

## Removal of Benzene from Oil Spill Wastewater Using Activated Carbon Developed from Watermelon Rind by Chemical Activation with $H_2SO_4$

Korie, C. I. Afolabi, E.A. Kovo, A. S. Muktar, A. Abdulkareem, A. S.

Department of Chemical Engineering, Federal University of Technology, Minnakoriechiemeka@gmail.com, elizamos2001@yahoo.com, Kovo@futminna.edu.ng Email: koriechiemeka@gmail.com

### Abstract

This study focused on developing activated carbon from watermelon rind for the removal of benzene from oil-spilled wastewater. The activated carbon was produced from biochar, which was produced from watermelon rind. The activated carbon was produced via chemical activation using sulphuric acid. The influence of process parameters like time, temperature, and impregnation ratio was investigated during the preparation of biochar and activated carbon respectively. The adsorbent was characterized Brunauer-Emmett-Teller (BET) analysis to determine the surface area, pore volume and pore diameter. The activated carbon was used in batch adsorption on oil wastewater. The effect of contact time, adsorbent dosage, and temperature on the adsorption process was studied and optimized using response surface methodology. The results showed that the yield was lowered when the carbonization temperature and impregnation ratio was increased. Maximum specific surface area of the activated carbon of  $900m^2/g$  was obtained at  $700^\circ C$  with activation duration of 15 minutes and at an impregnation ratio of 2/1. The results of the adsorption showed that benzene removal largely depended on the operating parameters, as the highest removal efficiency for benzene was recorded as 51% at optimal temperature, time, and adsorbent dosage parameters. This method of adsorption removal is economical as it utilizes the use of agricultural waste.

**Keywords:** Activated-carbon, Benzene, Impregnation-ratio, Rind, Watermelon

### 1. INTRODUCTION

EXPLORATION and oil activities have provided problem in the form of oil spills. Crude oil consists of a lot of unstable aromatic hydrocarbons (including benzene, toluene, ethyl benzene and the isomers of xylene known as BTEX), polycyclic aromatic hydrocarbons (PAHs), phenols and alkylated phenols, organic acids, radio nuclides, and metals (Dorea *et al.*, 2007).

Aromatic compounds, including mono aromatic hydrocarbons such as benzene, are amongst the significant aromatic contaminants of polluted water. Human exposure to benzene can lead to health that could affect the external organs like skin, eyes, and mucus membranes. Exposure can also affect internal organs like the nervous system, cause cancer as it is listed as a carcinogenic, and reduce bone marrow function. The levels of benzene drinking water as stated by the United States Environmental Protection Agency should be at a maximum of  $0.005\text{ mg L}^{-1}$  (benzene) (Costa *et al.*, 2012). Benzene is soluble in water and is unstable at room temperature, which means that once benzene is ingested it easily gets into the blood via the pulmonary vessels. Once the pulmonary vessels encourage the transportation of benzene in the system, it is easy for this hydrocarbon fraction to get absorbed in the lungs (Barton, 2014).

According to Ghannam and Chaalal (2003), there are different ways that oil spills can removed; skimmers, controlled burning, dredging, oil booms, dispersants, and vacuum techniques are some of the

ways to remove these spills. However, using sorption techniques to clean-up oil spills is considered as the best option, especially when dealing with mono aromatic hydrocarbons. Adsorption using activated carbon from biomass is an interesting approach to tackling hydrocarbon fractions. This is why a lot of research on the development of eccentric materials such as agricultural waste materials for remediating oil spillage is ongoing. Utilizing biomass is advantageous because these products are easily sourced, inexpensive; eco- friendly and can have high removal efficiencies because of functional groups like hydroxyl and carboxylic groups for the case of benzene removal (Qiu et al., 2008). It is important to tackle the challenges brought about by oil spillage, especially with regards the removal of soluble petroleum fractions, and it is even more important to discover and implement cost-effective and sustainable substitute technologies for bioremediation. Activated carbon as an adsorbent is more popular than other adsorbents among researchers, especially when being used for water treatment. Commercial grade adsorbents are usually expensive, and this increases with the increase in quality of the adsorbent. Small-scale industries are focusing on inexpensive ways to develop activated carbon; this explains the move towards agricultural waste. In order to consider biomass as activated, it has to show a large internal surface area and porous structure. Activated carbon has high levels of carbon mixed with other elements as stated by Lakshmipathy and Sarada (2013). Activated carbon is formed using a combination of pyrolysis or carbonization of the raw material and activation using an activating agent. Pyrolysis involves converting biomass into gas, oils, or biochar using temperature or pressure as determining constraints in an inert environment. To create this inert environment, nitrogen gas or argon gas is used. Other factors that are useful in determining interaction between biomass and the pyrolysis environment are the gas flow rate, and retention time (Oghenejoboh, 2018). Watermelon rind (WR) has components of phenols, cellulose, citrulline, proteins, and carotenoids. The polymers discovered in watermelon are copious in functional groups like hydroxyl (cellulose) and carboxylic (pectin) and can easily bind metal and treat the compounds for oil pollution (Queket et al., 2007). For a precursor to have good adsorption results, the adsorbent should have high content levels of oxygen and carbon. Some of the needed physical characteristics for a good precursor are small pores, which ultimately leads to more surface area for adsorption, thermal stability, and high abrasion resistance (Sabir, 2015).

