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# Comparison study of COD and ammoniacal nitrogen adsorption on activated coconut shell carbon, green mussel (*Perna viridis*), zeolite and composite material in stabilized landfill leachate treatment

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## ABSTRACT

Ammoniacal nitrogen and organic constituent (COD) are two crucial problematic parameters in leachate wastewater treatment. To achieve the objectives of this research, isotherm parameters for adsorption of COD and ammoniacal nitrogen by activated coconut shell carbon, green mussel (*Perna viridis*), zeolite and newly composite mass were investigated. All the adsorption materials were crushed and sieved to obtain the desired particle size of 150 μm. The optimum batch adsorption study condition was reported at 120 min contact time, 200 rpm shaking speed and pH 7. The result shows that all the adsorbents were well fitted with two different models namely Langmuir and Freundlich isotherm model whereas the coefficient of determination ($R^2$) value is greater than 0.9 for COD. A comparison research study indicates that the adsorption ability of composite adsorbents shows a higher tendency reduction of both COD and ammoniacal nitrogen than granular activated carbon, green mussel and zeolite. Generally, the results of the findings revealed that the Langmuir adsorption model was slightly better fitted and suitable for organic constituents while Freundlich was good for ammoniacal nitrogen. Langmuir adsorption coefficient of determination ($R^2$) for COD are 0.9968, 0.9914, 0.9944 and 0.9991 and Freundlich adsorption coefficient of determination ($R^2$) for ammoniacal nitrogen are 0.9514, 0.7136, 0.9568 and 0.9667, respectively.

*Keywords:* Activated coconut shell carbon; Green mussel shell; Zeolite; Composite adsorbent; Ammoniacal nitrogen and chemical oxygen demand (COD); Leachate wastewater

## 1. Introduction

Organic constituent (COD) and ammonia nitrogen both are problematic parameters for leachate treatment. The biological methods used for the treatment of leachate are quite effective to get rid of organic matters in the early stage [1] when the ratio of $BOD_5$/COD is relatively high in the leachate. Along with the age of landfill, this ratio of $BOD_5$/COD is decreasing [2] as well as the time of this process, which is becoming less efficient [3] because organic matter

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is present in the refractory. The high ammonia nitrogen concentration is often considered to inhibit the biological degradation by the microorganism [4,5]. The treatment of new leachate is usually easier and effective as compared to the older one [6]. Generally, biological processes are favorable for treating the leachates with a high $BOD_5$/COD ratio (biodegradability index). Such processes, however, cannot treat effectively mature landfill leachate that mainly contains recalcitrant organic matter and high strength of ammonia [7]. Due to the high ammonia concentration, the biodegradation processes and its organic compound tend to decreases thus, with the passage of time the landfills stabilized. Consequently, the biological treatment process effectiveness is also decreasing, therefore the physicochemical treatment process is an appropriate choice for stabilized landfills. The common future characterization of stabilizing leachates with a high concentration of ammoniacal nitrogen and a moderate value of COD might be ranged between (3,000–5,000 mg $L^{-1}$) and (5,000–20,000 mg $L^{-1}$) with the ratio of biodegradability less than 0.1 ($BOD_5/COD < 0.1$) [8,9].

Adsorption is one of the physicochemical processes commonly used for removing recalcitrant and toxic organic compounds from landfill leachates. Generally, adsorption method of mass transferring in which the substances transferred between fluid stages to the solid stage and then bounded through physical and chemical interaction. Adsorption utilized either commercial activated carbon or different adsorbent, for example, activated alumina, zeolite or inexpensive adsorbent namely clay, limestone, and micro-peat has been examined for the treatment of water, wastewater and landfill leachate [7]. Activated carbon adsorption has recently received significant attention to the removal of inorganic and organic pollutants from the landfill leachate, due to its large surface area, microporous structure adsorption, surface reactivity and its inherent physical properties [10].

Zeolite is commonly used as an excellent adsorbent for the reduction of ammoniacal nitrogen and inorganic pollutant from the landfill leachate as well as from other wastewaters. Generally, activated carbon does not have the adsorption ability to adsorb enough ammonia, as it typically has a nonpolar surface due to the high manufacturing temperature conditions, which is a drawback for certain applications because of poor interactions with certain polar adsorbate [11].

