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Ammoniacal nitrogen and COD removal from stabilized landfill leachate using granular activated carbon and green mussel (*Perna viridis*) shell powder as a composite adsorbent

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ABSTRACT

Leachate is a liquid that is produced when water percolates through solid waste and contain dissolved or suspended material from various dissolved materials and bio-decomposition process. This study describes the finding of ammoniacal nitrogen and chemical oxygen demand (COD) removal partially replacing the amount of activated carbon. Activated carbon is well known as a good adsorbent for the removal of COD but the cost is relatively very high. However green mussel shell is the most abundant source in the world. Due to its abundance, green mussel is one of the low-cost materials. The combination of both materials is believed to produce inexpensive and suitable composite to treat the leachate. The batch experiment results indicate that the leachate concentration of ammoniacal nitrogen was (148 mg L^{-1}) and that of COD (308 mg L^{-1}). The optimum conditions for removal of ammoniacal nitrogen and COD were determined at 120 min with 200 rpm at pH 7. The optimum ratio of activated carbon and green mussel shell is 2.5:1.5. The values of removal percentage of NH_3–N, and COD are 63% and 83%, respectively. The availability of low-cost adsorbent material like green mussel shells in the composite has helped to reduce the treatment cost, along with enhancing the adsorption capacity and is environment friendly. The Langmuir isotherm adsorption model showed a better fit with strong correlation R^2 = 0.9962 for COD and R^2 = 0.9918 for NH_3–N, respectively, which means that the adsorption of leachate on granular activated carbon-green mussel shell powder, in this study, is homogeneous with the monolayer.

Keywords: Activated carbon; Green mussel shell; Composite; Ammoniacal nitrogen; Chemical oxygen demand (COD); Low cost adsorbent

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1. Introduction

Currently, the high population density is increasing the growth of commercial and industrial development in most of the countries. The increasing growth of population as well as the increase in the rate of municipal solid waste (MSW) generation. MSW is produced from various sources such as residencies, industries, commercial areas, institutions, construction and demolition, municipal services, treatment plants and sites, agriculture, and biomedical activities; each of these is heterogeneous [1]. This huge amount of MSW generation implies serious issues regarding public health and the environment. Irregular dumping of MSW affects all stakeholders, especially the public. However, improper dumping of urban waste affects the land and creates serious environmental and public health problems. Especially, domestic waste, which is organic in nature, poses a serious risk, since it makes the condition ideal to the survival and growth of microbial pathogens [2].

Groundwater is one of the real sources of water supply for residential and industrialization purposes. However, its quality is undermined due to the manner in which the waste is disposed. Groundwater is safer and reliable to use than the surface water, as it is less exposed to various pollutants. However, groundwater quality is exposed to dangers because of the contamination caused by various sources. One of them is pollution by leachate through landfills; in most urban areas landfills are the final destination of most of the generated waste. Landfill leachate is defined as the liquid that permeated through waste; it emerged containing heavy minerals and suspended materials from various dissolved materials and bio-decomposition processes [3].

The most important part of urbanization planning is to classify the area for the disposal of solid waste. However, serious health hazards and environmental problems can arise because of the location of landfills and methods of disposal. The great concern related to landfill's environmental impacts is connected to its effects on surface and groundwater, soil, odor emissions, air, and issues with respect to the transportation of solid waste [4].

A huge amount of waste is generated and is dumped at different landfill sites. These wastes originate from residential, commercial, and various other activities. These landfill sites produce bad odor if; the waste is not properly stored and treated. It can contaminate the air as well as surface and groundwater alongside polluting the soil and the subsoil. The landfill is generally used for solid waste disposal; the use of landfills will always play a vital role in waste disposal up to 95% of solid waste is currently disposed-off in landfills [5].