In order to develop these physical characteristics, it is important to focus on how certain experimental parameters (temperature and time) affect elements like pore volume, surface area, and pore diameter during the production of activated carbon (Gin et al., 2014). Oghenejoboh (2018) carried out a research using watermelon rinds to develop activated carbon, which was then used to adsorb nickel (ii) ion from synthetic wastewater. This was done using response surface methodology (RSM) optimization approach. The aim of this research is to investigate the influence of temperature and time on biochar via pyrolysis, to investigate the influence of using this biochar to produce activated carbon by using an activating agent, while varying impregnation ratio and to test the removal efficiency of the activated carbon on benzene in oil-spilled wastewater.

## 1. METHODOLOGY

### 2.1 Material and Chemical Reagent

Watermelon rinds (WR) were collected from a fruit market in Federal university of technology, Minna, Niger State, Nigeria. The chemical precursor for the production of activated carbon from WR was sulphuric acid ( $H_2SO_4$ ). Benzene ( $C_6H_6$ , MW: 78.11 gmol<sup>-1</sup>) was the pollutant used to test the effectiveness of the WR activated carbon. The chemical reagent was analytical grade (AR) as supplied and it was utilized without any additional purification.

### 2.2 Development of Biochar and Activated Carbon

Fifty gram (50g) of the precursor was weighed and put in pyrolysis equipment. Nitrogen gas was purged into the reactor to create inert condition. Experimental conditions of approximately; time (2-4hrs) and temperature (400 - 600°C) were varied using response surface methodology. The influence of the parameters temperature and time were studied by using the response surface methodology (CCD design). The flow rate of nitrogen gas and the heating rate were held at 150 cm<sup>3</sup>/min and 10 °C/min, respectively. The dry char was kept in an airtight container awaiting further use after the carbonization process. The preliminary calculations for the response surface for biochar production are as seen in Table 1:

**Table1.** Preliminary Calculation for CCD for a Response Surface

| Parameters       | -α    | Lower(-1) | Midpoint(0) | Upper limit(+1) | +α    |
|------------------|-------|-----------|-------------|-----------------|-------|
| Temperature (°C) | 416.5 | 450       | 500         | 550             | 583.5 |
| Time(hr.)        | 2.165 | 2.5       | 3           | 3.5             | 3.835 |

The biochar was activated via chemical activation method using sulphuric acid which was poured into beakers containing the activated carbon and stirred until a paste was formed. The impregnation ratio (IR) used for this was 0.5/1, 1/1, 2/1, 3/1 and 4/1 and can be calculated by Eq. (1):

$$IR = \frac{W_1}{W_2} \quad (1)$$

Where, W1 is the dry weight in (g) of the activating agent (sulphuric acid) and W2 is the dry weight in (g) of the char

The char was mixed with sulphuric acid, placed in an oven, and activated at 700° C for about 15 mins. The samples was allowed to cool to room temperature, washed with distilled water, and soaked in 1% NaOH solution to remove any remaining acid. The sample was dried in an air oven at 105 °C for 2 hrs according to Kobya *et al.* (2005) and Madhumitha *et al.* (2016).

The char yield after the carbon has been activated was determined using Eq. (2):

$$\%yield = \frac{W_1}{W_2} * 100 \quad (2)$$

Where, W1 is the samples' weight after pyrolysis and W2 is the samples' weight before pyrolysis.

## 2.3 Raw Material Analysis

The physical properties of WR, bio char and activated carbon were determined via proximate analysis according to Gin *et al.* (2014).