For this reason, a lot of research works have focused on altering the surfaces of activated coconut (AC) and to produce composite adsorbents that can either interact with a polar or nonpolar adsorbate. Gao et al. [12] found a new zeolite–carbon composite material that combined the best property of zeolites and carbon. Zeolite surface is hydrophilic, regularly aligned the level of molecules pore and the potential for cation exchange, making it the best adsorbents for catalysts and metal ions [13]. However, the carbon surface is hydrophobic, the level of pore sizes within or above the nanometers range making it better suited for adsorption in organic substances [14]. Previously, the combination of AC, zeolite and low-cost material namely peat, clay, rice husk waste, limestone and ordinary Portland cement (OPC) were used to produce a composite adsorbent media. A most recent one was utilized as a binder [15].

Alharbi et al. [16] described the nanomaterial with high surface areas, active site and an abundance of the functional group may form large surface complexes with various types of pollutants thus effectively remove the pollutant from aqueous solution. The author mainly focused on recent studies about synthesized nanomaterial and application for effective removal of various inorganic and organic pollutants from wastewater. The adsorption and removal of photocatalytic organic pollutants and reduction/sorption of heavy metal ions are considered as the main method to minimize pollutant concentration in the natural environment. Thus, this study analysis highlighted new novel nanomaterial applications in environmental pollution.

Liu et al. [17] described graphitic carbon nitrides ($g\text{-}C_3N_4$) with moderate bandgap (2.7 eV), high chemical and thermal stability, which has been a hotspot in environmental photocatalysis. The adsorption performance was insufficient and unsatisfactory due to inadequate visible lights, lower surface area, poor electronic conductivity and higher recombination rate of photo-generated electron–hole pair. These problems could be overcome by modifying $g\text{-}C_3N_4$ to enhance the properties of photo-catalytic. Among the different modifying techniques, doping elements are an effective and easy technique for modifying the electronic structures and accelerate photocatalytic performances. Thus, this study highlighting the typical designing trend and progress, cost-effectiveness elements doped carbonized nitrogen and degradation ability of environment organic pollutant.

This study aimed to compare the adsorption efficiency of the new types of composite adsorbents toward COD and ammoniacal nitrogen.

## 2. Materials and method

### *2.1. Composite adsorbent preparation*

To develop a new composite adsorbent media with the combination of hydrophobic media such as activated coconut shell carbon (ACSC) with a low-cost waste material namely green mussel (GM); (*Perna viridis*), and hydrophilic media zeolite (ZE). These adsorbents are comprised of ACSC and green mussel at 62.5% (by weight, g) and zeolite at 37.5% (by weight, g), and the rest 30% is OPC which was used as a binder for the preparation of composite adsorbent. Adsorbent material was then grounded into particle size in powder form <150 µm diameter by using a ceramic ball mill and then mixed with OPC. Almost 60% of water by weight was added then the pasted mixture was left to harden for the next 24 h. After, it was submerged into water for 3 d for curing purposes. These composite materials were crushed and sieved to get the desired working particle size of (1.18–2.36 mm). The physicochemical properties of the composite adsorbent are presented in Table 1 [7].

Prior to the experiment, the chemical composition of ACSC, green mussel, and zeolite was determined by an X-ray fluorescence spectrometry instrument. Table 2 shows the properties of ACSC, GM, and ZE respectively.

### *2.2. Leachate sample*

This research study was carried out in Johore where the leachate sample was taken from Simpang Renggam

Table 1
Physicochemical characteristics of composite adsorption media

| Physicochemical properties of composite media | |
|---|---|
| Specific gravity (g cm$^{-3}$) | 2.45 |
| Brunauer–Emmett–Teller surface area (m$^2$ g$^{-1}$) | 95.95 |
| Porosity (%) | 57.40 |
| Water absorption (%) | 52.62 |
| Methylene blue number (mg g$^{-1}$) | 7.97 |
| Iodine number (mg g$^{-1}$) | 21.93 |
| Cation exchange capacity (meq g$^{-1}$) | 0.80 |
| $pH_{ZPC}$ | 11.25 |

Table 2
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 |
| $CH_2$ | 98.52 | $Al_2O_3$ | 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 | | | |

Landfill Site (SRLS). The SRLS was situated at 1° 53″41.64″ latitude N and 103° 22″34.68″ longitude E in Kluang, Johore, Malaysia. The SRLS covers a total area of about 6 acres and possesses one of the easiest processes used for the treatment of landfill leachate which is an aerated lagoon (or aerated pond). Currently, SRLS has now been operated for 12 y and has approximately 250-ton of solid waste collected on a daily basis. It also covers 3-areas which are Kluang, Simpang Renggam and Batu Pahat [18].

