
Color (Pt-Co)	4,685	4,788	4,736.5	100
BOD_5/COD	0.07	0.08	0.07	

(XRD) method. The finding results are presented in Table 2. Green mussel had a higher concentration of calcium carbonate which can be utilized in medication formulation, building constructions, or as filling in polymer material. These shells are made up of 95% $CaCO_3$ [32]. The results revealed that the composition of natural mussel shells mostly contain calcium oxide. In a previous study, Buasri et al. [33], reported that body of green mussel is made up to 98.36% of CaO.

Table 2
Chemical composition of green mussel (*Perna viridis*)

Composition	Percentage (%)
CaO	82.48
SiO_2	0.44
Al_2O_3	0.815
MgO	0.265
K_2O	0.375
Fe_2O_3	0.315
TiO_2	0.26
SO_3	0.688
Na_2O	0.028
SO_4	0.11
P_2O_5	0.163
SrO	0.158
ZrO_2	0.046
$CaCO_3$	95.6

3.3. Brunauer–Emmett–Teller analysis

The aim of Brunauer–Emmett–Teller (BET) theory explains the measurement of physical adsorption properties of the adsorbent of waste green mussel shell in this study was usually performed using BET method. This is conventional approach for determining the surface properties of adsorbent, catalyst and other materials. The surface characterization of the material effects the performance of material application. The BET surface of before and after treatment were 0.3579 and 9.7539 m^2/g. The finding result revealed that the BET value relatively higher surface area after treatment. The after treatment has a significantly making high effect on the material surface area.

3.4. Fourier-transform infrared spectroscopy analysis of mussel shells

The Fourier-transform infrared spectroscopy (FTIR) bands of before waste green mussel shell and after waste green mussel shell are represented in Fig. 1a and b in the range of 400–4,000 cm^{-1} wavenumbers. The FTIR analysis demonstrated that functional groups for after treatment of waste green mussel shell samples. The large spectra of before and after treatment at around 3,321.10 and 3,304.61 cm^{-1} in Fig. 1a and b is association to hydroxyl groups and the bond OH stretching mode. The band stretching at 1,471.81 and 856.10 cm^{-1} assigned to vibration of CH_n (aliphatic and aromatic) group.

3.5. Surface morphology of before and after mussel shells

Scanning electron microscopy (SEM) image of before and after waste green mussel shell are represented in Fig. 2a and b. The SEM morphology of the adsorbent material used to prepare was observed at x10, 000 magnification. From figure the SEM micrography, that observed morphology of the waste green mussel shell is not homogenous. It shows irregular shapes, sizes with low porosity structure and rudimentary pores.

3.6. Determination of optimum ratio

According to the static batch experiment, as presented in Fig. 3. The reduction percentage varied with waste mussel shell powder dosage for COD and ammoniacal nitrogen adsorption from leachate. Fig. 3 shows that shell powder was an efficient adsorbent to absorb COD. Thus, the surface of WGMS (*Perna viridis*) powder was more suitable to attract organic pollutants due to higher driving force increases the mobilization rate of organic from bulk fluid to film zone [34]. However, the optimum reduction percentage of COD and NH_3–N was 58% and 48% treating with 2.0 g of shell powder. The reduction percentage of COD and NH_3–N decreased on increasing the amount of waste mussel shell powder. The excess amount of shell powder in the batch reactor may cause to re-stabilization particles due to steric repulsion [35]. The optimal reduction ratio was used for the further experiment.

3.7. Determination of optimum shaking speed

The reduction percentage of COD and ammoniacal nitrogen are illustrated in Fig. 4, respectively, to achieve the

