



## Batch Study on COD and Ammonia Nitrogen Removal Using Granular Activated Carbon and Cockle Shells

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### ABSTRACT

Landfills generate leachate that contains elevated concentration of contaminants and is hazardous to human health and the ecosystem. In this study, the mixture of granular activated carbon and cockle shells was investigated for remediation of COD and ammonia from stabilized landfill leachate. All adsorbent media were sieved to a particle size between 2.00 and 3.35 mm. The optimum mixing ratio, shaking speed, shaking time, pH, and dosage were determined. Characterization results show that the leachate had a high concentration of COD (1763 mg/L), ammonia nitrogen (573 mg/L), and BOD5/COD ratio (0.09). The optimum mixing ratio of granular activated carbon and cockle shells was 20:20, shaking speed 150 rpm, pH level 6, shaking time 120 min, and dosage 32 g. The adsorption isotherm analysis reveals that the Langmuir isotherm yielded the best fit to experimental data as compared with the Freundlich isotherm. The media produce encouraging results and can be used as a good and economical adsorbent.

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### NOMENCLATURE

$C_e$	Equilibrium concentration of the adsorbate ( $\text{mg L}^{-1}$ )	$C_o$	initial concentration in leachate ( $\text{mg L}^{-1}$ )
$q_m$	maximum amount of adsorbate per unit weight of adsorbent ( $\text{mgg}^{-1}$ )	$C_f$	leachate concentration at equilibrium ( $\text{mg L}^{-1}$ )
$K_l$	Langmuir constant related to binding sites affinity with adsorbate ( $\text{L mg}^{-1}$ )	V	leachate volume (L)
$K_f$	Freudlich constant	m	adsorbent mass (g).
$n$	Constant relating to adsorption intensity.	$q_e$	Equilibrium sorption capacity ( $\text{mgg}^{-1}$ )

## 1. INTRODUCTION

Landfills are naturally employed for discarding of solid waste generated from municipal or industrial activities. Factors have been described in literature that affect solid waste disposal [1-3], Landfills and environment [4-14], degradation of leachate and adsorbents [15-19]. Activated carbon is a popular adsorbent for diverse fluid components because of its adsorption capacity, which is

enhanced by its large surface area. This feature makes activated carbon an effective, yet costly adsorbent. Many technologies have been developed to overcome the cost problem by applying marine polysaccharide-based adsorbents, such as cockle shells, which are low cost and effective [20-22]. Cockle shell is hydrophobic and comprises primarily of CaO of nearly equal percentage ratio (96.28%) as compared to that in limestone (97.67%); [17, 18]. Combining activated carbon with natural materials, such as cockle shells, may serve as a suitable alternative to using activated

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carbon alone and result in economically sustainable treatment. Considerable research has investigated the use of conventional medias involving combination of activated carbon and zeolite for remediation of stabilized leachate, but few of them focused on the application of cockle shell as substitute of conventional media in landfill leachate treatment. The present article presents the results from research progress on leachate treatment using alternative biomedias. A combined loose media was experimentally explored to analyze the optimum mixing ratio, shaking speed, shaking time, pH, and dosage for activated carbon and cockle shells in treatment of COD and ammonia nitrogen in a stabilized leachate. Experimentation was conducted through partial replacement of activated carbon with cockle shells.

## 2. MATERIALS AND METHODS

**2. 1. Sampling** The leachate sample was collected manually at Simpang Renggam municipal landfill in Johor. The study area is located at latitude 1° 53'41.64" N and longitude 103° 22'34.68" E in Kluang District. The landfill has been managed for numerous years by the government and is recipient of approximately 250 tons of waste daily [19, 20]. Samples of raw leachate were collected according to method described by [12]. All chemicals used for leachate characterization were of analytical grade.

**2. 2. Media** Two different mixed media types were used: coconut shell granular activated carbon (GAC) and cockle shells (CS). The GAC was purchased from Cabot Malaysia Sdn Bhd, Negeri Sembilan, Malaysia, at a price of RM4 per kilogram. The CS was obtained and prepared according to the method outlined by [11]. The chemical constituents of the media were determined with X-ray fluorescence spectrometer (Model Bruker S4 pioneer). The density of the medias was determined conventionally (dry weight/ volume). The GAC and CS were sieved to obtain particle sizes between 2.00 and 2.35 mm. Tables 1 and 2 show the general properties of the media.