These leachate samples were obtained from SRLS and were filled in a 30 L high-density polyethylene plastic container. The experimental study procedure and analysis were carried out according to the standard procedure of the APHA [19]. Therefore, the collected samples were transported to the research laboratory and kept at room temperature under 4°C. The chemical analysis was conducted the next following 2 d, based on the standard method for examining water and wastewater. All chemicals used for the analytical determination were of analytical grade. The initial physicochemical characterization of the obtained landfill leachate samples from the stabilized landfill site (Kluang, Johore, Malaysia) are presented in Table 3 [20]. The pH value exceeds 5 as well as the value of $BOD_5$/COD ratio, which is less than 0.1 where it is surpassed the standard leachate discharge is considered as stable leachate [21].

### 2.3. Batch adsorption experiments

Static batch experiments were performed at ambient temperatures to optimize reduction by the adsorbent using optimal conditions of all relevant factors, that is, agitation speed, pH, contact time, and adsorbent dosage. Subsequently, the only optimization of the process parameters is conducted for the adsorption experiment. The optimal condition used in the adsorption batch experiment of all relevant parameters such as pH 7, 200 rpm and 120 min of contact time taken from the previous research studies [8,22–25].

Adsorption isotherm test was conducted on a mixture of reactions consisted of 100 mL of leachate wastewater solution by varying the adsorbent weight. The closed reflux colorimetric method and nesslerization method were used to determine the organic constituents (COD) and ammoniacal nitrogen respectively [19].

## 3. Results and discussion

### 3.1. Adsorption equilibrium

Langmuir and Freundlich's models are usually applied to describe the adsorption processes between sorbate and sorbent molecules in an aqueous solution. In the work, isotherm models such as Freundlich and Langmuir were tested to access the sorption process. According to the Langmuir model; all the accessible sorption sites are homogeneous and morphologically uniform [26,27]. Langmuir model equation is given by;

$$q_e = \frac{QbC_e}{\left(1+bC_e\right)} \tag{1}$$

Whereas the equation can be described in another linear standard form;

$$\frac{1}{q_e} = \frac{1}{Q} + \frac{1}{QbC_e} \tag{2}$$

where $C_e$ represents the adsorbate concentration at equilibrium (mg L$^{-1}$), $q_e$ represents the equilibrium adsorption amount (mg g$^{-1}$), coefficient $Q$ represents the maximum adsorption capacity (mg g$^{-1}$), $b$ represents Langmuir constants that is related to the adsorption energy (L mg$^{-1}$). Langmuir isotherm constants were determined from the slope and intercept of the plot of ($1/q$) vs. ($1/C_e$).

Freundlich model assumes that uptake adsorbate occurs on heterogeneously surfaces by utilizing multilayers adsorption and increasing the adsorbing amount

Table 3
Physicochemical characteristics of the raw leachate wastewater collected from Simpang Renggam Landfill Site

| Parameter | Minimum | Maximum | Average | MEQA (1974) |
|---|---|---|---|---|
| COD (mg L$^{-1}$) | 1,595 | 1,829 | 1,712 | 400 |
| Ammoniacal nitrogen (mg L$^{-1}$) | 404.07 | 406.68 | 405.37 | 5 |
| pH | 7.65 | 8.27 | 7.96 | 6.0–9.0 |
| SS (mg L$^{-1}$) | 316 | 367 | 341.5 | 50 |
| Color (Pt-Co) | 4,685 | 4,788 | 4,736.5 | 100 |
| BOD$_5$/COD | 0.07 | 0.08 | 0.07 | – |
| BOD$_5$ (mg L$^{-1}$) | 140 | 163 | 138.66 | 20 |

continuously with an increase of concentrations [28]. The Freundlich equation is given as:

$$q_e = K_F C_e^{1/n} \tag{3}$$

where $K_F$ indicates the adsorption capacity, $1/n$ shows the adsorption intensity. The equation can be described in another linear standard form;

$$\log q_e = \log K_F + \frac{1}{n}\log C_e \tag{4}$$

where $K_F$ and $n$ are the Freundlich constants, the characteristics of the system. $K_F$ and $n$ are the indicators of the adsorption capacity and adsorption intensity, respectively. The exponent magnitude value $1/n$ provides an indication of the adsorption favorability of the system. The values of exponent greater than 1, as presented in Table 4 [29].

