

# Biosorption and Kinetic Studies on Oil Removal from Produced Water Using Banana Peel

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## Abstract

Crude oil production is accompanied by generation of large volumes of produced water. The produced water contains both dissolved and dispersed oil and these pose health hazards to human health and contamination of the environment. In this work, banana peel, an agricultural waste was used to remove the oil component from produced water. The peel samples were dried, size-reduced and characterized for functional groups, surface morphology, elemental composition and surface area using FTIR, SEM, EDS, and BET analytical equipment respectively. Standard methods were employed for the production of biosorbent and removal of oil using batch process. The result of EDS spectroscopy revealed that the peel contained 88.52% C; 5.69% O; 2.19% Si; 2.80% K; and 0.80% Ca. Biosorption study with the peel showed that it can be used in the removal of oil (dissolved and dispersed oil) from produced water providing almost complete removal at an equivalent dosage of 267 mg banana peel per liter produced water after 35 minutes equilibrium time and initial oil concentration as high as 194 mg/l. Solution pH, loading rate, sorption time, and temperature effects were studied. Several biosorption kinetic models were tested and the result showed that banana peel biosorbent followed pseudo-second-order kinetics. Equilibrium isotherm studies on the other hand revealed that banana peel closely followed the Langmuir isotherm model.

**Keywords:** banana peel, produced water, biosorption, kinetic models

## 1.0 Introduction

Treatment of oil spills and oil contaminated water remains one of the major challenges to environmental scientists and technologists. Among the existing techniques used for oil treatment, sorption is a popular technique because it is cheap, simple and effective (Banerjee *et al.*, 2006). Among the various sorbents used, banana peel appears to be attractive material in terms of cost, versatility and abundance (Muhammad, 2012). Oil removal by biological waste materials have been reported by several authors [Khaled *et al.*, (2011); Lin *et al.*, (2011); Othman *et al.*, (2008); Pasila, (2004); Suni *et al.*, (2004); Tania *et al.*, (2000); Vlaev *et al.*, (2011); Banerjee *et al.*, (2006); Muhammad *et al.*, (2012) and Muhammad *et al.*, 2012]. During the 1970s increasing environment awareness and concern led to search for new techniques to replace expensive/hazardous chemical treatment methods. These demands had led to increasing interest in biosorption. The use of biosorbent has been proposed as an alternative, as it has proven effective on low concentrations.

The biosorption process involve a solid phase (sorbent or biosorbent; biological material) and a liquid phase (solvent, normally water) containing a dissolved species to be sorbed (sorbate, metal ions). Due to higher affinity of the sorbent to the sorbate species, the later is attracted and removed by different mechanisms. The process continues till equilibrium is established between the amount of the solid-bound sorbate species and its portion remaining in the solution. The degree of sorbent affinity for the sorbent determines its distribution between the solid and liquid phases. The major advantages of biosorption over conventional treatment methods include low cost, high efficiency, minimized chemical and biological sludge, and regenerability. Many workers have reported using biological waste materials to treat different pollutants ranging from metals to organic dyes found in waste water. For example, AjayKumar *et al.*, (2009) studied biosorption of Cadmium, Copper, Nickel, and Zinc in aqueous solution on activated sludge. Nowicki *et al.*, (2010) investigated the sorption properties of walnut shell while Suni *et al.*, (2004) utilized cotton grass fibre as sorbent for oil spills. Kaewsam *et al.*, (2008) used banana peel as sorbent in the removal of cadmium ion from aqueous solution. Kaur, (2011) used sugarcane bagasse for the removal of erythrosim B and methylene blue from aqueous solution. Mahmoud *et al.*, (2012) studied the kinetic of Ni (II) using activated clay minerals. Hasana *et al.*, (2010) investigated the selenium remediation through up-flow-fixed-bed column.

In this study, banana peel was conditioned and tested for oil uptake from produced water. Isotherms equilibria and kinetic studies on the data generated were carried out. Sorption of oil was made at various temperatures, bio-waste loading and sorption times. Characterization of the biosorbent was made using FTIR, EDS-SEM and surface area measurement using nitrogen adsorption technique by BET method.