### 2.3.1 Moisture Content

Three grams (3 g) of the carbonized precursor was put in a clean, dry petri-dish and dried in an oven at 105°C for 90 mins. The precursor was cooled in a desiccator for 30 mins. The samples' weight was determined before and after heating. The percentage moisture content was calculated using Eq. (3):

$$Moisture\ content = \frac{(W_1 - W_2) \times 100}{W_1} \quad (3)$$

Where, W1 = samples' weight before drying, g and W2 = samples' weight after drying, g

### 2.3.2 Ash Content

Three grams (3 g) of watermelon peel was weighed in a crucible and oven dried at 400 °C for 1 hr. The precursor was cooled in a desiccator and the weight was determined. The ash content was calculated with Eq. (4):

$$\frac{W_1}{W_2} \quad (4)$$

Where W1 = weight of the ash and W2 = weight of the dry precursor

### 2.3.3 Volatile Matter Content

Four grams (4 g) of activated carbon was put in a crucible and oven dried at a temperature of 450°C for 10 mins. The sample was weighed before and after drying to determine the amount of volatile matter as shown in Eq. (5):

$$\text{Percentage volatile content} = \frac{(W_1 - W_2)}{W_1} \times 100 \quad (5)$$

Where, W1 = weight of precursor before drying (g), and W2 = weight of precursor after drying (g).

### 2.3.4 Fixed Carbon Content.

This was gotten using Eq. (6):

$$100 - (\text{Ash content} + \text{volatile matter content} + \text{moisture content}) \% \quad (6)$$

## 2.4 Characterization of Activated Carbon

The Brunauer - Emmett- Teller (N<sub>2</sub> BET) method was used to determine the pore volume, surface area, and pore diameter of the activated carbon produced from watermelon rinds. The equipment used was a NOVA 4200e surface area and pore analyzer instrument. 100 mg each of the activated carbon was introduced into the equipment with flowing N<sub>2</sub> at 90 °C for 1 hr, and then held at 350 °C for 2 hrs.

## 2.5 Batch Adsorption for Benzene Removal

Batch adsorption experiments were carried out by placing activated carbon of varying dosages of about 0.3 - 1 g in 100 mL conical flasks with 10 mL of oil spill wastewater. The flasks were tightly covered to avoid likely losses of the aromatic compounds because of volatilization that can happen during experimentation. These covered flasks were placed in thermostatic shake water bath which was shaken at an agitation rate of 200 rpm for different agitation times ranging from 95-145 mins. The influence of varying constraints such as temperature, adsorption dosage, and contact time were investigated via response surface methodology, to determine optimal conditions for the removal of benzene. The benzene concentration in solution before and after the adsorption was determined using UV spectrophotometer using the wavelength 252 nm for benzene. The calibration curves for benzene were designed by diluting the solution to several known concentration and documenting their absorbance.

The removal efficiency was calculated using Eq. (7):

$$\text{Removal efficiency (\%)} = \frac{C_o - C_t \times 100}{C_o} \quad (7)$$

Where C<sub>o</sub> is the initial concentration and C<sub>t</sub> is the concentration at time t in (mg/L). The preliminary calculations for the response surface methodology are as seen in **Table 2**.

**Table 2.** Preliminary Calculation for CCD for a Response Surface

| Name             | Units | Minimum | Maximum | Coded Low   | Coded High  |
|------------------|-------|---------|---------|-------------|-------------|
| Temp             | C     | 19.89   | 45.11   | -1 ↔ 25.00  | +1 ↔ 40.00  |
| time             | min   | 94.77   | 145.23  | -1 ↔ 105.00 | +1 ↔ 135.00 |
| Adsorbent dosage | g     | 0.3636  | 1.04    | -1 ↔ 0.50   | +1 ↔ 0.90   |