Table 4 illustrates the result data of the COD and ammoniacal nitrogen adsorption experiment where it shows that the Langmuir model gives a better fit as compared to the Freundlich model. It showed a higher coefficient of determination and adsorption capacity. The adsorption equilibrium for COD and ammoniacal nitrogen adsorption onto the zeolite, green mussel, activated carbon, and composite adsorbent is indicated in Table 4. Thus, findings also indicated that adsorption was better fitted represented by mono-layer coverage of COD and ammoniacal nitrogen onto the adsorbent as shown in Table 4.

### 3.2. Organic constituents

Fig. 1 indicates that ACSC was the most efficient for organic constituents (COD) reduction closely follows by the composite while green mussel and zeolite adsorbent were comparatively the lowest. Comparison of Isotherms constant and coefficient of determination between ACSC,

Table 4
COD and ammonia isotherm constant for activated coconut shell carbon, green mussel, zeolite and composite

| Isotherm (Langmuir) COD | Activated coconut shell carbon | Green mussel | Zeolite | Composite |
|---|---|---|---|---|
| $R^2$ | 0.9968 | 0.9914 | 0.9944 | 0.9991 |
| $Q$ | 555.56 (mg g$^{-1}$) | 243.90 (mg g$^{-1}$) | 250 (mg g$^{-1}$) | 2,000 (mg g$^{-1}$) |
| $b$ | 0.027 (L g$^{-1}$) | 0.571 (L g$^{-1}$) | 1.06 (L g$^{-1}$) | 0.651 (L g$^{-1}$) |
| $q_e$ | $54.04C_e/1 + 0.0270C_e$ | $142.75C_e/1 + 0.571C_e$ | $588.90C_e/1 + 1.06C_e$ | $158.78C_e/1 + 651C_e$ |
| Isotherm (Langmuir) NH$_3$–N | | | | |
| $R^2$ | 0.9963 | 0.9909 | 0.9974 | 0.9982 |
| $Q$ | 90.90 (mg g$^{-1}$) | 166.67 (mg g$^{-1}$) | 200 (mg g$^{-1}$) | 208 (mg g$^{-1}$) |
| $b$ | 10 (L g$^{-1}$) | 16 (L g$^{-1}$) | 0.175 (L g$^{-1}$) | 3.793 (L g$^{-1}$) |
| $q_e$ | $2,000C_e/1 + 10C_e$ | $3,333.34C_e/1 + 16C_e$ | $29.32C_e/1 + 0.175C_e$ | $344.878C_e/1 + 3.793C_e$ |
| Isotherm (Freundlich) COD | | | | |
| $R^2$ | 0.9965 | 0.8116 | 0.8202 | 0.9974 |
| $K_F$ | 15,980.85459 | 4,027,170.343 | 6,520,786.785 | 3,379,091.874 |
| $1/n$ | 0.7937 | 0.3411 | 0.1709 | 0.3362 |
| $q_e$ | $15,980.85C_e^{0.7937}$ | $4,027,170.342C_e^{0.3411}$ | $6,520,786.785C_e^{0.1709}$ | $3,379,091.874C_e^{0.3362}$ |
| Isotherm (Freundlich) NH$_3$–N | | | | |
| $R^2$ | 0.9514 | 0.7136 | 0.9568 | 0.9667 |
| $K_F$ | 276,312.1614 | 269,649.7364 | 2.30E+51 | 85,921.134 |
| $1/n$ | 0.1281 | 0.1431 | 8.0567 | 0.2995 |
| $q_e$ | $276,312.1614C_e^{0.1281}$ | $269,649.7364C_e^{0.1431}$ | $2.30E+51C_e^{8.0567}$ | $85,921.134C_e^{0.2995}$ |

green mussel, zeolite, and composite are represented in Table 4. ACSC has the advantage of adsorbed organic pollutants from the leachate. Also, it has a high surface area, large pore-size distributions, and hydrophobic surfaces. The same finding result revealed that powdered activated carbon (PAC) is more effective in removing COD at 5 g $L^{-1}$ of dosage than zeolite, while zeolite has better-removing efficiency for ammonia as compared to (PAC) at 1 g $L^{-1}$ dose [6]. Generally, activated carbon adsorption application is ideal for GAC or PAC and was significantly more efficient for reduction of the nonbiodegradable compound from landfill leachate wastewater, but not suitable for ammoniacal nitrogen [30].