Malaysia is a south-east country, where landfilling is significantly important and the waste management standard system needs improvement. It consists of 13 states and 3 federal areas, with a total area of 329,700 km^2. Kuala Lumpur, officially the Federal Territory of Kuala Lumpur and commonly known as KL, is the national capital and the largest city in Malaysia. Being one of the fastest-growing economies in Asia, the generation rate of MSW increased from 292 kg/capita, in 2000, to 511 kg/capita in 2025 [1].

At present, landfilling methods play a vital role in disposing-off MSW around the world, significantly the best choice in the present and future, particularly for low and middle-income countries. Among the developing Asian countries like Malaysia, India, Vietnam, Indonesia, and Thailand), 70% to 90% of waste is disposed-off in the landfill as shown in Fig. 1 [6].

Worldwide various research studies have been carried out on the treatment of landfill leachate which utilized various types of treatment technologies like individual and combined such as, physicochemical and combined physicochemical and biological. However, consider these methods probably incur very high operational costs and most importantly are unsuitable for small scale industries. These also produce unsatisfactory results. Landfill leachate treatment is very expensive, complicated, and most importantly requires numerous processes. A few technologies, for example, reverse osmosis, chemical precipitation, membrane filtration, air stripping, oxidation, and adsorption are useful in the treatment of landfill leachate. Among these technologies, physicochemical treatment is one of the most important and useful ways for the treatment of stabilized leachate via adsorption [10]. All these different treatment strategies have been used in water treatment technologies, for example, bioremediation [11], electrochemical degradation [12], cation exchange membranes [13], Fenton chemical oxidation [14], constructed wetland [15] and photocatalysis [16–18].

Adsorption by activated carbon has gained considerable attention as it has a greater surface area, high adsorption capacity, and better thermal stability [19] and it is effective for adsorption of non-polar contaminants [20]. Additionally, with the presence of carbon–oxygen surface groups, polar compounds, and metal cations can also be absorbed. With these applications, it is widely used as an adsorbent in the process of water and air purification [21].

Using the activated cow dung ash (ACA) as well as cow dung ash (CA) to treat the wastewater for the removal of chemical oxygen demand (COD) using the batch mode. According to the batch mode result in the removal of COD using ACA has managed to get rid of 79% whereas using of CA were remove 66% at the same working range [22].

However, the use of activated carbon is not feasible in developing countries due to its high production cost and the need for the regenerative process of re-activating activated carbon columns [23]. There are various treatment processes available ion exchange and are considered

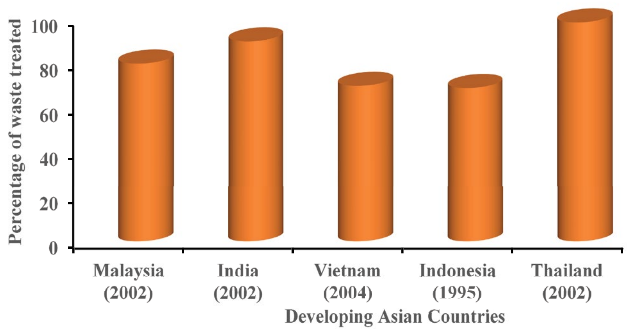


Fig. 1. Percentage of municipal waste treated [7–9].

cost-effective if low-cost ion exchangers, such as zeolites, are used [24].

In addition, this method seems to be more effective and user-friendly, if proper adsorbent and regeneration steps are combined. Nowadays, a variety of low-cost adsorbents have been used for the removal of various types of pollutants in the leachate arising from various types of wastewater and aqueous solutions for the past few decades. A wide range of materials has been utilized as a low-cost alternative to activated carbon [25]. Adsorption is one of the best and effective processes for removing organic and inorganic pollutants from water even at concentrations as low as 1 mg L^{-1}. Numerous non-conventional adsorbents have been used for water remediation, including organic and inorganic adsorbent and biomass. Granular activated carbon (GAC) is a well-known adsorbent for the treatment of water and wastewater because of its greater surface area, porous structure, capacity for proficient adsorption of a wide scope of adsorbates and ease of design [26].