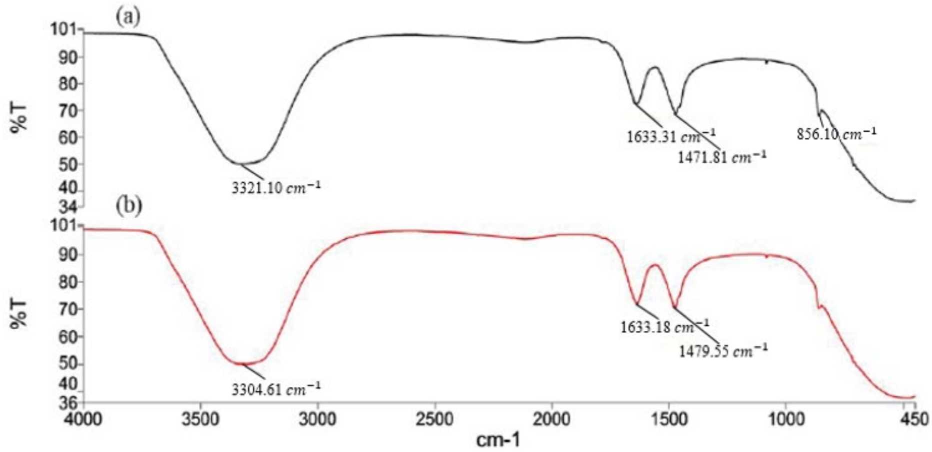

Fig. 1. FTIR spectra of waste green mussel shell (a) before and (b) after treatment.

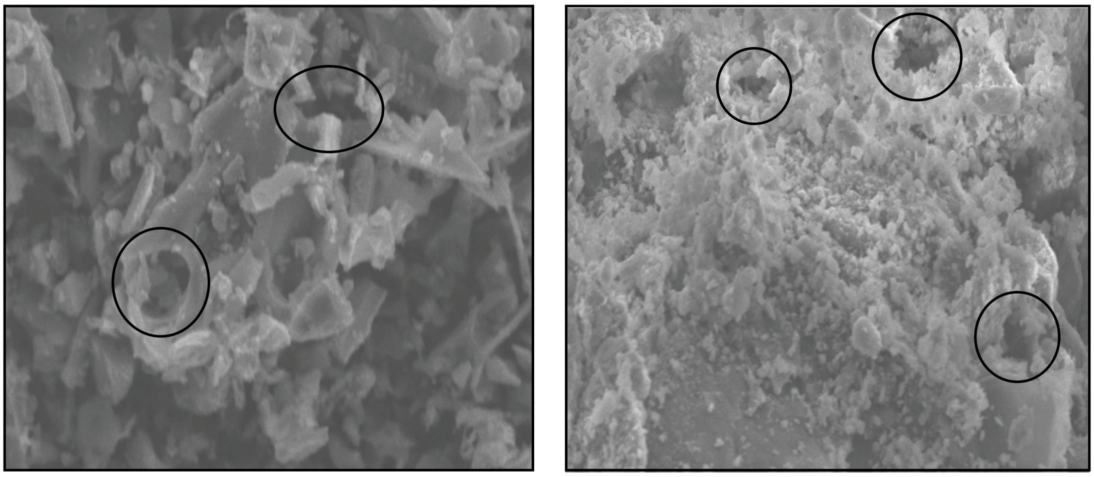

Fig. 2. Surface physical morphology of WGMS (a) before and (b) after adsorption.

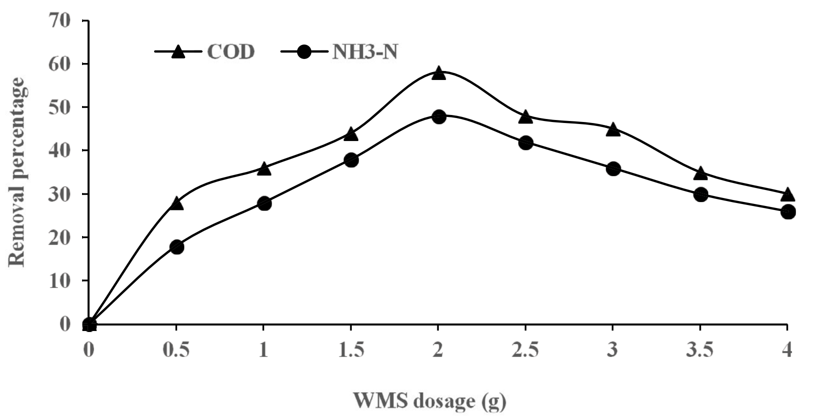

Fig. 3. Percentage of COD and NH_3–N removal vs. dosage of WMSP (g).

optimum shaking speed in the samples using waste green mussel powder and shaken at different rotational speeds (50 to 300 rpm). The most favorable initial condition for the reduction of two parameters (COD and NH_3–N) were achieved at 200 rpm are 45% and 42%, respectively. The optimal reduction of COD and NH_3–N are acceptable at the ratio of 2.0 g at 200 rpm speed. The optimal reduction result was used for the further experiment.