### 3.3. *Ammoniacal nitrogen*

Fig. 2 apparently indicates that the reduction percentage of ammoniacal nitrogen compared between ACSC, green mussel, zeolite, and composite by varying dose. The result describes that zeolite media and composite do not show any difference in removing ammoniacal nitrogen, but obviously much better than ACSC and green mussel. The isotherm constants and coefficient of determination for adsorption of ammoniacal nitrogen from stabilized leachate to ACSC, green mussel, zeolite, and composite are described in Table 4. The composite media was the highest in terms of ammoniacal nitrogen adsorption capacity (208 mg $g^{-1}$), follows by zeolite (200 mg $g^{-1}$), green mussel (166.67 mg $g^{-1}$) whereas ACSC (90.90 mg $g^{-1}$) was the lowest. These findings results demonstrated that ACSC doesn't have sufficient adsorption capacity for ammoniacal nitrogen. Particularly, it possesses a nonpolar surface due to the preparation at higher temperature conditions, which is a disadvantage for some applications due to poor interactions with some polar adsorbate [11]. Due to its high cation exchangeability as well as the physical adsorption mechanism, zeolite is a well-known adsorbent used for removing ammonium ions from an aqueous solution.

However, physical adsorption is dominants for activated carbon due to its wide ranges of pore size available on its surfaces. Moreover, zeolites are hydrophilic surfaces and have a greater affinity to ammonium ion and ammoniacal nitrogen in an aqueous solution compared to the hydrophobic surfaces on activated carbon. Both the models (Langmuir isotherm and Freundlich isotherm) were acceptable for all media based on the higher value of *R*-square (>0.9).

The values of isotherm model parameters for the reduction of COD and ammoniacal nitrogen are presented in Table 4. The model parameters are directly related to the variations in the system properties. The result shows the best fitting models and the relative parameters determined for the reduction of COD and ammoniacal nitrogen by the adsorbent. The table also shows that the Langmuir model was in accordance with experimental data for the reduction of contaminants investigated with ($R^2$) values of 0.9968, 0.9914, 0.9944 and 0.9991 for COD and ($R^2$) for ammoniacal nitrogen are 0.9514, 0.7136, 0.9568 and 0.9667 respectively. Therefore, the Langmuir model was more appropriate to be used to describe the process than the Freundlich model as shown in Figs. 3–10, respectively.

## 4. Conclusion

In this research study, a combination of ACSC, green mussel, and zeolite as an adsorption composite media providing both hydrophobic and hydrophilic surfaces for the reduction of organic and inorganic contaminants especially COD and ammoniacal nitrogen. The static batch experiment condition was found at 120 min contact time, 200 rpm shaking speed, and pH 7 respectively. The findings of the comparative analysis indicate that organic constituents (COD) adsorption was the best on composite 2,000 mg $g^{-1}$, follows by ACSC 555.56 mg $g^{-1}$, zeolite 250 mg $g^{-1}$, and green mussel 243.90 mg $g^{-1}$. The results were different for ammoniacal nitrogen where zeolite shows the best adsorption capacity 208 mg $g^{-1}$, follows by the composite 200 mg $g^{-1}$, green mussel 166.67 mg $g^{-1}$, and ACSC 90.90 mg $g^{-1}$.

A comparison research study indicates that the adsorption ability of granular activated carbon and composite adsorbent shows a higher tendency reduction of organic constituents (COD) and ammoniacal nitrogen than green mussel, and zeolite. Generally, the findings results revealed that the Langmuir model adsorption was slightly better fitted and suitable for organic constituents (COD) and Freundlich was good for ammoniacal nitrogen reduction in terms of coefficient of determination ($R^2$). Langmuir adsorption coefficient of determination ($R^2$) for COD are 0.9968, 0.9914, 0.9944, and 0.9991 and Freundlich adsorption

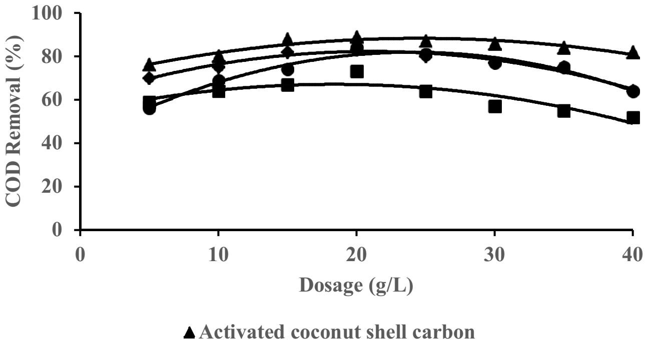

Fig. 1. Removal percentage of COD using activated coconut shell carbon, green mussel, zeolite and composite media as adsorbent.