**2. 3. Batch study** Optimum mixing ratio experiment between GAC and CS was determined for the removal of ammonia nitrogen and COD. A total mixture of 40 cm<sup>3</sup> was introduced in each conical flask with 100 mL of leachate, and each flask was shaken at maximum speed of 200 rpm for 105 min at ambient temperature using orbital shaker (model Daiki) and pH 7. The samples were filtered using a 0.45-µm filter paper and analyzed for the two notable pollutants in the leachate, namely, ammonia nitrogen and COD. The ammoniacal nitrogen in mg/L was determined using

ultraviolet visible spectrophotometer model HACH DR6000 and COD was assessed by the closed reflux titrimetric method 14. The sequence of media mixing ratios used (for GAC:CS) were 4:56, 8:48, 12:40, 16:32, 20:24, 24:16, and 28:8 based on mass (g). The optimum values for the combination of the two medias were obtained by plotting mixing ratio against removal percentages. The mixing ratio with high removal percentage was selected as the optimum.

Optimization of shaking speed experiments were conducted in a series of 250-mL conical flasks containing 100 mL of raw leachate, and a measured mass of the media as obtained earlier in ratio was introduced into each flask. Optimum shaking speed was analyzed by varying the shaking speed from 50 rpm to 200 rpm. The flasks were shaken for 105 min. The rotational speed with the maximum depletion of ammonia and COD was selected as the optimum speed.

The influence of pH in the minimization of COD and ammonia in the samples was assessed by modifying the pH levels to 4, 6, 7, 8, and 9. The pH adjustment was administered with 97% H<sub>2</sub>SO<sub>4</sub> and 1M NaOH.

**TABLE 1.** Chemical composition of activated carbon

Formula	Concentration (Ppm)
Al	0.101
Ca	0.2053
CH <sub>2</sub>	98.6
Cl	0.0539
Cr	0.0021
Cu	0.0015
Fe	0.2322
K	0.4539
Mg	0.0252
Mn	0.0066
Mo	0.001
Si	0.0619

**TABLE 2.** Chemical composition of cockle shells

Formula	Cockle Shell (%)
CO <sub>2</sub>	0.10
SiO <sub>2</sub>	0.30
Al <sub>2</sub> O <sub>3</sub>	-
Fe <sub>2</sub> O <sub>3</sub>	0.80
K <sub>2</sub> O	3.55
CaO	92.00
MgO	-
Na <sub>2</sub> O	0.92

After every adjustment to a particular pH, experimentation was performed at a predetermined optimum shaking speed and optimum mixed ratio. The optimum pH was then selected depending on the pH that yielded the maximum decrement of COD and ammonia.

To assess the optimum shaking time, 250-mL conical flasks containing 40 mL of predetermined media mixture (GAC:CS) and pH and 100 mL of leachate were agitated at the predetermined shaking speed on an orbital shaker. The conical flasks were collected from the shaker after every 60, 120, 180, 240, 300, and 360 min. The samples were then filtered and analyzed.

Optimum media dosage was investigated by applying the range of adsorbent from 4 g to 56 g. Experimentation was conducted at predetermined shaking speed, mixed ratio, pH, and shaking time. The dosage that yielded the maximum removal was selected. The quantity of the adsorbed COD and ammonia per unit mixed media was evaluated using Equation (1).

$$q_e = (C_o - C_f)V/m \tag{1}$$

$$\text{Removal (\%)} = (C_o - C_f)/C_o \times 100 \tag{2}$$

The percentage removal (%) was evaluated using Equation (2).

### 3. RESULTS AND DISCUSSION

**3. 1. Chemical Analysis of Leachate** Several studies have described variations in the quality of the leachate obtained from different landfills [16]. The result suggests that the leachate had a high amount of COD and ammonia nitrogen. The average values of BOD<sub>5</sub>, ammonia nitrogen, pH, and COD for the leachate were 164, 573, 8.11, and 1763, mg/L, respectively; and the biodegradability ratio (BOD<sub>5</sub>/COD) of raw leachate was 0.09. The data of the BOD<sub>5</sub> and COD indicate that the leachate was stabilized. Stabilized leachates are usually high in NH<sub>3</sub>-N (>400 mg/L) and COD (<3000 mg/L) and have low biodegradability ratio. pH of leachate usually increases with time, indicating the decrease in concentration of the partially ionized free volatile fatty acids. Previous research has revealed that pH of stabilized leachate is higher than 7.5 [12].

