

Adsorptive Removal of Mn(II) ions from Aqueous Solution on Activated Carbon

Produced from Bambara Nut Shells Using Physical Activation with Steam

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ABSTRACT

Activated carbon derived from Bambara nut shell (BSAC) by physical activation with steam was used for the removal Mn(II) ions from aqueous solution. The result of physicochemical properties of the adsorbent indicated that the percentage yield, moisture and ash contents were $24.59 \pm 1.24\%$, $0.17 \pm 0.06\%$ and $8.1 \pm 0.01\%$ respectively. The value of pH and pHPzc were 8.25 ± 0.05 and 7.5 respectively. Batch adsorption studies showed that the uptake of Mn(II) ions onto BSAC was dependent on the initial adsorbate concentration, contact time and pH of the solution. The maximum uptake capacity at 60 mg/L was 173.13 mg/g. Adsorption isotherm studies revealed that Langmuir isotherm model had higher correlation coefficients, R^2 (0.968) compared to Freundlich isotherm (0.379) indicating that the uptake of Mn(II) ions onto BSAC showed better fitting to Langmuir adsorption isotherm. The value of separation factor, R_L (0.005) indicates favourable adsorption process. The study concluded that adsorbent derived from agricultural waste by steam activation could be used for the removal of heavy metals from aqueous solution.

Keywords: Activated carbon, Adsorption, Bambara nut, steam

INTRODUCTION

Industrial wastewater containing harmful substances like heavy metals resulted in rapid increase in environmental pollution. Heavy metals are non-biodegradable materials which occur naturally in the earth crust as a result of natural and man-made activities and usually persist in the environment and enter into the body of living organisms leading to severe threats to human and other organisms [1]. The concentrations of these metals in the industrial wastewaters are usually higher than the permissible discharge limits. Therefore, appropriate

treatments before releasing them to the environment become necessary [2]. Adsorption technique using agricultural based adsorbents has been the most effective and commonly used method for removing wide range of organic and inorganic pollutants from aqueous solution [3].

Activated carbon is an amorphous or crystalline material with high degree of porosity and large surface which can be produced from wide varieties of agricultural waste materials [4]. The most commonly used methods of preparation are physical and chemical methods. Physical activation is a two-step process that involves carbonization (pyrolysis) of the starting material in a neutral atmosphere at a high temperature followed by activation of the carbonized material by using steam, carbon dioxide or air mixtures [5]. The advantage of this method is that it does not require the use of chemicals and can effectively produce an activated carbon with a good porous structure and it requires less washing, but high reaction temperature is required [6, 7]. In chemical activation, the raw material is usually impregnated using oxidizing and highly dehydrated chemicals. After impregnation, the mixture is allowed to dry and the remaining mixture will be heated at temperatures ranging from 400 – 900 °C depending on the type of the precursor material [8].

Activated carbon has been widely used for the removal of pollutants from aqueous solutions produced activated carbon from maize cob for effective removal of manganese(II) and cobalt(II) ions from aqueous solution [1]. Activated carbon derived from rice husk ash was used for the removal of manganese and iron from aqueous solution [9]. Nharingo, et al [10] reported adsorption isotherm studies on the biosorption of Cu(II) ions using low cost adsorbent derived from *Vigna Subterranea* (L.) *Verdc* hull.

In the present study, agricultural waste derived from Bambara nut (*Vigna Subterranea* (L.) *Verdc*) shell was utilized to produce less expensive adsorbent via physical activation with steam and its adsorption performance for the removal of manganese(II) ions from aqueous solution was investigated using batch adsorption experiments.

MATERIAL AND METHODS

Sample collection

The Bambara nut shells were collected from Dawakin- Kudu Town, Dawakin- Kudu Local Government Area Kano State, Nigeria, with geographical coordinates of 11°50'05''N 8°35'53''E. The shells were washed with distilled water to remove all adhered materials and other impurities.

Preparation of Bambara nut shell activated carbon by steam activation

A 40 g of the sample was loaded in a hollow quartz glass tube and placed in a tubular reactor furnace under flow of N₂ gas at a flow rate of 750 mL/min reactor for 30 minutes. The sample was then pyrolysed at 600 °C for 30 min in the presence of N₂ at flow rate of 1000 mL/min and heating rate of 20 °C/min. The resulting char was physically activated under the presence of nitrogen/steam mixture that was achieved via nitrogen supply with flow rate of 750 mL/min through a heated water bath kept at constant temperature of 90±2 °C to final temperatures of 600 °C for 30 min. The activated carbon sample was cooled inside quartz tube under flow of nitrogen/steam mixture, washed with de-ionized water and dried at 105 °C for 24 hours [5, 11].