## 1. RESULTS AND DISCUSSION

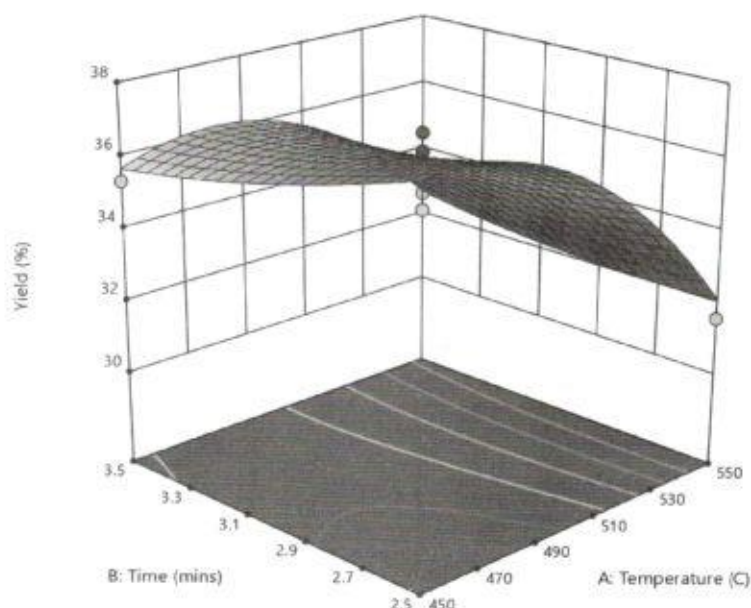
### 3.1 Influence of Temperature and Time on Psychochemical Properties of Carbon Yield.

Temperature and time were the parameters used to produce biochar from activated carbon. The temperature and time were determined from response surface methodology (RSM) (central composite design). This section focuses on how these process parameters affect certain elements like carbon yield. ANOVA for the model is discussed in this section. The biochar yield reduced as temperature and time increased. For example, at 450°C and 2.5hrs, the carbon yield was 37.46% but dropped to 31.56% at 550°C and 2.5 hrs. It was discovered that temperature affected the carbon yield more than time, as there was a massive drop in carbon yield 30.22% at the highest temperature 583.5°C and time of 3 hrs. It was observed, that while the biochar yield was dropping, the volatile matter was affected in a directly proportional manner, while the fixed carbon was affected in an inversely proportional manner; this is due to the decomposition of biomass during the pyrolysis process and the release of volatile matter. These results correspond to Junna *et al.* (2016). Response surface methodology analysis for CCD gave the Eq. (8):

$$\text{Yield} = +36.03 - 215A - 0.3677B + 0.6675AB - 1.70A^2 + 0.2426B^2 \quad (8)$$

Design-Expert Software

3D Surface



**Figure1:** A plot of temperature and time vs. Yield for WRBC

From the ANOVA results, the model is significant at Model F-value of 30.88 (as seen in Figure 1). There is only a 0.01% chance that an F-value this large could occur due to noise. This model is significant because it has P-values less than 0.0500. In this case A, A<sup>2</sup> is significant model terms. Values greater than 0.1000 indicate the model terms are not significant. The model has a Lack of Fit F-value of 2.36 which means that the Lack of Fit is not significant comparative to the pure error. There is a 21.24 % chance that a Lack of Fit F-value this large could occur due to noise. Non-significant lack of fit is good. The model gave a predicted R<sup>2</sup> of 0.7784 and an adjusted R<sup>2</sup> of 0.9257. These two values agree with each other, as the difference of the R<sup>2</sup> values is less than 0.2.

### 3.2 Pore Development and Characteristic of Activated Carbon

Activated carbon was developed via chemical activation using an activating agent (sulphuric acid) and biochar from watermelon rind as the precursor. The impregnation ratio was varied and the influence of impregnation ratio on pore volume, surface area, and pore diameter and on the activated carbon was studied. It was discovered that the impregnation ratio that produced activated carbon with the largest surface areas and pore volumes for the reagent sulphuric acid was at an IR of 2/1. As impregnation ratio increased from 0.5/1 to 2/1, the surface area of watermelon rind activated carbon (WAC) increased, but increasing impregnation ratio from 2/1 to 4/1 led to decrease the surface area of WAC. The surface area for the activated carbon reached a maximum of 900 m<sup>2</sup>/g when the impregnation 2/1 but reduced drastically to 447 m<sup>2</sup>/g at 3/1; this shows that as the impregnation ratio increased the surface area reduced. This increase in surface area might be as a result of degradation of the lignocellulose sample's matrix, which gives rise to more surface area, meanwhile, a reduction in surface area as the impregnation ratio increases could be as a result of shrinking of carbon in the activated carbon (Zakaria et al., 2021).