3.8. Determination of optimum contact time

The reduction percentage of COD and ammoniacal nitrogen are illustrated in Fig. 5, respectively, to achieve the optimum shaking time in the samples using waste green mussel powder and shaken at different time intervals (5–360 min). The most favorable initial conditions for the reduction of two parameters (COD and NH_3–N) were achieved at 120 min, at 45% and 40%, respectively. The optimal reduction of COD and NH_3–N are acceptable at the mixing ratio of 2.0 g, 200 rpm speed at 120 min shaken time. The optimal reduction result was used for the further experiment.

3.9. Determination of optimum pH

The reduction percentage of COD and ammoniacal nitrogen are illustrated in Fig. 6, respectively, to achieve the optimum pH level in the samples using WGMS (*Perna viridis*) powder and shaken at different pH intervals (4–12). The most favorable initial condition for the reduction of two parameters (COD and NH_3–N) were achieved at pH 7 of 48% and 40%, respectively. The optimal reduction of COD and NH_3–N are acceptable at the mixing ratio of 2.0 g, 200 rpm speed, 120 min shaken time, and pH.

Figs. 7 and 8 show the values of ammonia nitrogen and COD removal parameters for isotherm models. The parameters of the model are directly related to the system properties variation. The isotherm model results are best of fit and relatively determine the parameter for the removal of ammonia and COD by the adsorbent. Accordance with experimental data for the removal of contaminants investigated with R^2 value of 0.9944 and 0.9925 for COD and NH_3–N respectively. Therefore, the Langmuir model was more appropriate to be used to describe the process than the Freundlich model.

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

In this study the use of WGMS (*Perna viridis*) is consider as novelty as well as to produce the low cost adsorbent for replacement of commercial material (activated carbon and

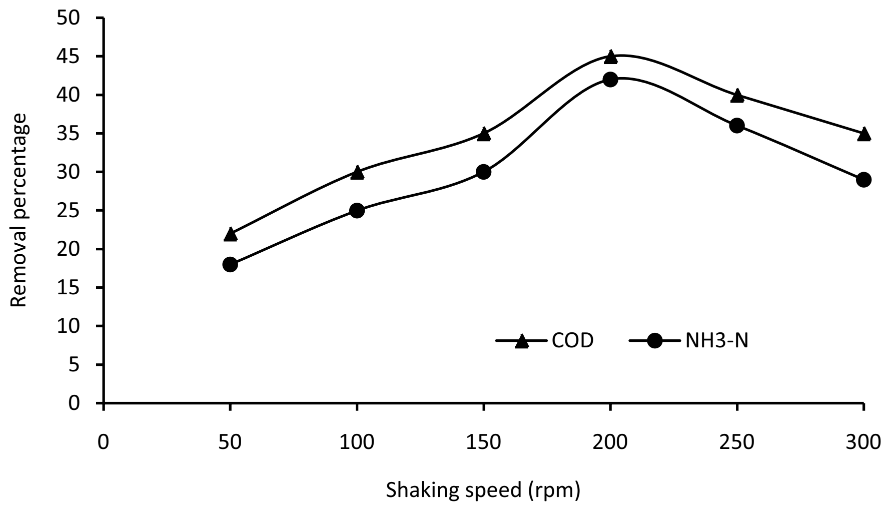

Fig. 4. Percentage of COD and NH_3–N removal against different shaking speed.