Adsorption method for wastewater treatment has been generally utilized in various industries. The activated carbon is a widely known adsorbent and is used in removing organic, inorganic and heavy metals. In developing countries, however, the utilization of activated carbon is not in practice due to its high cost [27]. The adsorption capacities of various low-cost adsorbents created by premise material of marine shells, for example, crab shell, cockle shell, and clam shell can possibly be promising, as adsorbents, for the removal of pollutants from contaminated water [28–30].

Natural, industrial, and agricultural materials are locally available which may be used as cheap sorbents. In Malaysia, mussel shell is available in large quantities which originate from the seafood industry and is mostly left at dumpsites to natural deteriorate. The experimental data proposed that the use of green mussel shell powder, as an adsorbent, is good for removing heavy pollutants from the wastewater. The present study attempted to show the capability of green mussel shells in improving the quality of wastewater.

Daud et al. [31] state that a combination of two materials improves the adsorption capacity. The main objective of this study is to examine the characteristics of the leachate and investigate the removal concentration of NH_3–N and COD. In this study, two minerals namely activated carbon and green mussel shell are combined to improve the capacity of adsorption, reduce cost, and replace a part of activated carbon with the green mussel shell. The impact of activated carbon and green mussel shell ratio towards NH_3–N and COD removal is investigated in this study.

2. Materials and methods

2.1. Leachate characteristics

A raw leachate sample was collected for this study from Simpang Renggam landfill site (SRLS) in Johor, Malaysia [32] and was kept into a chiller for future use, according to the Standard Methods for the Examination of Water and Wastewater [33]. Leachate sample was collected from the landfill site and was put into a clean airtight container high-density polyethylene (HDPE). Once the leachate sample arrived at the laboratory, it was stored at 4°C to reduce further changes that might occur in its physicochemical and biological properties until the experimental analyses is carried out. All the chemical analysis were carried out within 48 h. The samples were tested for COD, biochemical oxygen demand (BOD_5), suspended solids (SS), ammoniacal nitrogen (NH_3–N), color, iron(Fe), and pH.

2.2. Media

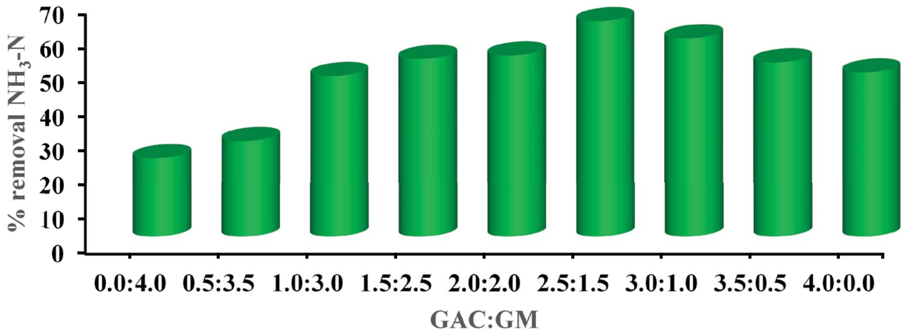
In this study, two different types of media were used: coconut shell GAC and green mussel shell powder (GM). The GAC was obtained from Cabot Malaysia Sdn Bhd, Negeri Sembilan, Malaysia. The green mussel was collected from Ceria Maju Restaurant located in Parit Raja, Johor. Both GAC and GM were pulverized and sieved to obtain a particle size of 75 to 150 μm (passed through the 100 sieves and retained on 200 sieves) using a ceramic ball mill. The chemical composition analysis of the mixed media was determined via X-ray fluorescence spectrometry (Model Bruker S4 Pioneer, Manufacturer Company, Singapore). The specific density of the media was determined conventionally (dry weight/volume) as shown in Table 1.