Preparation of Mn(II) stock solution

A 1000 mg/L of Mn(II) stock solution was prepared by dissolving 2.88 g of KMnO₄ in a small quantity of de-ionized water in a 250 mL beaker and then transferred into a 1000 mL standard flask and was made up to the mark with de-ionized water. Other lower concentrations were prepared by serial dilution of the stock solution [12].

Batch adsorption experiments

The effect of initial concentration was investigated by contacting 25 mL solutions of the adsorbate at varying concentration from 10 to 80 mg/L in 100 mL conical flask with a fixed amount of the adsorbent (0.01 g). The effect of contact time from 5 to 120 minutes at optimum concentration was investigated while the effect of pH of the solution was conducted between pH 3 to 6. The mixtures were agitated for 4 hours in a mechanical shaker at room temperature. The mixtures were filtered and the residual filtrates were analysed using atomic absorption spectrophotometer (AAS), Buck Scientific model 210 VGP [13, 14]. The amount of metal ions adsorbed onto BSAC was determined using Eq. 1:

$$Q_e = \frac{(C_0 - C_e)V}{M} \quad \text{Eq. (1)}$$

Where Q_e (mg/g) is the adsorption capacity, C_0 (mg/L) is the initial concentration of the adsorbate in the solution, C_e (mg/L) is the equilibrium concentration of the adsorbate in the solution, V (L) is the volume of the solution used in the flask and M (g) is the amount of adsorbent used.

Adsorption isotherms study

Langmuir isotherm

The Langmuir isotherm model is based on the assumptions that the adsorption takes place in the form of monolayer and the general expression of Langmuir isotherm is given by Eq. 2:

$$\frac{C_e}{q_e} = \frac{1}{K_L q_m} + \frac{C_e}{q_m} \quad \text{Eq. (2)}$$

Where C_e is the equilibrium concentration of adsorbate (mg/L), q_e is the amount of the adsorbate adsorbed per gram of the adsorbent at equilibrium (mg/g), q_m is the maximum adsorption capacity to complete monolayer coverage on the adsorbent surface (mg/g) and K_L is the Langmuir constant related to the energy of adsorption (L/mg). The values of q_m and K_L can be obtained from the plot of $\frac{C_e}{q_e}$ against C_e [15].

The value of separation factor R_L , Eq. 3 is very useful in predicting whether the adsorption process is favourable or not.

$$R_L = \frac{1}{1 + K_L C_0} \quad \text{Eq. (3)}$$

Where R_L is the separation factor, C_0 is the initial concentration (mg/L). When the R_L value is greater than 1 it indicates unfavourable adsorption, when R_L value equal to 1, the adsorption is linear. R_L value between 0 and 1 it indicates favourable adsorption and if R_L value is equal to 0, the adsorption is irreversible [16].

Freundlich Isotherm

Freundlich isotherm model is based on the assumption that the adsorption takes place on a heterogeneous surface. The general expression of Freundlich isotherm is given by Eq. 4.

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \quad \text{Eq. (4)}$$

Where C_e is the equilibrium concentration of the adsorbate (mg/L), q_e is the amount of adsorbate adsorbed per unit mass of adsorbent (mg/g), K_F (mg/g) and n (L/mg) are Freundlich constants representing the adsorption capacity and intensity of adsorption respectively. The values of n and K_F can be obtained from the slope and intercept of the plot of $\log q_e$ against $\log C_e$. When the value of $1/n$ is below 1, it indicates a favourable normal adsorption at low

dilution while if the value of $1/n$ is above 1 it indicates cooperative adsorption or unfavourable adsorption [17].

RESULTS AND DISCUSSION

The physicochemical properties of BSAC are presented in Table 1.