## 2.0 Materials

Banana peels were collected from Muda Lawal market, Bauchi Nigeria. Crude oil was obtained from Kaduna Refinery and Petrochemical Company (KRPC), Kaduna-Nigeria. 1,1,1-tri-chloroethane was purchased from Chuzz Bond International, Jos-Nigeria. All chemicals and reagents were of analytical grade. Distilled water was obtained from Gubi Dam Water Treatment Plant Laboratory, Bauchi-Nigeria. Oven was used to dry the sorbent materials (manufactured by Regaterm, Italy). Separating funnels were used to extract out the oil from water and DR/2000 spectrophotometer (HACH, Colorado, U.S.A) was used to test the presence of oil in the extract. Hanna pH meter was used to determine the pH of the mixtures. A JJ-4 Six couplet Digital electric mixer (Search Tech Instrument, England) was used for the sorption study. Laboratory mortar and pestle were used to convert the banana peel to powder and sieves were used to classify it into different sizes (212-63 microns). Perkin Elmer Spectrum 100 FTIR spectrometer was used for the infra-red spectroscopic studies at wave numbers 4000-400  $\text{cm}^{-1}$ . The X-ray diffractometry was done on a BRUKER AXS D8 Advance (Cu-K $\alpha$  radiation  $\lambda K\alpha_1=1.5406\text{\AA}$ ) 40kV. The Hitachi X-650 Scanning Electron Microscope (Tungsten filament, EHT 20.00kV) and LEO 1450 Scanning Electron Microscope (Tungsten filament, EHT 20.00kV) were used for the SEM imaging. The chemical composition was determined using energy dispersive spectroscopy (EDS) and surface area and pore sizes were determined using TriStar 3000 V6.05 A BET equipment.

## 3.0 Method

### 3.1 Biosorbent Preparation

Banana peel was first washed with water several times and then sun-dried. The dried banana peel was ground to particle sizes between 2-3 mm and later treated with n-hexane to remove all coloured pigment in a reflux condenser. The n-hexane treated banana peel was later washed with distilled water and dried in an oven at 70°C for 24 hours. This was further crushed and sieved through 212-63 microns sieve and then stored in air tight sealed plastic containers as raw banana peel (RBP).

### 3.2 Characterization

The raw banana peel (RBP) biosorbent was characterized using FT-IR, SEM, and EDS. The spectrograms are presented in Figures 1, 2, and 3 respectively.

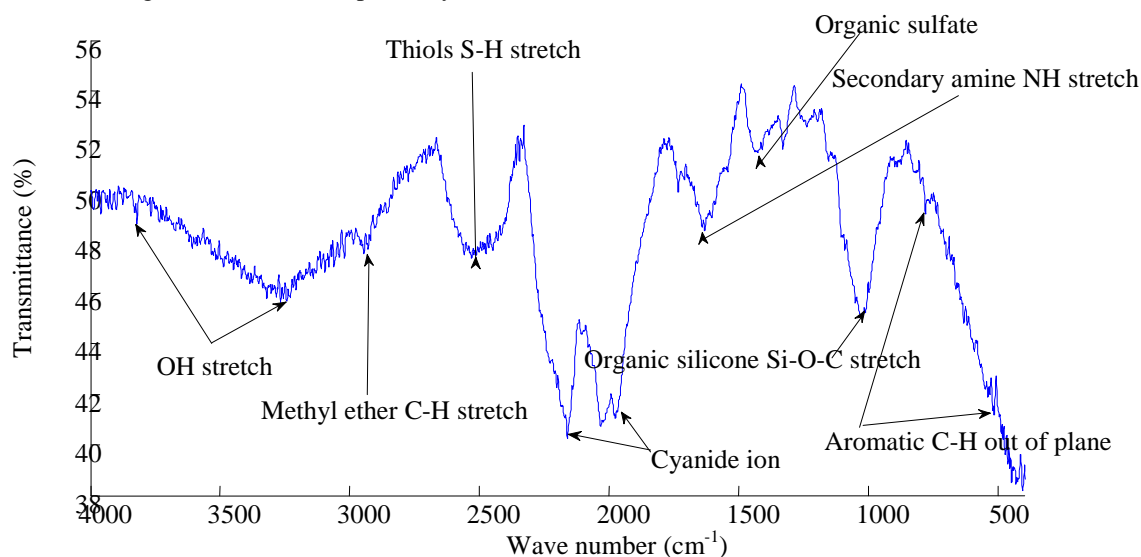


Figure 1: FT-IR spectrum of banana peel (RBP) biosorbent

From the FTIR spectra in Figure 1 above, it can be seen that many peaks exist indication of many functional groups. A shift at (Figure 1) wave number 539-800  $\text{cm}^{-1}$  was observed and was assigned to aromatic C-H out-of plane. The shift observed at 1035  $\text{cm}^{-1}$  was assigned to Si-O-Si silicon stretching while the absorption at 1450  $\text{cm}^{-1}$  was assigned to organic sulfate stretch. The shift at 1600  $\text{cm}^{-1}$  was assigned to secondary amine NH stretch. Shift at 1988-2200  $\text{cm}^{-1}$  was as a result of cyanide ion stretching while shift at 2500  $\text{cm}^{-1}$  was due to thiol S-H stretch. Methyl ester C-H stretch was observed at 2950  $\text{cm}^{-1}$ . The last shift at 3250-3900  $\text{cm}^{-1}$  was assigned to OH stretch. These assignments are in agreement with Kamsonlian et al., (2011) but slightly differ from Kaewsam et al., (2008) and Kumar et al., (2010).