**3. 2. Media Mixing Ratio** Figure 1 exhibits the percent removal in the sample after mixing activated carbon with CS and realizing optimum mixing ratio shaking for the removal of COD and ammonia nitrogen. From the figure, the most favorable conditions were at 5:35 and 20:20. These mixing ratios obtained COD removal rates of 31.98% and 31.99%. Therefore, the mixing ratio of 5:35 contains a more significant amount of activated carbon and is costlier than the mixing ratio

of 20:20. The main purpose of using the alternative media is to minimize cost while assuring quality. Accordingly, 20:20 was selected because this mixing ratio implies less conventional media content. Figure 2 shows the removal of ammonia nitrogen after mixing at different ratios.

The optimum removal occurred at 20:20 (27.82% removal). The selected optimum conditions were used for further experiments.

A one-tailed two-sample t-test was also used to determine whether the mix ratio of 20:20 and 5:35 participate more in parameter removal compared to the overall mix ratio. The t-test results for 20:20 and 5:35 mix ratio was summarized in Table 3. The results show that on average, the mix ratio 20:20 and 5:35 removed more parameter than general mix ratios. The results also revealed that a significant difference exists for the two mix ratios and combined data. It is therefore evident from data that statistically, mix ratio 20:20 is significant in parameter removal more compared to the overall mix ratio.

**3. 3. Effect of shaking speed** The media mixing ratio of 20:20 was applied to determine the optimum shaking speed, which plays a significant part in parameter reduction.

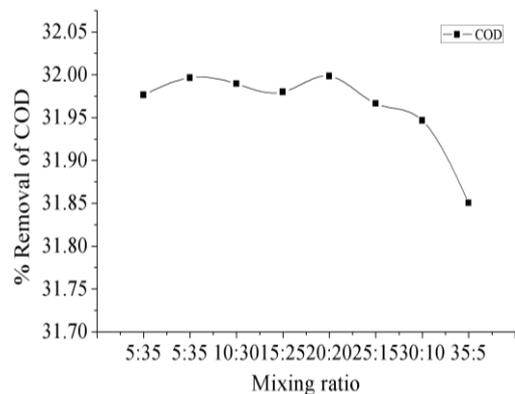


Figure 1. Mixing ratio between AC and CK in COD removal

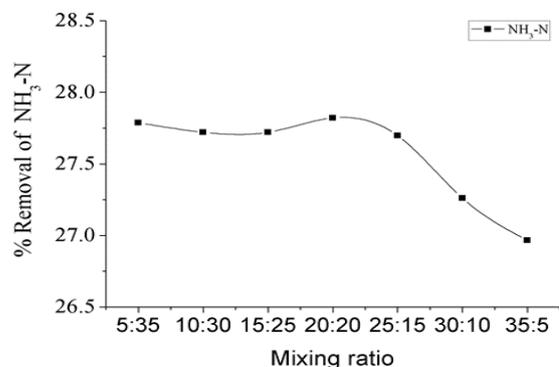


Figure 2. Mixing ratio between AC and CK in ammonia nitrogen removal.

TABLE 3. t-Test result

Mix ratio	N	mean	All mix ratio mean	T-value	P-value
20:20	7	31.998	31.9610	-1.890	0.041
5:35	7	31.996	31.9610	-1.794	0.049

From Figures 3 and 4, the percentage of COD and  $\text{NH}_3\text{-N}$  decreased with the increase in agitation rate in the range of 50–150 rpm. This result may be attributed to the diffusion of the adsorbate in the liquid bulk to the adsorbent surface, which increased with the increase in shaking rate. Another reason may be the reaction occurring on the surface of the CS as a result of the attractive forces between the ions present in the fluid and the CS and the surface of the activated carbon by the adsorbate. At agitation speed of above 150 rpm, the removal decreased with further agitation. This finding is possibly due to the decrease in vacant adsorption sites and the reduction of concentration gradient between the adsorbate and the adsorbent in the fluid. The optimum shaking speed adopted for further experiment was 150 rpm. This shaking speed obtained COD and  $\text{NH}_3\text{-N}$  removal rates of 49% and 38%.