At low impregnation ratios, pore formation is dominating mechanism and pore widening becomes important with an increase in impregnation ratio. Therefore, pore volume values become higher until the impregnation ratio of 2/1. Then, the pore volume values become lower at the impregnation ratio of 3/1. The maximal value of the total pore volume of WRAC developed with an impregnation ratio of 2/1 is 0.84 cm<sup>3</sup>/g. The pore volume drastically reduced at 0.382 cm<sup>3</sup>/g when the impregnation ratio was increased to 3/1. The reduction in pore volume as the IR increases shows that mesopores open when temperature is utilized in conjunction with the IR for activation. The entities of the activating agent become more voluminous, accentuating further the hydrolysis and thus the swelling of the activating material. It appears that at higher acid concentrations the generation of meso-pores and macro-pores occurred at the expense of micro-pores. This is in line with Angin (2014).

**Table 3** shows the influence of varying impregnation ratios on the surface area pore volume and pore diameter for activated carbon produced with sulphuric acid. In the column for the pore diameter, it can be observed that there was an increased until the impregnation ratio of 1/1 at 3.000 nm but was reduced to 2.940 nm at IR of 2/1.

**Table 3.** Influence on impregnation ratios on surface area, pore volume and pore diameter.

| IR    | Surface area (m <sup>2</sup> /g) | Pore volume (cm <sup>3</sup> /g) | Pore diameter (nm) |
|-------|----------------------------------|----------------------------------|--------------------|
| 0.5/1 | 475.120                          | 0.423                            | 2.960              |
| 1/1   | 530.422                          | 0.468                            | 3.000              |
| 2/1   | 900.361                          | 0.843                            | 2.940              |
| 3/1   | 477.203                          | 0.382                            | 2.760              |
| 4/1   | 574.505                          | 0.407                            | 2.140              |

### 3.2. Preliminary Analysis on Watermelon Rind, Biochar and Activated Carbon

**Table 4** shows the properties of raw watermelon rind (WR), biochar (BC) and activated carbon (WAC). Ash content (AC) has a big responsibility on the quality of activated carbon. Excess ash usually blocks the pores in the activated carbon, reducing the surface area of the activated carbon and ultimately affecting the adsorption efficiency of the activated carbon. The proximate analysis shows that the ash content for BC at 31.9% was higher than WR at 2% but the ash content for activated carbon at 13.05% was lower than BC at 31.9%. The higher content in BC than WAC and WR could be as a result of mineral salt formation during the pyrolysis process (Ultrasonik, 2017). High ash content is undesired in WAC processing because it represents amount of inorganic material from the precursor activation and a numerous treatment needed to take place. This is consistent with Ultrasonik (2017).

The results show that volatile matter was reduced from 60.07% for WR, to 27.3% for BC to 20.70% for WAR. Volatile matter (VM) for WR got lower during pyrolysis due to the decomposition of biomass and reduced even lower for activated carbon because the addition of activators causes modifications in the properties and structure of activated carbon.  $H_2SO_4$  as an activator causes degradation of organic matter which weakens the surface structure of activated carbon. In addition, this activator also releases volatile substances and develops an active carbon micro-pore structure. The activator is able to clean and improve the pore surface of AC by releasing non-carbon elements, especially oxygen, hydrogen, and nitrogen which is known as tar and gases. These results are in accordance to (Maulina, 2018).

**Table 4.** Proximate analysis of watermelon rind (WR), biochar and activated carbon (WAC)

| S/N | Property         | WR, (% wt) | Biochar, (% wt) | WAC, (% wt) |
|-----|------------------|------------|-----------------|-------------|
| 1   | Moisture Content | 15.38      | 1.4             | 5.8         |
| 2   | Volatile Matter  | 60.07      | 27.3            | 20.70       |
| 3   | Ash Content      | 4.00       | 31.9            | 13.5        |
| 4   | Fixed Carbon     | 20.55      | 39.4            | 60          |

Fixed carbon (FC) is the solid flammable matter that is around when biomass is heated. Volatile matter is usually removed during the process of combustion. Fixed carbon content is influenced by lignin and cellulose content in biomass as these elements can be transformed to carbon. Temperature and time affects fixed carbon levels during pyrolysis while activator concentration and temperature have effect on fixed carbon for activated carbon. The fixed carbon saw a tremendous increase from 22.55% for WR to 39.4 for BC and 60% for activated carbon. The increase in activated carbon is due to the lower volatile content and ash content. These results correspond to Junna *et al.* (2016).