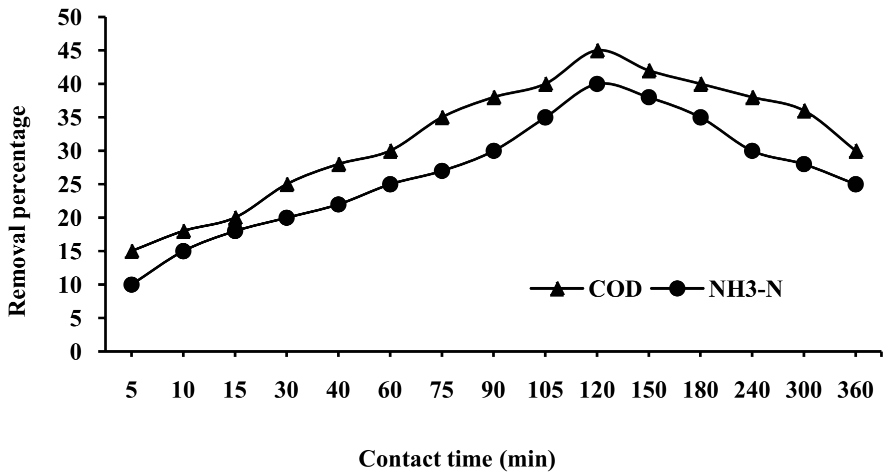

Fig. 5. Percentage of COD and NH_3–N removal against different contact time.

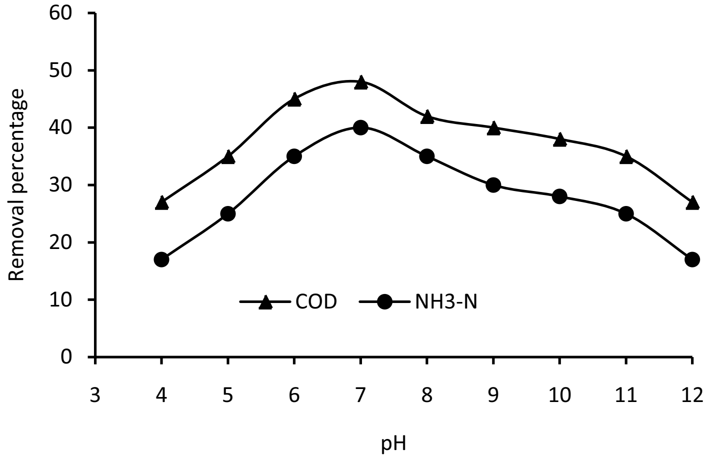

Fig. 6. Percentage of COD and NH_3–N removal against different pH.

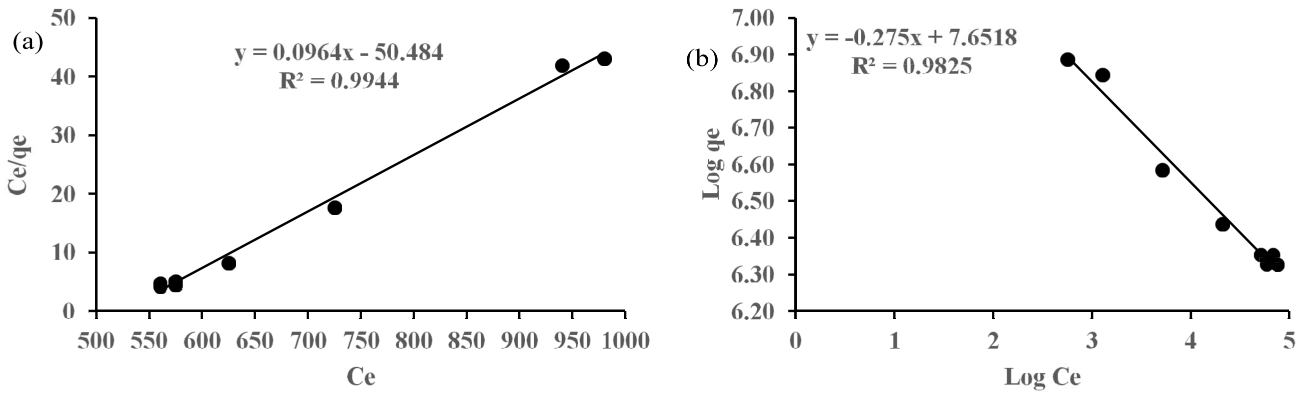

Fig. 7. Isotherm models for COD removal (a) Langmuir and (b) Freundlich.