2.3. Optimum ratio

The optimum ratio is determined between GAC and GM based on the previous research studies which proposed various amounts by weight [31]. The total weight of the media mixture utilized for each conical flask was 4.0 g. The mixture ratios of GAC and GM used in this experiment are 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, and 4.0:0.0, as shown in Fig. 2. The static experiment was carried out to determine the GAC and GM performance in the removal of pollutants from the leachate. Ammoniacal nitrogen and COD experiment were carried out by batch experiments in a 100 ml conical flask. The batch experiment is performed in a 100 ml conical flask with varying amounts of media ratio (measured in terms of 4 g) as shown in Fig. 2. Optimum ratio determination

Table 1
Chemical composition of granular activated carbon and green mussel

Formula	Granular activated carbon	Formula	Green mussel
Al	0.109	SiO_2	0.81
Ca	0.2171	Al_2O_3	0.26
CH_2	9,850.00	Fe_2O_3	0.37
CI	4.07	CaO	74.54
Cu	0.21	MgO	0.31
Fe	8.18	K_2O	0.26
K	30.28	Na_2O	0.44
Mg	3.61	SO_3	0.52
Mn	0.18	Cl	0.02
Mo	0.16	SO_4	0.11
Si	0.52	$CaCO_3$	95.6
Bulk density (g cm^{-3})	0.619		1.56



Mixing ratio	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
Initial NH_3–N	406.68	406.68	406.68	406.68	406.68	406.68	406.68	406.68	406.68
Final NH_3–N	315	293	215	196	190	149	170	199.6	210.31
% removal	23	28	47	52	53	63	58	51	48

Fig. 2. Optimum ratio of granular activated carbon-green mussel for NH_3–N removal at pH 7, shaking speed 200 rpm and 120 min contact time.

between GAC and GM was based on the extent to which the mixture removed ammoniacal nitrogen and COD. In each conical flask, 100 mL of the leachate sample was added and was shaken for 2 h with shaking speed of 200 rpm at the pH value of 7 [34]. Three replicates of each sample were tested and the average results were used. The percentage removal of all parameters in the solution was evaluated by using Eqs. (1) and (2).

$$q_e = \frac{V\left(C_0 - C_e\right)}{M} \tag{1}$$

where q_e is the equilibrium adsorption capacity in (mg g^{-1}), C_0 and C_e are the initial and equilibrium ammoniacal nitrogen and COD concentrations in the leachate (mg L^{-1}). V is the volume of leachate solution in (L) and M is the weight of the adsorbent waste mussel shell adsorbent in (g). Besides that, the removal percentage efficiency of ammoniacal nitrogen and COD in the solution calculated by using the following equation:

$$E(\%) = \frac{\left(C_0 - C_e\right)}{C_0} \times 100 \tag{2}$$

2.4. Analysis method

pH was measured by using a portable pH meter HACH Sension, Manufacturer Company, Singapore. Total suspended solids were determined by the gravimetric method of the residue dried to a constant weight from 103°C to 105°C. The COD and NH_3–N were assessed by the closed reflux and Nessler Method respectively using the atomic adsorption spectrophotometer (Model DR6000, HACH Manufacturer Company, Singapore). BOD was measured with the method of 5220D (closed reflux, colorimetric method) and BOD measure of oxygen consumed in a 5 d test period respectively. The color was measured using HACH/DR6000 spectrophotometer and reported as platinum-cobalt (Pt-Co) method.

3. Results

3.1. Leachate characteristics analysis

Various studies describe the variation in the quality of leachate of different landfills. The characterization of the leachate indicates the degree of leachate stability, which is important to choose the most applicable treatment method. The characteristics of the collected raw leachate are presented in Table 2. The average values of COD and BOD_5, in the sample from SRLS are 1,829 and 163 mg L^{-1}, respectively. The ratio of BOD_5/COD is 0.08, which is less than 0.1 and the leachate is difficult to further degrade biologically [35].

Table 2
Simpang Renggam landfill site leachate characteristic

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

Meanwhile, the ammoniacal nitrogen also has a high concentration of about 406.68 mg L^{-1}. The data indicate that the leachate is stabilized. Therefore, the physicochemical treatment method is deemed the most applicable for the treatment.