Dissolved oil from the produced water polarizes in water. The charged particles initiate a reaction by opening the double and triple bonds in the banana peel structure and exchange their ions to neutralize the charges. Where the pollutants do not dissociate in solution, adsorption is by affinity of the surface to bind with the pollutant through the porous structure of the sorbent material.

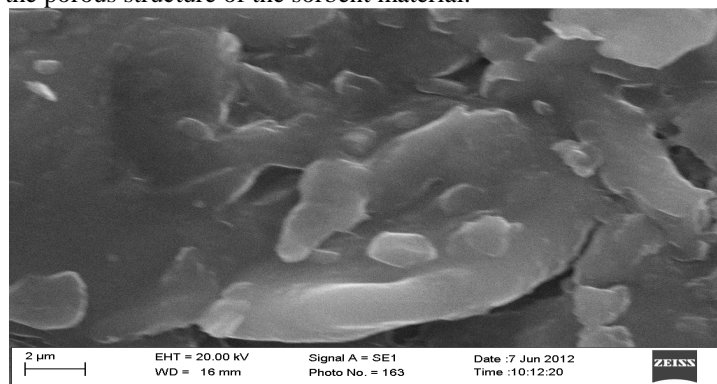


Figure 2: SEM of banana peel sample

Figure 2 shows the SEM of RBP. However, it can be said to consist of layered materials clustered together in which the sorption may take place. Particles seen on the image were measured to have equivalent spherical diameter from  $37.357\text{-}179.619 \times 10^{-6}$  m and cross sectional area of  $62 \times 10^{-12}\text{-}206 \times 10^{-12}$  m<sup>2</sup>.

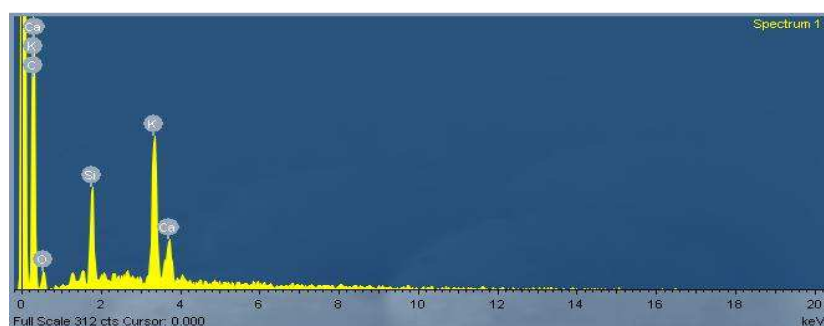


Figure 3: EDS of banana peel (RBP)

Table 1: Elemental analysis of RBP

Element	Weight, g	Atomic, %	Weight, %
C	4.65	88.52	78.41
O	0.40	5.69	6.74
Si	0.27	2.19	4.55
K	0.48	2.80	8.09
Ca	0.14	0.80	2.36
Totals	5.93	100	100

The use of peels and other bio-waste materials not only would provide less costly adsorbent or synthetic ion exchangers but is also easily available and save the environment. Banana peel has an average particle size of 0.89 nm as reported by Kamsonlian *et al.*, (2011) and a BET surface area of 1.856 m<sup>2</sup>/g. The surface area of RBP by BET and Langmuir methods were found to be 0.1135 and 0.1307 m<sup>2</sup>/g respectively. Its average pore size was determined to be 58.33421 nm while its pore volume determined to be 0.001655 cm<sup>3</sup>/g. However, the surface area did not agree with what was reported by Kamsonlian *et al.*, 2011. This may be attributed to peculiarities of the particular banana species used.

### 3.3 Batch adsorption experiments

The experiments were carried out by taking 300 mL of 194 mg/l produced water and different quantity of RBP in a 600 ml beaker. The contents were then agitated at 700 rpm for 30 minutes using mechanical shaker at room

temperature. The biosorbent and sorbate were separated by 63 micron sieve. Studies on the effects of agitation time, and biosorbent dose were carried out by using known amounts of biosorbents of particle size 212-63 microns. Oil solutions (300 mL) with different amounts of biosorbents were taken to study the effect of adsorbent dosage on the removal of oil. The biosorption experiments were carried out at room temperatures.

The laboratory synthesized produced water (oil-in-water mixture) was prepared by mixing crude oil with distilled water. The pH was kept constant during the experiment. The already prepared oil-water mixture was treated differently with various quantities of RBP for a period of 30 minutes and a stirring speed of 700 rpm. At the end of the treatment, RBP was removed from the oil/water mixture by passing through 63microns sieve; the residual oil in the water was determined using 1-1-1-tri-chloroethane as solvent. The extract was analyzed for oil content using HACH DR/2000 spectrophotometer at a wavelength of 450 nm. The test was repeated until optimum loading point was identified. With the optimum loading kept constant, the time was varied to determine the optimum time of the sorption study.