**3. 4. Effect of pH** The pH of an aqueous solution is a key factor in the adsorption process and influences the surface charge of the ionizable surface groups of the adsorbent and the adsorbate particles.

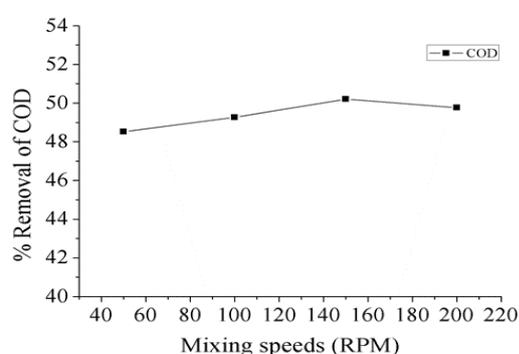


Figure 3. Optimum mixing speed for AC and CK mixture in COD removal

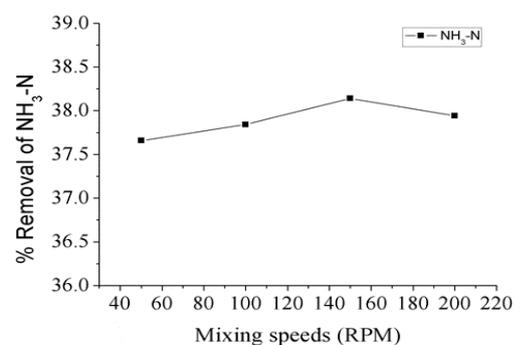


Figure 4. Optimum mixing speed for AC and CK mixture in ammonia nitrogen removal.

The influence of pH on the adsorption process was studied by varying the pH of the solution from 2 to 9. Figure 5 shows the removal rates of COD and ammonia at different pH levels. Obviously, the adsorption rate continuously improved with the increase in pH until the pH reached 6, and then gradually decreased until the pH reached 9. This behavior may be ascribed to the electrostatic attraction occurring between the solid adsorbent on which the ionizable surface groups presented a positive charge because of acidic condition and that of the organic functional groups of the adsorbate in the bulk liquid. Accordingly, the COD diminished until equilibrium state at pH 6. A similar interactive force may have drawn inorganics, such as ammonia, to the adsorbent surfaces.

Thus, the organic and inorganic ions may accumulate in a thin film on the adsorbent. In addition, molecules of the same charges usually repulse from one another and cannot crowd together on the adsorbent [23]. Beyond pH 6, the adsorption process had decreased. This decrease is probably because the surface of the adsorbent had become less positively charged owing to the predominance of  $\text{OH}^-$  ions on the surface of the adsorbent. Such predominance in turn causes the dissociation between the negatively charged surface and anionic elements in the aqueous solution, thereby decreasing the adsorption phenomena.

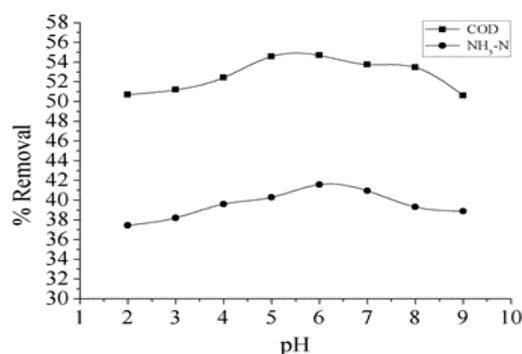


Figure 5. Optimum pH for AC and CK mixture in COD and ammonia nitrogen removal.