3.2. Optimum ratio

The best optimum ratio was determined based on the extent to which ammoniacal nitrogen and COD were removed. The best combination ratio of GAC and GM, that gives maximum removal of ammoniacal nitrogen and COD, is 2.5:1.5 whereas the maximum removal percentage of ammoniacal nitrogen and COD is 63% and 83% respectively, as shown in Figs. 2 and 3.

Activated carbon is known as a popular adsorbent because of its high adsorption capacity which is enhanced by its large specific surface area. However, the use of activated carbon is an effective but costly adsorbent.

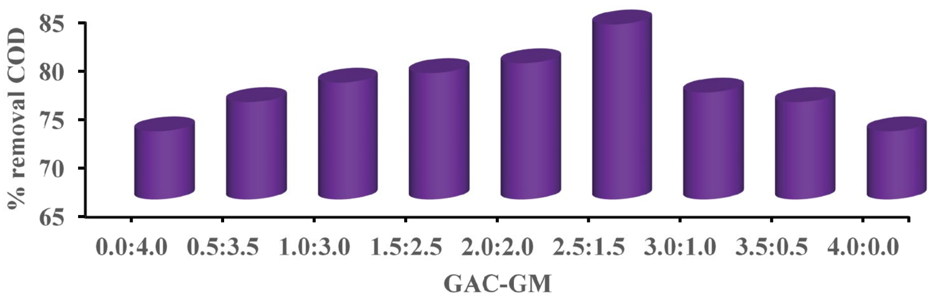
3.3. Isotherm analysis

Langmuir and Freundlich isotherm models are generally used to represent the processes of adsorption in an aqueous solution between the sorbate and the sorbent molecules. In this work, to test the sorption method, isotherm models such as Langmuir and Freundlich were applied. All accessible sorption sites are homogeneous and morphologically uniform according to the Langmuir model [36].

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

The Freundlich model defines adsorption over a heterogeneous surface as a reversible multilayer process [37]. The equation with the model is represented by;

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



Mixing ratio	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
Initial COD	1,829	1,829	1,829	1,829	1,829	1,829	1,829	1,829	1,829
Final COD	506	449	418	399	384	308	437	449	510
% removal	72	75	77	78	79	83	76	75	72

Fig. 3. Optimum ratio of granular activated carbon-green mussel for COD removal at pH 7, shaking speed 200 rpm and 120 min contact time.

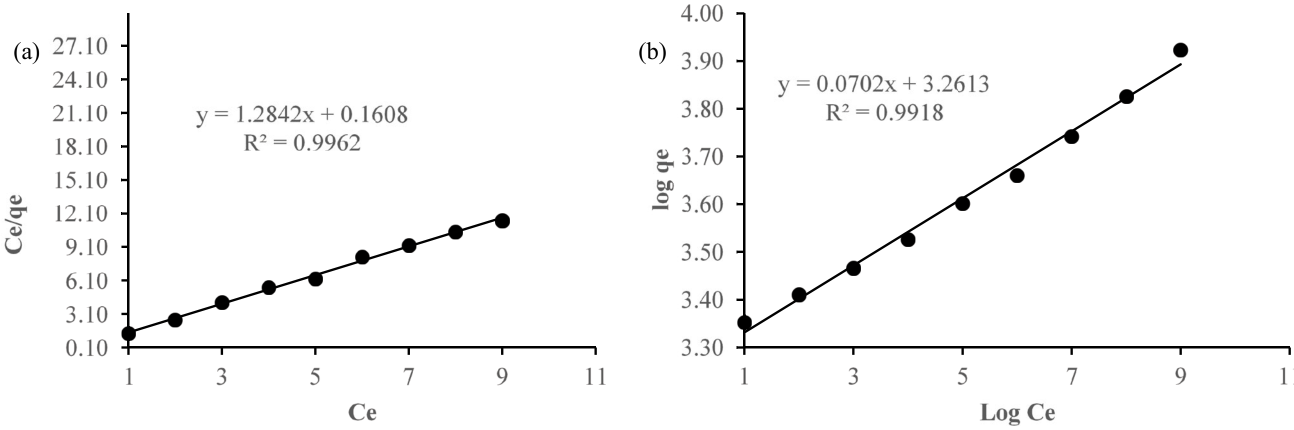


Fig. 4. Isotherm models for COD removal (a) Langmuir and (b) Freundlich.