### 3.4 Adsorption isotherm

Equilibrium isotherm equations are used to describe experimental sorption data. Among several models that have been published in the literature to describe experimental data of adsorption isotherms Langmuir and Freundlich are the most frequently used models. The equation parameters and the underlying thermodynamic assumptions of these equilibrium models often provide some insight into both the sorption mechanism and the surface properties and affinity of the sorbent. The symbols and coefficients used in the equations are defined.

#### 3.4.1. The Freundlich Isotherm

In 1906, Freundlich presented the earliest known sorption isotherm equation (Freundlich, 1906). This empirical model can be applied to non-ideal sorption on heterogeneous surfaces as well as multilayer sorption and is expressed by the following equation (Ho, *et al.*, 2001):

$$q_e = a_F C_e^{b_F} \quad (1)$$

Where,  $a_F$  Freundlich isotherm constant;  $C_e$  solution phase oil concentration at equilibrium,  $\text{mg dm}^{-3}$ ;  
 The Freundlich isotherm has been derived by assuming an exponentially decaying sorption site energy distribution. It is often criticised for lacking a fundamental thermodynamic basis since it does not reduce to Henry's law at low concentrations. The linearized Freundlich equation can be used (equation 2):

$$\ln q_e = \ln a_F + b_F \ln C_e \quad (2)$$

A plot of  $\ln q_e$  vs  $\ln C_e$  is a straight line with slope,  $b_F$  and intercept,  $\ln a_F$  (Figure 4).

#### 3.4.2. The Langmuir Isotherm

Langmuir developed a theoretical equilibrium isotherm relating the amount of gas sorbed on a surface to the pressure of the gas (Langmuir, 1916). The Langmuir model is probably the best known and most widely applied sorption isotherm. It has produced good agreement with a wide variety of experimental data and may be represented as in equation 3 (Ho *et al.*, 2001): The Langmuir adsorption isotherm assumes that adsorption takes place at specific homogeneous sites within the adsorbent, and it has been used successfully for many monolayer adsorption processes. The linearized Langmuir can be used (equation 4):

$$q_e = \frac{k_L b C_e}{1 + k_L C_e} \quad (3)$$

$$\frac{C_e}{q_e} = \frac{1}{b k_L} + \frac{1}{b} C_e \quad (4)$$

Where,  $C_e$  is the equilibrium concentration of the sorbate (milligrams per litre),  $q_e$  is the amount of sorbate per unit mass of biosorbent (milligrams per gram),  $k_L$  is a constant representing the strength with which the solute is bound to the substrate (L/mg) and  $b$  is the adsorption capacity of the substrate (gram solute/gram adsorbent). When  $\frac{C_e}{q_e}$  was plotted against  $C_e$  for RBP, a straight line with slope  $\frac{1}{b}$  will be obtained and an intercept  $\frac{1}{b k_L}$  while  $q_e$  is expressed as given in Equation (5).

$$q_e = \frac{V(C_i - C_e)}{m} \quad (5)$$

Where,  $V$  is the biosorption volume (litres),  $C_i$  is the initial concentration of the sorbate (milligrams/litre), and  $m$  is the mass of biosorbent (gram).

The essential characteristics of the Langmuir isotherm can be expressed in terms of a dimensionless equilibrium parameter,  $R_L$  which is defined by.

$$R_L = \frac{1}{1 + bC_o} \quad (6)$$

Where,  $C_o$  is the highest initial solute concentration,  $b$  the Langmuir's adsorption constant (L/mg). The value of  $R_L$  indicates the type of the isotherm to be either unfavorable ( $R_L > 1$ ), linear ( $R_L = 1$ ), favorable ( $0 < R_L < 1$ ) or irreversible ( $R_L = 0$ ).

#### 4.0 Results and Discussions

##### 4.1 Results

Tables 2 and 3 present the results obtained on biosorption of oil from water using RBP.

Table 2: Effect of RBP loading rate on oil removal from produced water

Dosage (mg)	Residual oil ( $C_e$ ) (mg/L)	Oil removed (mg)	% Removal
0	194.00	0	0
10	10.66	183.34	94.51
20	8.52	185.48	95.61
30	4.26	189.74	97.80
40	2.13	191.87	98.90
50	0	194.00	100.00

Table 3: Effect of contact time on the biosorption of oil from produced water

Time (minutes)	$C_e$ Oil left (mg/L)	Oil removed (mg/L)	% Removal
0	194.00	0	0
5	85.27	108.73	56.04
10	83.96	130.04	67.03
15	57.56	136.44	70.33
20	56.69	134.31	69.23
25	38.37	155.63	80.22
30	2.13	191.87	98.90
35	0.00	194.00	100.00
40	0.00	194.00	100.00

The linearized form of Freundlich and Langmuir isotherms (equations (2) and (5)) were used. Figures 4 and 5 in conjunction with Tables 2 and 3