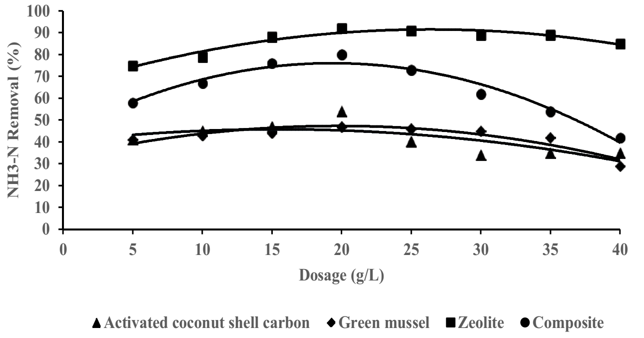

Fig. 2. Removal percentage of $NH_3$–N using activated coconut shell carbon, green mussel, zeolite and composite media as adsorbent.

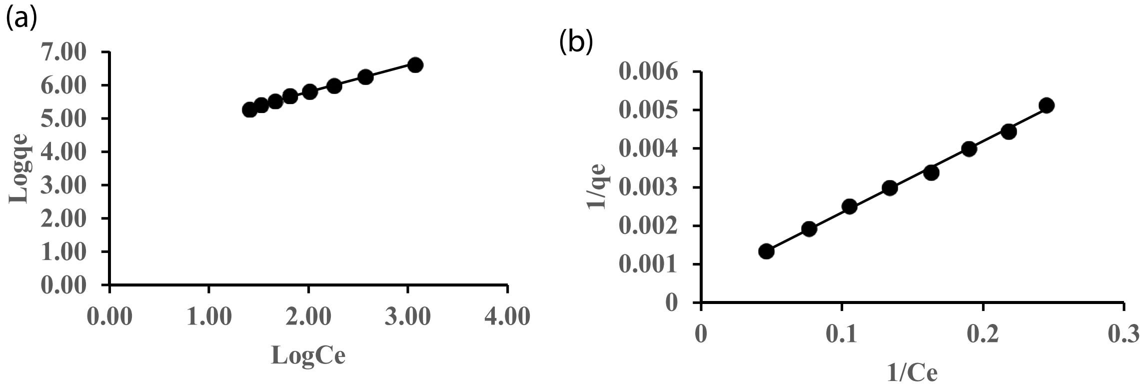

Fig. 3. ACSC: isotherm models for COD (a) Langmuir and (b) Freundlich.

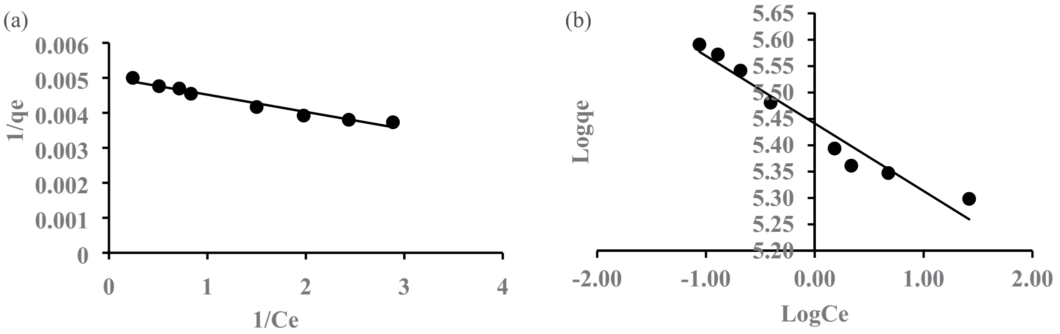

Fig. 4. ACSC: isotherm models for $NH_3$–N (a) Langmuir and (b) Freundlich.