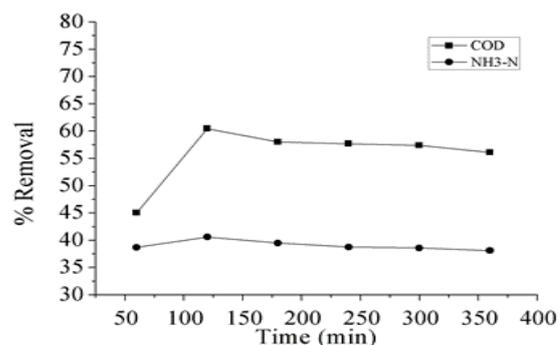
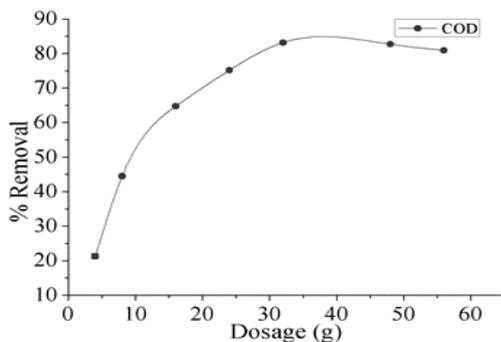


Figure 6. Optimum shaking time for AC and CK mixture in COD and ammonia nitrogen removal.

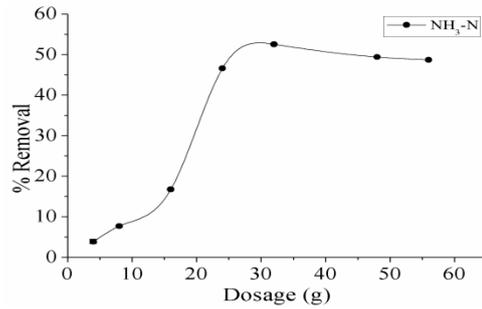
The result reveals that the optimum removal rates of 54% and 38% for COD and ammonia occurred at pH 6. Accordingly, pH 6 was adopted for further experiments.

**3. 5. Optimum Shaking Time** The concentration of adsorbate species in the bulk fluid and the interaction time between the adsorbent and adsorbate play dominant roles in minimizing pollutants from effluent water. The time duration needed to achieve maximum removal efficiency is the time when the sorption system attains equilibrium. The result of shaking time on COD and ammonia nitrogen removal by the adsorbent is illustrated in Figure 6. The removal of COD was increasing with the increment in time and attained equilibrium in about 120 min. The ammonia nitrogen removal was nearly the same from shaking time of about 105 min to 120 min. This phenomenon at the early stages is attributed to the vacant sorption sites on the media that initially became increasingly abundant and then slowed down owing to increasing competition for available vacant sites as a result of adsorbate saturation on the adsorbent and repulsion between similarly charged species. The plot exhibited a single, even, and continuous profile, suggesting the likelihood of monolayer coverage on the surface of the adsorbent. Similar sorption traits have been reported in literature [23-26]. The shaking time of 120 min was adopted, which obtained COD and ammonia nitrogen removal rates of 60% and 49%.

**3. 6. Optimum Dosage** Inadequate dosage or excess dosage of media may lead to an inadequate capacity in parameter removal. Consequently, the optimum dosage of media must be determined to achieve economic savings and increased efficiency. The influence of adsorbent concentration on adsorption of COD and ammonia was investigated by varying the dosage in the range of 4-56 g at predetermined optimum parameters (mixed ratio of 20:20, shaking speed of 150 rpm, pH level of 6, and shaking time of 120 min).



**Figure 7.** Optimum dosage for AC and CK mixture in COD removal



**Figure 8.** Optimum dosage for AC and CK mixture ammonia nitrogen removal.

From the results, the percentage removal of the adsorbate expeditiously increased with the increase in the adsorbent dose until it reached an equilibrium condition when the dosage was 32 g. This condition obtained COD and ammonia nitrogen removal rates of 83% and 52%. Afterward, the percentage removal decreased with additional increment in the adsorbent dose (Figure 7 and 8). This behavior can be accredited to an increase in the amount of available sorption sites from the onset until the optimum mass is reached; thereafter, any further increase in the adsorbent dose may result in aggregation, which can decrease the probability of molecules contacting all available adsorption sites [19-21]. This attribute is similar to the one reported by [27] in an investigation of the removal of COD by CS in jar test, and they reported a removal efficiency of 38.8% using 3g/L adsorbent.