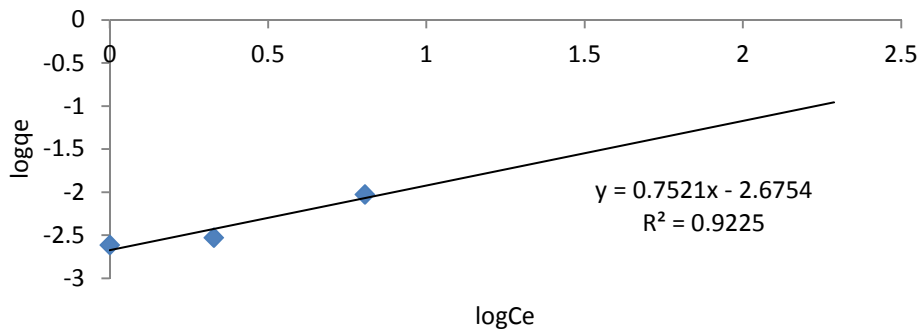


Figure 4: Freundlich Isotherm of RBP

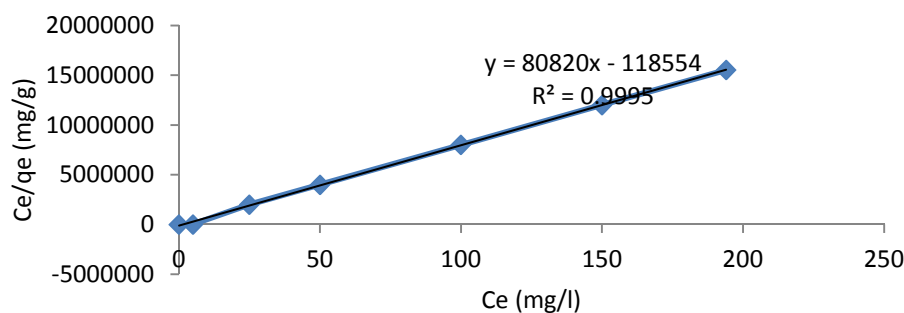


Figure 5: Langmuir plot of RBP

As can be seen from the figures, the  $R^2$  values for the charts were satisfactory. The various constants evaluated can be seen in Table 4, from which it is clear that the sorption process is better described by Langmuir isotherm. This indicates that sorption by banana peel is mono-layer as suggested by Langmuir.

Table 4: Evaluated constants from Langmuir and Freundlich isotherms by RBP

Isotherm	$R^2$	B	$k_L$	$a_F$	$b_F$	$R_L$
Langmuir	0.999	1.237E-5	-6.817	-	-	0.9975
Freundlich	0.9225	-	-	0.0609	0.7521	-

$R_L$  was evaluated from Figure 5 and the value obtained was 0.9975. Although the value of the dimensionless equilibrium parameter,  $R_L$  is satisfactorily good, the value falls within favorable range of  $0 < R_L < 1$ . The adsorption hence favored mono-layer as it agreed with Langmuir isotherm  $R^2=0.999$ .

#### 4.2 Kinetic studies

A number of kinetic models have been proposed to elucidate the mechanism of solute adsorption. These kinetic models are useful for the design and optimization of effluent treatment process. In order to investigate the mechanism of oil biosorption by RBP, the following kinetic models were considered. Pre-equilibrium kinetic profiles were characterized in order to determine the rate limiting steps involved in the process of biosorption of oil onto RBP. The first order (Eq. (7)), Lagergren pseudo-first order (Eq. (8)) and pseudo-second order (Eq. (9)) kinetic models were applied in the biosorption of nickel on spent activated clay (Mahmoud *et al.*, 2012), reactive black 5 dye by *Aspergillus foetidus* (Patel and Suresh, 2008), biosorption of Acid Red 57 by dried *Cephalosporium aphidicola* cells (Kiran *et al.*, 2006) and biosorption of  $Cu^{2+}$  unto banana peel (Hossain *et al.*, 2012).

$$\frac{1}{q_t} = \frac{1}{q_e} + \frac{k_1}{q_e t} \quad (7)$$

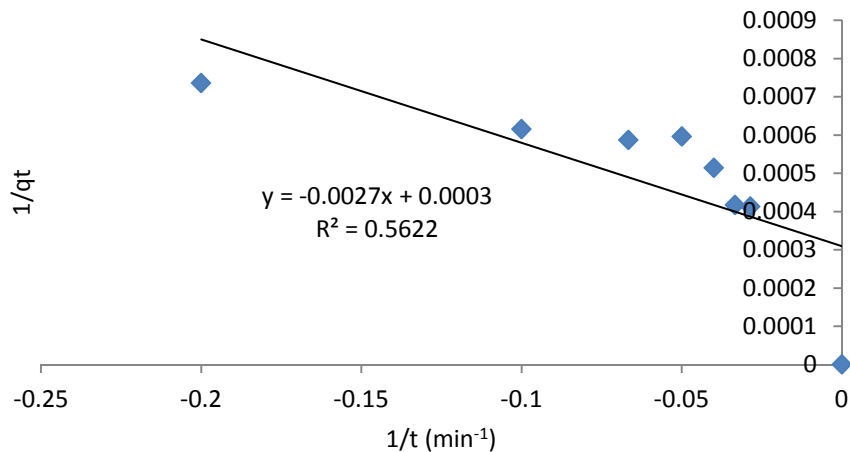


Figure 6: First order kinetic model for RBP

#### 4.2.2 Lagergren Pseudo first order kinetic model

The integrated linear form of pseudo first order kinetic model the model proposed by Lagergren is