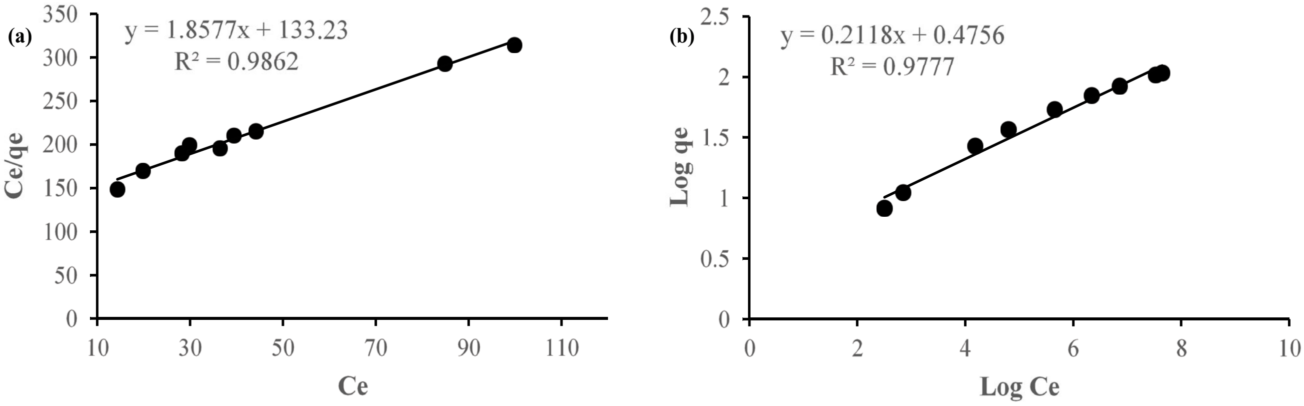


Fig. 5. Isotherm models for NH_3–N removal (a) Langmuir and (b) Freundlich.

The parameters values for COD and NH_3–N in isotherm models are identified to be directly related to the variation in system properties. The results show that the Langmuir and Freundlich models are best fitting and the relative parameters were determined by the adsorbent removing COD and NH_3–N as shown in Figs. 4 and 5, respectively. The Langmuir models for COD and NH_3–N were based on experimental data of pollutant removal, which were investigated with R^2 values of 0.9962 and 0.9862. Therefore, the Langmuir model was more fitting than the Freundlich model to be used to describe the process, respectively.

4. Conclusion

In this study, the adsorption efficiencies of NH_3–N and COD were investigated. GAC and green mussel were used together in a composite media with green mussel as an alternate adsorption medium to replace a part of activated carbon. The ideal mixing ratio condition was obtained to be at 2.5:1.5 at the pH value of 7, contact time of 120 min and shaking speed of 200 rpm. With this composite media, the removal percentages of NH_3–N and COD are 63% and 83% respectively, which are higher as compared to the other ratios. Therefore, based on this ratio (2.5:1.5), the main advantage of green mussel waste is that it partially replaces the amount of activated carbon thereby reducing the waste and producing a low-cost adsorbent for the treatment of contaminated water. The Langmuir adsorption isotherm could well be fitted by the Langmuir model. The R^2 value, investigated in the present research, is less than one. A good indicator of the GAC-GM potential, for its use as an adsorbent, was the adsorption efficiency obtained for COD and NH_3–N. For further research, therefore, it is proposed that kinetic adsorption be taken into consideration to examine the processes of COD and NH_3–N adsorption on GAC-GM.

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