**3. 7. Adsorption Isotherms** The evaluation of the adsorption isotherms is essential for the optimisation of sorption experimentation. The two renowned adsorption isotherms (Langmuir and Freundlich) are used in this study. The Langmuir isotherm analyzes the formation of a monolayer adsorbate onto the adsorbent surface. This isotherm is based on the premise that finite adsorption sites (homogenous) exist on the adsorbent surface, in which no further adsorption can occur when this surface is fully occupied. On the contrary, the Freundlich isotherm model explains heterogeneous surface adsorption, in which the surface concentration of the adsorbate on the adsorbent increases with the elevation in the initial concentration of the solution [27, 28]. The Langmuir isotherm is represented by Equation (3).

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

The linearized Langmuir equation is expressed as follows:

$$1/q_e = 1/q_m + (1/q_m K_l)(1/C_e) \tag{4}$$

The maximum amount of adsorbate ( $q_m$ ) collected in a given system can be evaluated from the isotherm. A linear plot of  $\frac{1}{q_e}$  versus  $\frac{1}{C_e}$  yields the slope as  $\frac{1}{q_m}$

and intercept  $\frac{1}{q_m \cdot K_L}$ . The Freundlich isotherm can be denoted by Equation (5).

$$\log q_e = \log K_f + \frac{1}{n} \log C_e \tag{5}$$

A linear plot of  $\log C_e$  versus  $\log q_e$  yields the slope as  $\frac{1}{n}$  and intercept  $\log K_f$ .

Adsorption isotherm defines the estimation of absorption capacity of the adsorbent materials and is thus essential in the analysis for wastewater treatment. Figures 9 and 10 represent COD reduction for Langmuir and Freundlich isotherms. Figures 11 and 12 illustrate NH<sub>3</sub>-N reduction for both isotherms using the media at the optimum of adsorption parameters obtained. The correlation coefficient and constant from Equations (3)–(5) are presented in Table 4. The measure of the exponent 1/n indicates the favorability of adsorption.

Values of 1/n approaching 1 signify favorable adsorption, whereas values higher than 1 mean unfavorable adsorption. The constant K<sub>L</sub> is related to magnitude of adsorption energy and K<sub>f</sub> the adsorption capacity [29-31]. The correlation coefficients R<sup>2</sup> demonstrate or differentiate suitability of each equation. The correlation value obtained from the Langmuir and Freundlich equations for COD shows that the value of R<sup>2</sup> was higher in Langmuir than in Freundlich. Similarly, the R<sup>2</sup> values for NH<sub>3</sub>-N were higher in Langmuir (Table 4). This finding indicates that the Langmuir isotherm model is more suitable than Freundlich isotherm for evaluating the adsorption equilibrium required for COD and NH<sub>3</sub>-N uptake by the media in contrast, the study by [30] indicated that Freundlich model is suitable in the case for CS. This a monolayer on surface of the adsorbent. result also implies that the molecules adsorbed formed