$$\ln(q_e - q_t) = \ln q_e - kt \quad (8)$$

Where,  $q_e$  is the amount of dye adsorbed at equilibrium (mg/g),  $q_t$  is the amount of oil adsorbed at time  $t$  (mg/g),  $k_1$  is the first order rate constant ( $\text{min}^{-1}$ ) and  $t$  is the time (min). Hence, a linear trace is expected between the two parameters  $\ln(q_e - q_t)$  and  $t$ , provided the biosorption follows first order kinetics. The values of  $k_1$  and  $q_e$  can be determined from the slope and intercept. Even though  $q_e$  and the  $R^2$  value suggest that the biosorption data badly fitted to pseudo first order kinetics. Hence, the biosorption of oil onto RBP may not follow the pseudo first order rate expression.

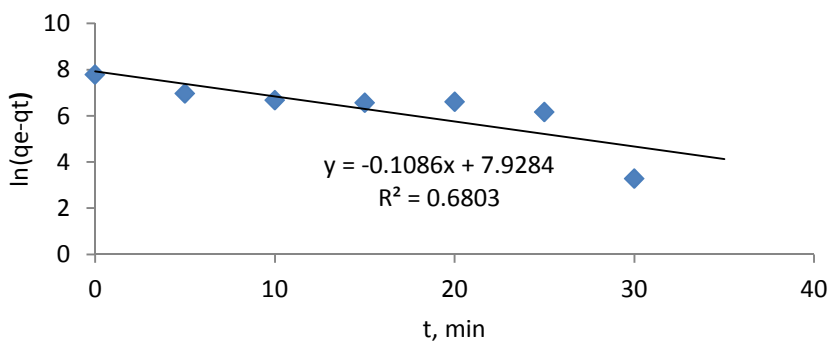


Figure 7: Pseudo-first-order kinetic model of RPB

#### 4.2.3 Second order and pseudo second – order kinetics

The biosorption may also be described by a second order or pseudo second order kinetic model. The linearized form of the second order and pseudo second order models are:

$$\frac{1}{c_e} = k_2 t + \frac{1}{c_0} \quad (9)$$

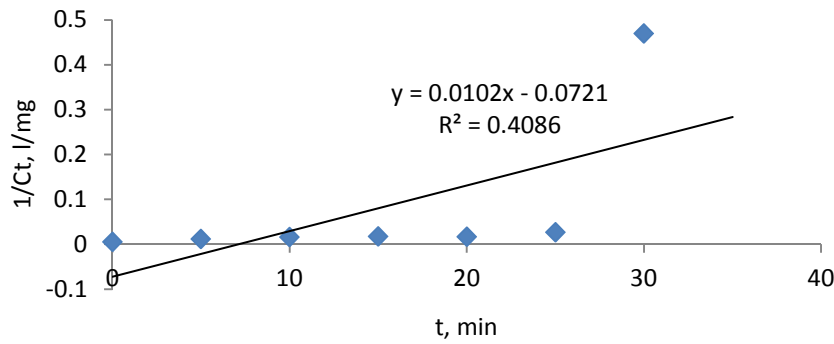


Figure 8: Second Order Kinetic Model of RBP

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \quad (10)$$

Where,  $k_2$  is the second order rate constant (g/mg min). A plot of  $1/q_e$  vs  $t$  and  $t/q_t$  vs  $t$  should be linear if the adsorption follows second order or pseudo-second order.  $q_e$  and  $k_2$  can be calculated from the slopes and intercepts of the plots.