### 3.4 Effect of Adsorbent Dosage on Adsorption Removal Efficiency

The adsorbent dosage is an important factor during adsorption studies as it provided the adsorption capacity of the adsorbent for a known starting concentration of the adsorbate. To determine their influence of adsorbent dosage on removal efficiency, different adsorbent dosages in the range of (0.3-1g) were used. The removal percentage of benzene increased as the adsorbent dosage increased as is seen in **Figure 2**. This could be because of the rise in adsorption sites. The peak removal efficiency was discovered at 0.7 g, and after this peak, any alteration in adsorbent dosage was insignificant because it attained equilibrium at the peak.

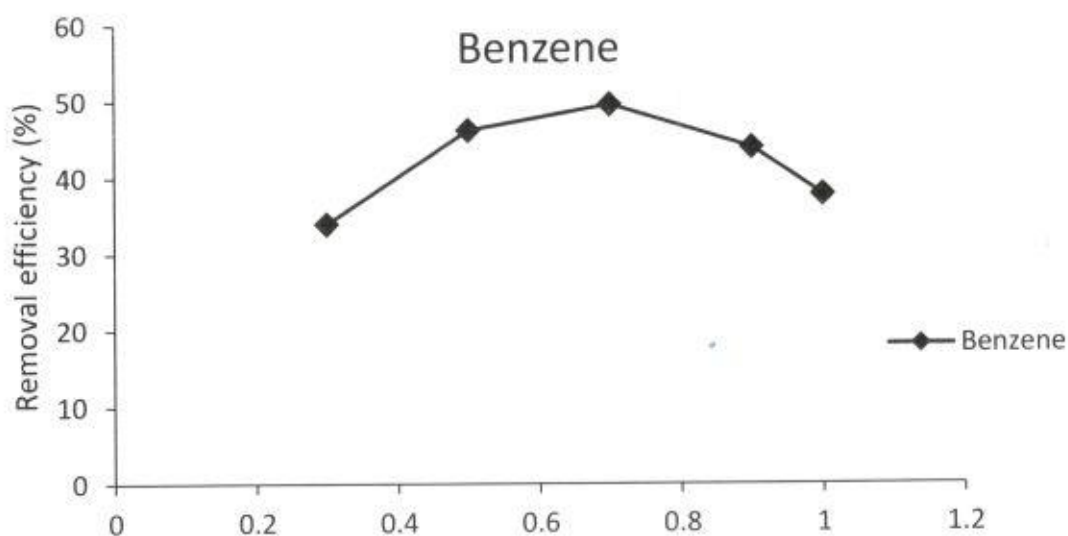


Figure 2: A plot of adsorbent dosage vs removal efficiency.

The reason for this could be that the active sites on the activated carbon are saturated with the pollutant at the peak adsorption dosage, such that an increase in adsorbent does not make any difference to the removal efficiency (Nourmoradi *et al.*, 2013).

### 3.5 Effect of Temperature on Adsorption Removal Efficiency

The effect of temperature on removal efficiency was studied for the removal of benzene in oil spill wastewater. Temperature ranging from about 25°C to 45°C was used to determine the removal efficiency of these soluble hydrocarbons. As seen in **Figure 3**, it was discovered that at lower temperatures, removal efficiency was higher than at higher temperatures with the peak removal efficiency at 51% for benzene.