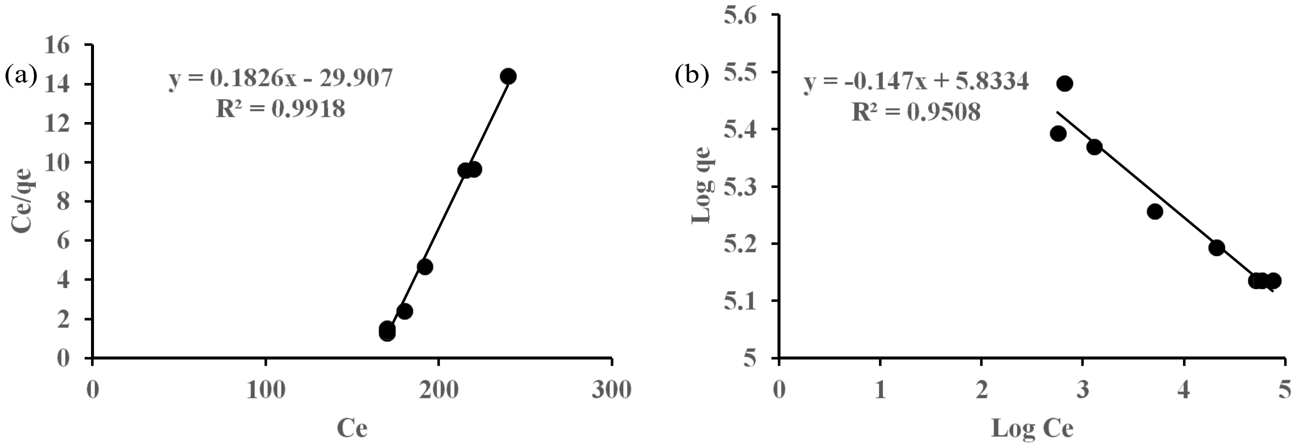

Fig. 8. Isotherm models for NH_3–N removal (a) Langmuir and (b) Freundlich.

zeolite). Mainly, in this research, the WGMS (*Perna viridis*) adsorbent material are analyzed and compared with the other researcher. While from the analysis, the WGMS (*Perna viridis*) have shown better performance as compared to other adsorbent materials. Additionally, the overall removal percentage comparison of WGMS (*Perna viridis*) adsorbent is also included in this listed Table 3. From the table, it can be concluded that WGMS (*Perna viridis)* show better removal percentages in terms of COD and ammoniacal nitrogen as compared to other adsorbent material.

Table 3
Performance comparison with various other researchers

References	Type of wastewater	Absorbent	Parameter	Removal efficiency (%)
[18]	Lake wastewater	Green mussel shell	NH_4^+	31.28
			COD	44.45
[36]	Leachate	Cockle shell	COD	55
[24]	Leachate	Activated carbon and green mussel	COD	83
			NH_3–N	63
[37]	River water	Cockle shell	COD	38.8
Present study	Leachate	Waste green mussel shell	COD	58
			NH_3–N	48

4. Conclusion

In this study, the effectiveness and regeneration ability of waste green mussel shell using as an adsorbent for to minimize COD and NH_3–N from stabilized leachate. The pattern of removal percentage of pollutants follows in the order of COD > NH_3–N with the value of removing percentage were (58% > 48%). On the basis of the above, it can be concluded that the optimum removal percentage of COD and NH_3–N was achieved at dosage 2.0 g is more efficient in reducing the pollutant from landfill leachate. The removal percentage of pollutants follows in order of COD > NH_3–N with the value of removing percentage were at shaking speed 200, contact time 120 min at pH 7 is more efficient in reducing the pollutant from landfill leachate. The good adsorption capability to remove pollutant from landfill leachate water demonstrated that WGMS would be promising in practical surface water treatment. The isotherm adsorption study of Langmuir and Freundlich isotherm shows best adsorption for the removal of COD and NH_3–N. According to the coefficient of determination, COD with the value of (R^2 = 0.9944) was better fit to Langmuir model while NH_3–N with the value of (R^2 = 0.9825) was best use for Freundlich model. This implies that adsorbate adsorption occurs by monolayer adsorption on a homogeneous surface. Therefore, it is proposed that kinetic adsorption be taken into account for further research to examine the COD and NH_3–N process on to the WGMS (*Perna viridis*) media respectively.

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