Table 1: Physicochemical properties of BSAC

Properties	BSAC
Yield (%)	24.59±1.24
Moisture content (%)	0.17±0.06
Ash content (%)	8.1±0.01
Bulk density (g/cm ³)	0.33±0.006
Volatile matter (%)	31.30±0.12
Fixed carbon (%)	60.43
pH	8.25±0.05
pHpzc	7.5

The percentage yield of BSAC was 24.59±1.24%, lower than the reported value (41.4 - 58.6%) required for industrial [18]. Percentage yield of 38.4±2.35% has been reported [19]. The moisture content is the amount of water present in the activated carbon and its presence has no significant influence on the adsorption performance, but high moisture content in the sample usually results in additional amount of adsorbent during adsorption process [20]. The value of moisture content of BSAC was 0.17±0.06%, lower than the reported value of 14.37% obtained from rubber-seed shell activated carbon by physical activation with steam [21].

Ash content gives information about inorganic constituents present in the sample and low ash contents leads to good adsorption capacity [22]. The ash content of BSAC was found to be 8.1±0.01%. Ash content of 41.15% has been reported from steam based activated carbon derived from black liquor lignin [23].

The bulk density value of BSAC was 0.33±0.006 g/cm³, which is comparable to the reported value (0.37±0.01 g/cm³) obtained from activated carbon derived from maize cob [1]. Bulk density value of 0.45 g/cm³ has been reported elsewhere in the literature [24]. Volatile matter is the amount of combustible material present in the raw sample and the value obtained was 31.30±0.12%, lower than the reported value of 40.34% [23]. The fixed carbon represents the total combustible residue that is left in the sample.

The amount of carbon residue (60.43%) in BSAC was relatively high compared to the reported value of 13.97% [21].

The pH is the degree of acidity or alkalinity in the activated carbon, and the value of pH of BSAC was 8.25 ± 0.05 indicating that the pH of the adsorbent is alkaline in nature which could be due to dissociation of the hydroxyl functional group on the surface of the adsorbent when dissolved in water [25]. pH value of 5.97 has been reported in the literature [23].

The pH_{pzc} is the pH at which the total charge of the adsorbent is zero. The pH_{pzc} of BSAC was 7.5, lower than its corresponding pH value. At pH greater than pH_{pzc} , the surface of the adsorbent is negatively charged and this favours the adsorption of cations [26]. pH_{pzc} value of 6.8 has been reported elsewhere in the literature [1].

Adsorption studies

Effect of initial concentration

The result of the uptake of Mn(II) ions onto BSAC is shown in Fig. 1. It could be observed that as the concentration increased from 10 to 60 mg/L, there was a corresponding increase in the uptake capacity of Mn(II) ions onto BSAC. The maximum adsorption capacity was 173.12 mg/g. The increase in uptake capacity with increase in concentration could be due to the availability of metal ions for binding with active sites of the adsorbent. But after attaining the equilibrium concentration, there was a decrease in adsorption capacity with increase in concentration which could be due to saturation of the available binding sites. A similar trend has been reported in the literature [27].

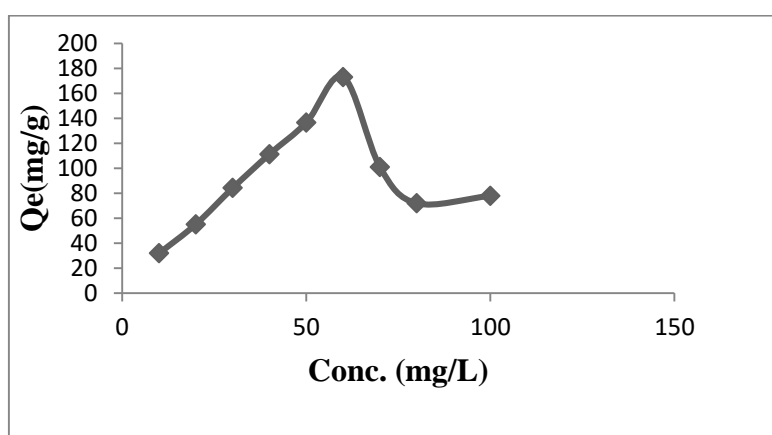


Fig. 1: Effect of initial concentration on the uptake of Mn(II) ions onto BSAC

Effect of contact time

The influence of contact time on the uptake of Mn(II) ions onto BSAC is shown in Fig. 2. It was observed that the adsorption of Mn(II) ions onto BSAC was very rapid at the initial stage as a result of vacant binding sites on the surface of the adsorbent. [28]. The equilibrium was reached at 5 minute with uptake capacity of 101.42 mg/g and then a decrease in uptake capacity was observed with increase in contact time after attaining equilibrium which could be due to saturation of available binding sites. A similar observation has been reported [28].