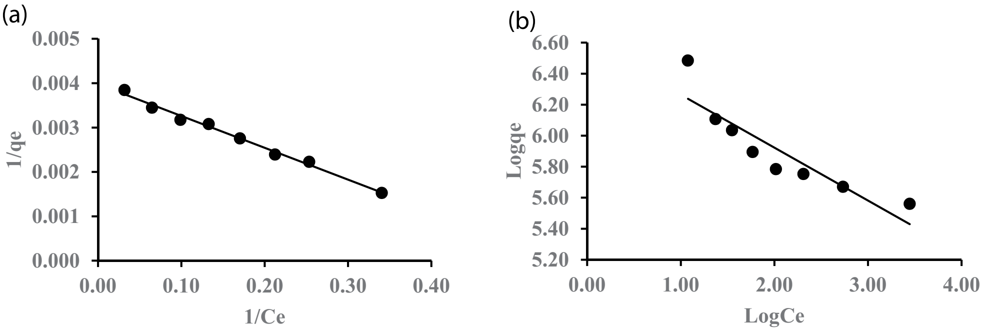

Fig. 5. GM: isotherm models for COD (a) Langmuir and (b) Freundlich.

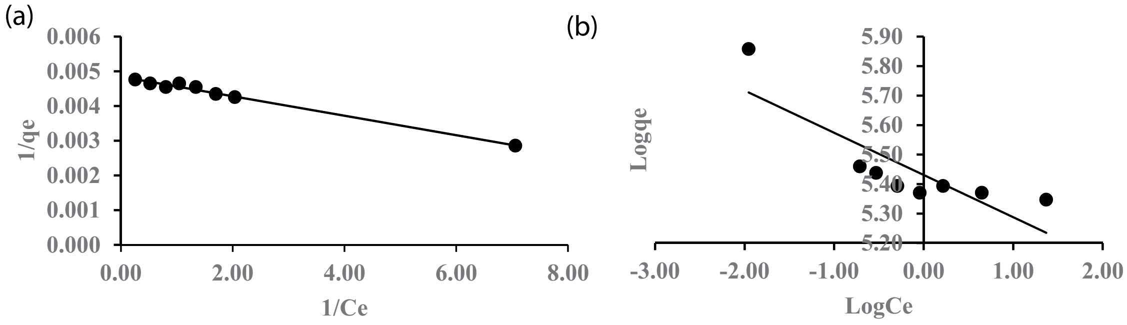

Fig. 6. GM: isotherm models for $NH_3$–N (a) Langmuir and (b) Freundlich.

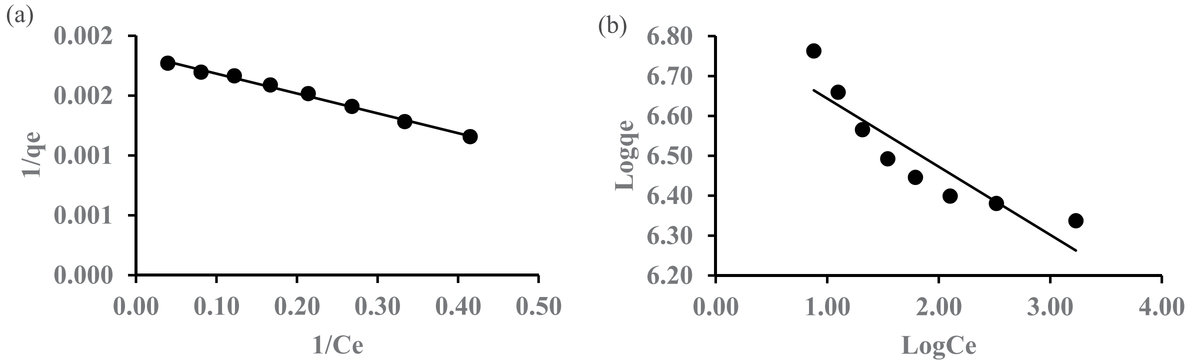

Fig. 7. ZE: isotherm models for COD (a) Langmuir and (b) Freundlich.