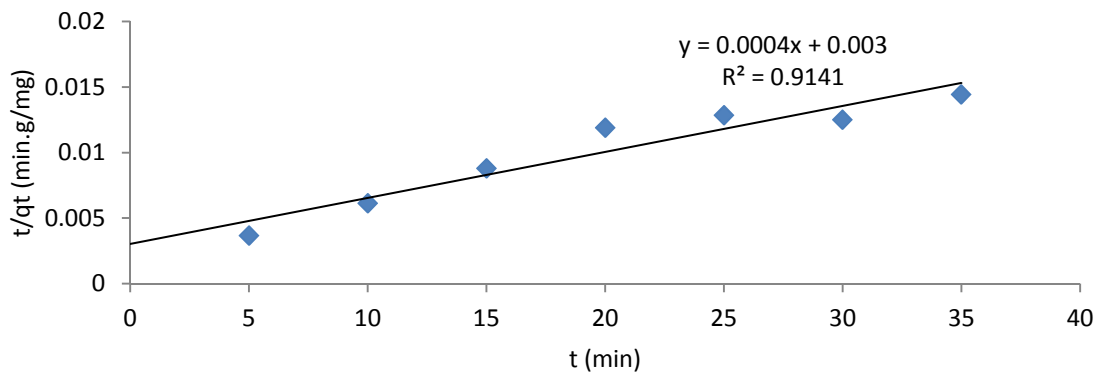


Figure 8: Pseudo-second-order Kinetic Model of RBP

#### 4.2.4 Elovich model

The Elovich equation is mainly applicable for chemisorption process. The equation is often valid for systems in which the adsorbing surface is heterogeneous. The Elovich model is generally expressed as

$$q_t = \frac{1}{b} \ln(ab) + \frac{1}{b} \ln t \quad (11)$$

Where,  $a$  is the initial biosorption rate (mg/g min) and  $b$  is related to the extent of surface coverage and the activation energy for chemisorption (g/mg). A plot of  $q_t$  vs  $\ln t$  gives a straight line with a slope of  $1/b$  and an intercept of  $1/b \ln(ab)$  with good correlation coefficients.

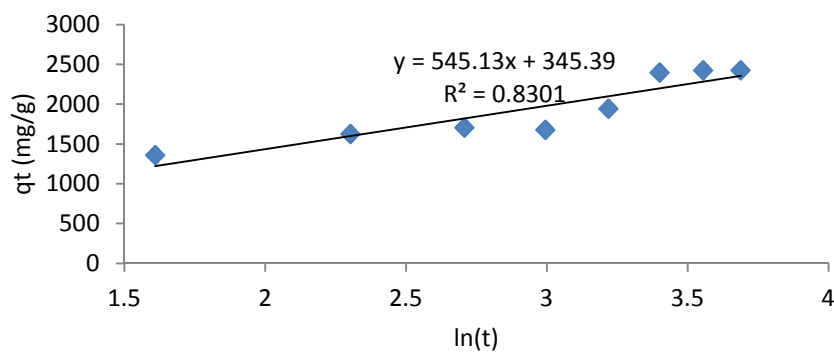


Figure 9: Elovich Kinetic Model of RBP



#### 4.3: Effect of initial pH of water on oil removal

pH is one of the most important parameters which control the sorption process of adsorbents, functional groups and ionic state of adsorbate. The adsorption capacities/ removal efficiency of oil from water onto banana peel were strongly affected by the pH (Figure 10). The adsorption capacities/efficiency at pH 0.7, 1.1, 10.0, and 11.6 were investigated. The results were found to be 84.54, 82.99, 87.63, and 87.63% removal efficiency respectively. There was significant increased in removal efficiency from acidic to alkaline region. The increased from 82.99% to 87.63% was recorded at pH from 1.1 to 10.0. However neutral pH was not investigated. The removal efficiency (87.63%) tends to be constant at pH greater or equal to ten ( $\text{pH} \geq 10$ ).

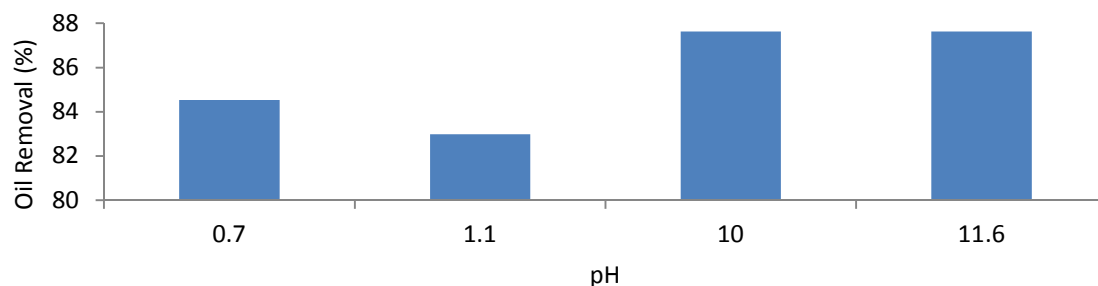


Figure 10: Effect of pH on the Biosorption of Oil using Banana Peel

#### 4.4: Effect of initial temperature of water on oil removal

Normally temperature stimulates the molecules, functional groups and surface morphology of the adsorbent and adsorbate during adsorptions processes. To determine the thermal effects of oil adsorption onto banana peel, temperature variation experiments were conducted at 40, 50, 60 and 80°C, with an initial oil concentration 30 mg/l. The experimental data show that the equilibrium adsorptions ( $q_m$ ) were increased with an increase in temperature up to 60°C when it started decreasing (Figure 11), suggesting that higher temperature helped to desorb the oil or retard the oil adsorption onto banana peel. The pattern followed by oil sorption with change in temperature can best be explained by Figure 12 and is a polynomial curve of power 3.