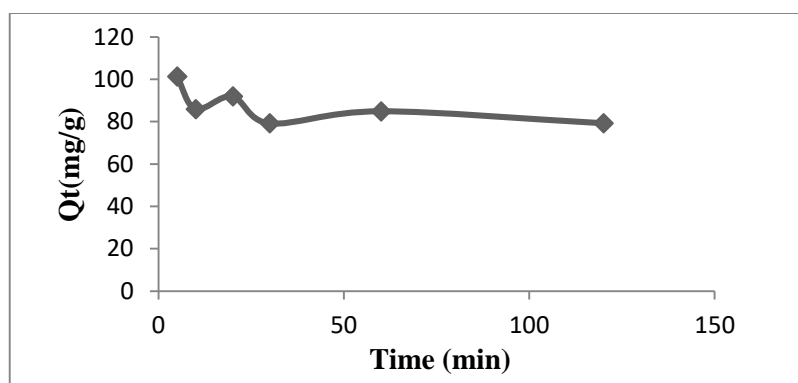


Fig. 2: Effect of contact time on the uptake of Mn(II) ions onto BSAC

Effect of pH

The pH of the solution strongly affects the uptake capacity. The maximum uptake capacity, 91.25 mg/g for the uptake of Mn(II) ions onto BSAC was reached at pH 3 as shown in Fig. 3. At low pH, there is availability of positive charges in the solution and the pH_{pzc} of the adsorbent was found to be lower than its corresponding pH thereby making the surface of BSAC negatively charged and thus enhance strong attraction between surface and the protons in the solution which eventually results in increase in uptake capacity. A similar observation has been reported in the literature [9, 29].

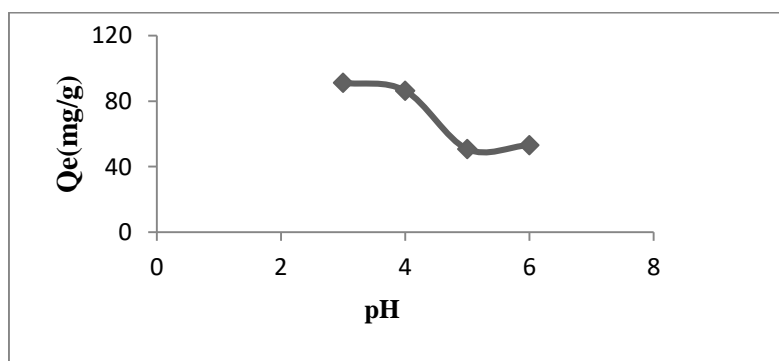


Fig. 3: Effect of pH on the uptake of Mn(II) ions onto BSAC

Effect of adsorbent dosage

The influence of adsorbent dosage on uptake of Mn(II) ions is shown in Fig 4. It was observed that the uptake capacity decreases with increase in adsorbent dosage in which the highest uptake capacity corresponding to 65.65 mg/g was achieved using 0.01 g of BSAC adsorbent. The decrease in the uptake capacity with increase in adsorbent dosage could be attributed to the reduction of surface areas of the adsorbent as a result of overlapping of the various adsorption sites [30].