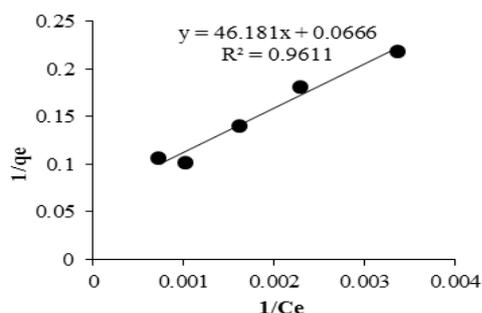


Figure 9. Langmuir isotherm for COD adsorption

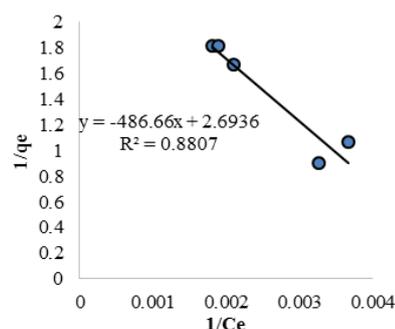


Figure 11. Langmuir isotherm for NH<sub>3</sub>-N adsorption

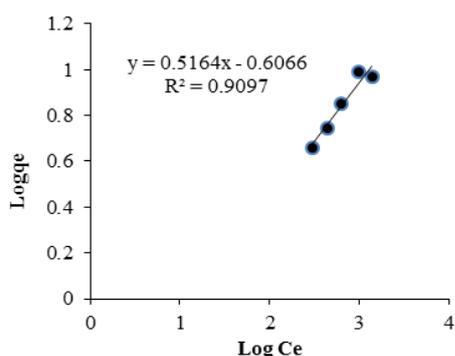


Figure 10. Freundlich isotherm for COD adsorption

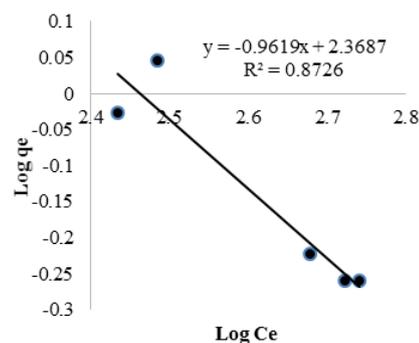


Figure 12. Langmuir isotherm for NH<sub>3</sub>-N adsorption

TABLE 4. Coefficients of Langmuir and Freundlich isotherm models for COD and NH<sub>3</sub>-N uptake using the mixed media

	Langmuir constant			Freundlich constant		
	q <sub>m</sub>	K <sub>L</sub>	R <sup>2</sup>	logK <sub>f</sub>	1/n	R <sup>2</sup>
COD	46.181	0.0666	0.9611	0.6066	0.5164	0.9097
NH <sub>3</sub> -N	-486.66	2.6936	0.8807	2.3687	0.9619	0.8726

#### 4. CONCLUSIONS

In this study, the capability of the mixture of CS and GAC as adsorbents was investigated for the removal of ammoniacal nitrogen and COD from a stabilized landfill leachate. The functional parameters including optimum mixing ratio, shaking speed, shaking time, pH, and adsorbent dose influenced the adsorption efficiency of the mixed media. The ideal conditions were established as follows: mixing ratio of 20:20, shaking speed of 150 rpm, pH level of 6, shaking time of 120 min, and dosage of 32 g. The adsorption isotherm analysis reveals that the Langmuir isotherm yielded the best fit to experimental data as compared with the Freundlich isotherm. The mixed media can be used as a good and economical adsorbent for stabilized landfill effluent treatment. The current findings add to the growing body of literature on alternative to conventional media for leachate treatment. Additionally, research is in progress to determine other factors such as desorption and disposal of exhausted adsorbents that may affect the field of study

#### 5. ACKNOWLEDGEMENTS

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## Batch Study on COD and Ammonia Nitrogen Removal Using Granular Activated Carbon and Cockle Shells

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دفع زباله‌های شیلات حاوی موادی تولید می‌کنند که غلظت بالایی از آلاینده‌ها دارند که برای سلامتی انسان و اکوسیستم خطرناک است. در این مطالعه، مخلوطی از کربن فعال گرانول و پوسته کوکول برای بازسازی COD و آمونیاک از محلول زباله تثبیت شده بررسی شد. تمام ذرات مواد جاذب تا اندازه‌ی بین ۲،۰۰ و ۳،۳۵ میلی متر غربال شدند. نسبت مخلوط بهینه، سرعت تکان دادن، زمان تکان دادن، pH و دوز تعیین شد. نتایج نشان می‌دهد که شیلات با غلظت COD بالا (۱۷۶۳ میلی گرم در لیتر)، نیتروژن آمونیاک (۵۷۳ میلی گرم در لیتر) و نسبت BOD5 / COD (۰،۰۹) است. نسبت مخلوط مطلوب کربن فعال و کربن فعال ۲۰:۲۰، سرعت تکان دادن ۱۵۰ دور در دقیقه، سطح pH 6، زمان تکان دادن ۱۲۰ دقیقه و دوز ۳۲ گرم بود. تحلیل ایزوترم جذب نشان می‌دهد که ایزوترم لانگمویر بهترین نتایج داده‌های آزمایشی را نسبت به ایزوترم فروندلیچ به دست می‌دهد. این عوامل واسط نتایج دلگرم کننده تولید می‌کنند و می‌توانند به عنوان یک جاذب خوب و اقتصادی استفاده شوند.

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