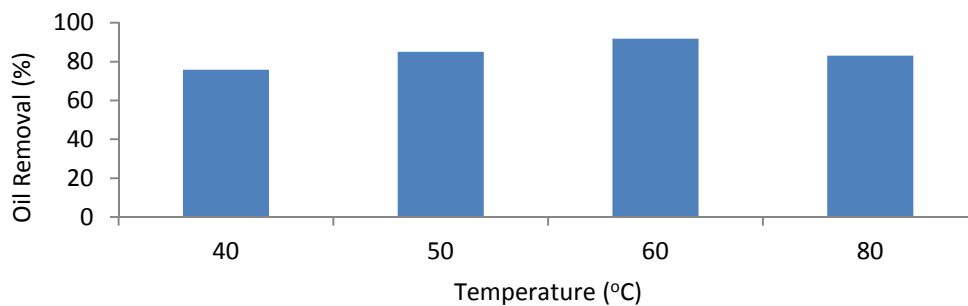


Figure 11: Effect of temperature on the Biosorption of Oil using RBP

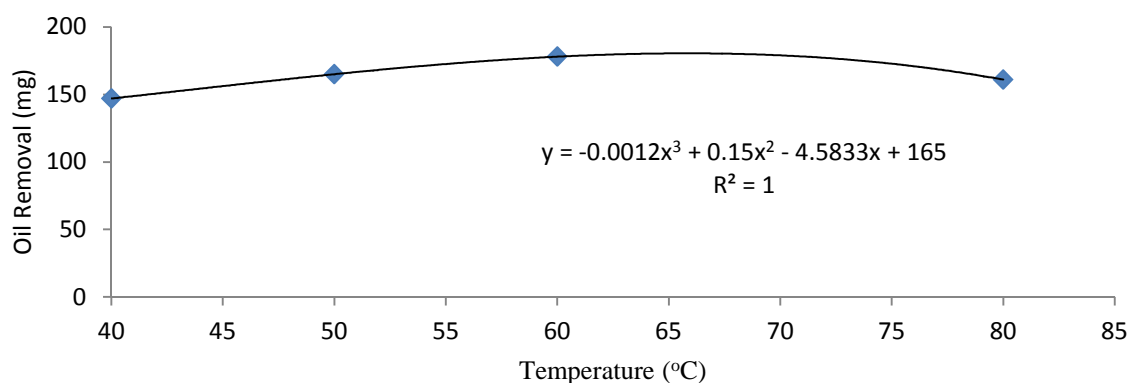


Figure 12: Biosorption of Oil on RBP with Temperature

From results of Figures 6-9, it can be deduced that oil sorption from produced water using RBP fits pseudo-second-order kinetics. Comparison of the tested kinetics and calculated constants are shown in Table 6.

Table 5: Comparison of the biosorption kinetics and evaluated constants

Kinetics	R <sup>2</sup>	Intercept	Slope	Rate constant (k)	q <sub>e</sub>	C <sub>o</sub>	a	b
1 <sup>st</sup> order	0.5622	0.0003	-0.1027	-340.67	3333.33	-	-	-
Pseudo-1 <sup>st</sup> order	0.6803	7.9284	0.1086	0.1086	2774.98	-	-	-
2 <sup>nd</sup> order	0.4086	-0.6721	0.0102	0.0102	-	1.4879	-	-
Pseudo-2 <sup>nd</sup> order	0.9141	0.003	0.0004	5.333E-3	2500	-	-	-
Elovich	0.8301	345.39	545.13	-	-	-	1046.87	0.0018

## 5.0 Conclusion

RBP was characterized using FTIR, SEM, EDS, and BET equipment. The characterization using FTIR revealed the functional groups present on the RBP while SEM revealed the surface morphology of the sorbent. EDS revealed the elemental composition of the biosorbent to contain carbon, oxygen, silicone, potassium, and calcium. Surface area of the RBP was measured using nitrogen adsorption by BET method and found to be 1.86 m<sup>2</sup>/g with average pore size of 58.3nm. Biosorption studies with the peel showed that it can be used in the removal of oil (both dissolved and dispersed) from produced water providing almost 100% removal at an equivalent dosage representing 267 mg banana peel in produced water after 35 minutes exposure and initial oil concentration as high as 194 mg/l. Biosorption factors such as effects of loading rate, contact time, solution pH, and temperature were studied. pH and temperature effect were seen to follow a polynomial path of the third order. Several biosorption kinetic models were tested and the results showed that RBP biosorbent followed pseudo-second-order kinetics. Langmuir and Freundlich isotherms were tested with the data the results showed that Langmuir isotherm gave the best fit indicating mono-layer adsorption of oil on the RBP surface.

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