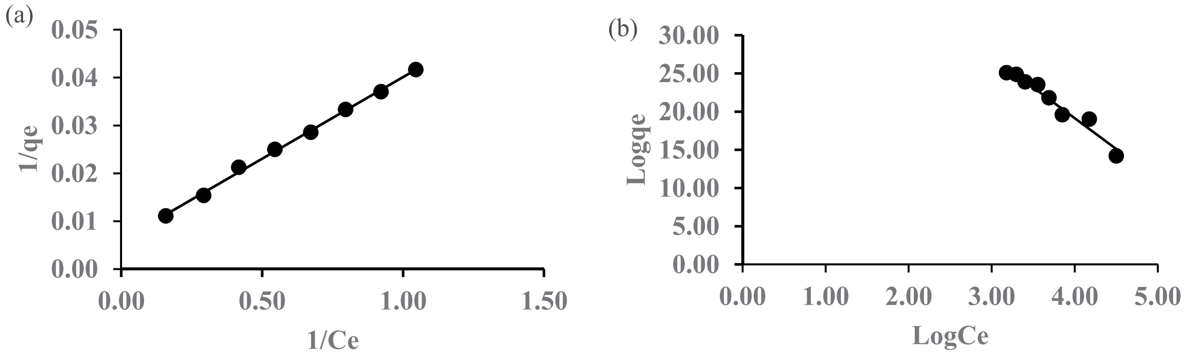

Fig. 8. ZE: isotherm models for $NH_3$–N (a) Langmuir and (b) Freundlich.

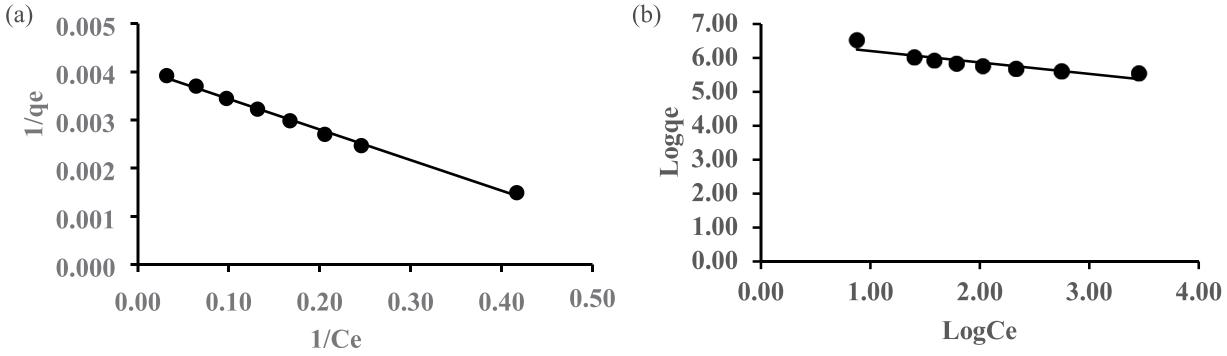

Fig. 9. COMP: isotherm models for COD (a) Langmuir and (b) Freundlich.

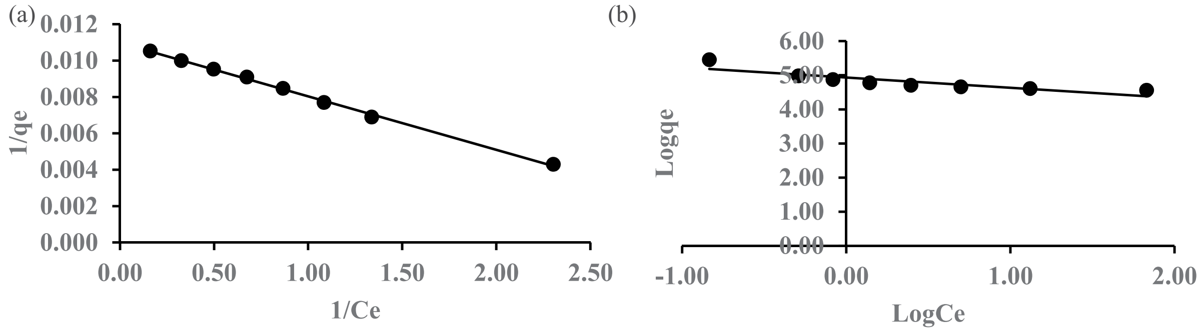

Fig. 10. COMP: isotherm models for $NH_3$–N (a) Langmuir and (b) Freundlich.

coefficient of determination ($R^2$) for ammoniacal nitrogen are 0.9514, 0.7136, 0.9568, and 0.9667 respectively. Therefore, it is proposed that kinetic adsorption be taken into account for further research to examine the organic constituents (COD) and ammoniacal nitrogen process onto the ACSC, green mussel, zeolite, and composite media respectively.

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