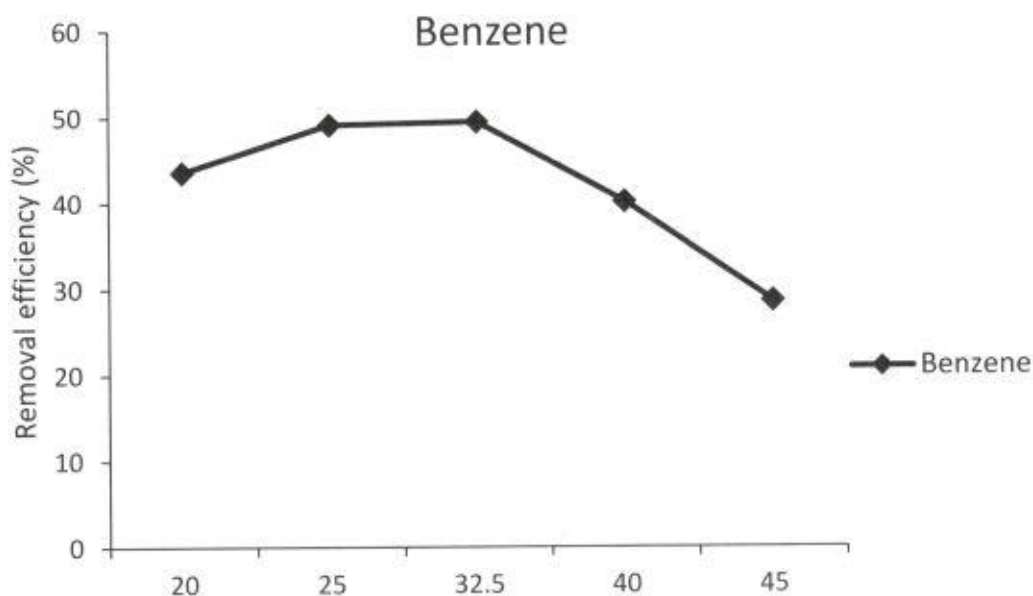


Figure 3: A plot of temperature vs removal efficiency.

The removal efficiency was drastically reduced after the peak was achieved. This result is in line with Nourmoradi *et al.* (2013).

### 3.6 Effect of Time on Adsorption Removal Efficiency

The adsorption efficiency of benzene using adsorbent from watermelon rinds showed that there was a rapid increase as contact time was increased. This increase might have something to do with how many available adsorption sites were available during the beginning of the experiment (Nourmoradi *et al.*, 2013). The contact time for the adsorbent on benzene was from 90-200 minutes approximately. The removal efficiency increases slowly up to about 120 minutes and after that, the removal efficiency was slowed down, this is shown in Figure 4.

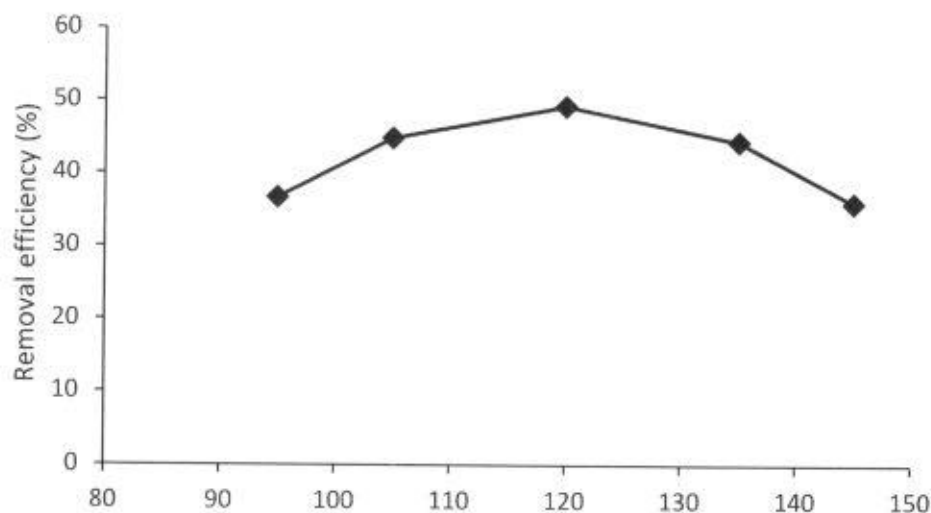


Figure 4: A plot of time vs removal efficiency.

## 4. CONCLUSION

The activated carbon prepared by chemical activation with  $H_2SO_4$  attained a maximum value of  $900 \text{ m}^2/\text{g}$  at a carbonization temperature of  $700^\circ\text{C}$  with activation duration of 15 mins and at an impregnation ratio of 2/1. It was discovered that as impregnation ratio increased, pore volume and surface area increased for the activating agent, until it reached equilibrium at impregnation ratio of 2/1, after which the surface area and volume reduced. The increase in the temperature and time decreased yield while increasing the fixed carbon content for the biochar. The use of chemical activation method for the watermelon rind improved fixed carbon content for the watermelon rind, while reducing the ash content.

The results of the adsorption showed that benzene removal largely depended on the operating parameter as the highest removal efficiency for benzene was 51% was recorded at optimal temperature, time, and adsorbent dosage parameters. The results obtained from this work, conclude that using watermelon rind as activated carbon for the adsorption of benzene shows promise.

## ACKNOWLEDGEMENT

This research was made possible through the facilities from Chemical Engineering Department of Federal University of Technology, Minna, Nigeria and support from an institution based research fund with grant number: TETFUND/FUTMINNA/2016-2017/6<sup>th</sup> BRP/02.

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