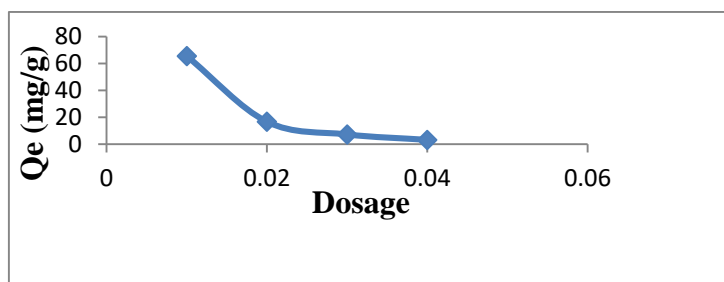


Fig. 4: Effect of contact time on the uptake of Mn(II) ions onto BSAC

Adsorption isotherms

The results of Langmuir and Freundlich adsorption isotherm plots are shown in Figs. 5 and 6 while the value of various parameters are presented in Table 2. The Langmuir isotherm plot showed a good correlation coefficient, R^2 value of 0.968 (Fig. 5) while that of Freundlich adsorption isotherm as shown in Fig. 6 showed very poor R^2 value of 0.379 which indicates that the adsorption of Mn(II) ions onto BSAC showed better fitting to Langmuir isotherm compared to Freundlich adsorption isotherm model and the linearity of the plot with R^2 value close to unity suggest the applicability of the model implying that the adsorption process is in the form of monolayer coverage [31]. The value of Langmuir monolayer coverage was 0.014 mg/g. The value of separation factor, R_L was 0.005 and fell between 0 and 1, indicating that the adsorption process was favourable [32].

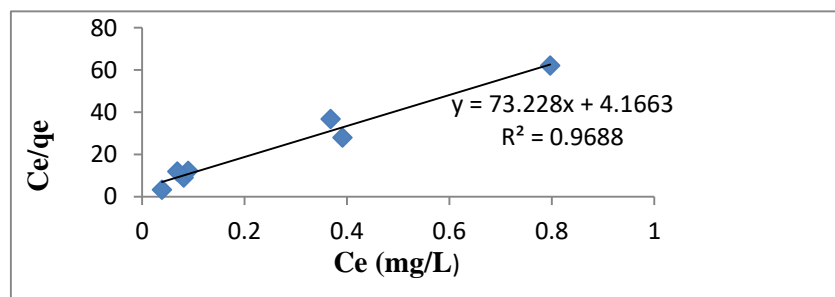


Fig. 5: Langmuir plot on the uptake of Mn(II) ions onto BSAC

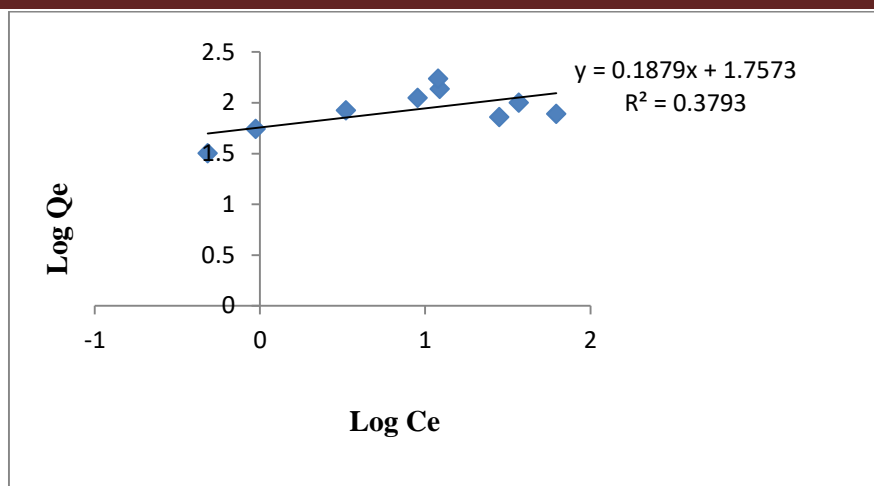


Fig.6: Freundlich plot on the uptake of Mn(II)ions onto BSAC

Table 2: Adsorption isotherm parameters on the uptake of Mn(II) ions onto BSAC.

Isotherm	Parameter	Value
Langmuir	Q_m (mg/g)	0.014
	K_L (L/mg)	17.15
	R_L	0.005
	R^2	0.968
Freundlich	K_F (mg/g)	57.19
	n	5.26
	$1/n$	0.19
	R^2	0.379

CONCLUSION

Adsorptive performance of activated carbon obtained from Bambara nut shells using physical activation method with steam has been investigated for the removal of Mn(II) ions from aqueous solution. The results of the physicochemical properties indicated that the percentage yield, moisture and ash content of the produced adsorbent were very low. The adsorption of Mn(II) ions onto BSAC were found to be dependent upon the initial concentration, contact time as well as the pH of the solution. Adsorption isotherm studies revealed that the uptake of Mn(II) ions was best described by Langmuir adsorption isotherm model and the adsorbent could be utilised for the treatment of heavy metals from aqueous solution.

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