

Evaluation of AGO Adsorption Rate Isotherms Using Adsorbent Formulation of Plantain Agbagba¹ and Clay of Different Mix Ratio in Pollutant Remediation in Salt Water

Nnadi Vincent Gilbert¹, Ukpaka Chukwuemeka Peter^{2,*}

Abstract

The application of some agro-based materials in combination with day soil, for the production of adsorbents were investigated in relationship to their performance in AGO (Diesel) treatment in a batch process unit. The research allows the model concept of Langmuir isotherm, Frundlich isotherm and Temkin Isotherm for the determination of the adsorption rate of the various isotherms with respect to the effect of the particle size and the mixed ratios of the clay soil and the agro-based materials used in this research. The particle size investigated were 1.18 mm, 50 µm, 300 µm, 600 µm and the performance of the adsorbent was rated with the particle size as observed on the obtained data. The characteristics of AGO adsorbed with respect to the quantum rate of adsorption shows that the smaller the particle size the more the rate of contaminants removal will increase. Mixed ratios the day soil to the agro-based materials used were R1:9, R:2:8, R3:7, R4:6, R5:5, R6:4, R8:4 and the isotherms value determine by establishing the regression linear lien of the plots. The process allows the evaluation of the slope and intercepts values, which was subjected to the formulated isotherm models of Temkins, Freudlich and Largmuir. The adsorbents were produced by using clay soil mixed with each of the agro – based materials of plantain agbagba. The research revealed that the adsorption rate of the output response increases with increase in the dependent parameter of plot in the x-coordinate and this behaviour was experienced in all the plot showing the significance of the produced adsorbent as a good formulation for treatment of AGO (diesel product) in salt water medium.

Keywords: Salt, Water, Medium, AGO, Contaminants, Adsorption, Treatment.

INTRODUCTION

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This research reviewed the synthesis and absorbing properties of a wide range of porous material including silica gels, zeolites, organoclays and natural sorbents relative to their use as in removal or organics in oil spill clean-up. Their structure permits the penetration of compounds into the solid. They have large surface areas, high porosity, low density and thermal conductivity [1-3]. However, the collapse of their structure due to adsorption of water makes it an obstacle for commercialization as good adsorbent. Zeolite: Hydrophobic zeolites have a small percentage of aluminium atoms in their structure which shifts their affinity from polar molecules like water towards non polar substances like organic solvents. They could be treated and modified and adapted to suit specific purposes of adsorption for oil spill

cleanup. While hydrophobic zeolites have the advantage of its ability for regeneration, its disadvantage of having much less adsorption compared to active carbon solvents [4–5].

This study reviewed the adsorption of petroleum compounds on four types of sawdust (Walnut, Poplar, Beech and Pine). The adsorption ability was examined at different pH, temperature and H₂O₂. Groundwater was collected from water well close to a refinery. A complete analysis of the water samples was done. The hydrocarbon content of the water samples was compared with the range of compounds in gasoline. The sawdust (adsorbent) was sieved, dried and the various parameters such as specific area, density, moisture content, ash content and volatile matter were obtained [6–8].

According to this study, it was found that the walnut sawdust has higher adsorption capacity than the other types. It was also found that the equilibrium adsorption capacity was a function of pH, temperature and H₂O₂. The maximum adsorption of petroleum compounds was obtained at pH 8. The adsorption of petroleum compounds increased by decreasing the temperature and H₂O₂ concentration. The kinetics were studied with two well-known models (Freundlich and Langmuir). The Freundlich model fitted the data obtained. This work is more in the same direction of my research of looking into the best local products that could be used to substitute synthetic products for adsorption [8–11].

This study examined the sorption efficiency and capacity of raw and modified fibres for diesel, crude oil and vegetable oil. Natural sorbents have been found to be more effective than synthetic sorbents because of their low costs and biodegradability. This work therefor examined the efficiency and capacity of using Palm fibres as a natural adsorbent. To evaluate this, the effect of sorption time, oil concentration, particle size, adsorbent dosage and temperature of the crude oil were examined [1].

The experiment included preparation of the adsorbent, exposure of the adsorbent to the crude samples. The sorption efficiency and capacity were calculated. The physical properties (density and viscosity) of the crude oil, diesel oil and vegetable oil. To improve their efficiencies of removing oil, the adsorbent was treated with acid and alkaline solutions. At the end of the experiment, the adsorption capacity of the palm fibres was found to increase with time, thickness of oil film, temperature and particle size. The adsorption efficiency remained constant after both acid and alkali treatment. The maximum adsorption capacity of palm fibres was found to 35.71, 22.73 and 21.74 g oil/g adsorbent for diesel, crude oil and vegetable oil respectively. These natural adsorbents were found to be eco-friendly because of its biodegradability and cost effective [10].

This research presented at the 16th International Conference “Chemistry and Chemical Engineering in XXI century” investigated the adsorption activity of natural sorbents to be used for clean-up of water surface from hydrocarbon under various temperatures [3]. The experiment determined the adsorption capacity, oil capacity and water absorption. It was determined that oil capacity is a function of density, molecular weight, temperature and viscosity. The experiment also concluded that natural sorbents have good buoyancy and great oil capacity and better sorption properties when compared with sawdust. It also has the advantage of being eco-friendly. The adsorbents degrade over time without requiring special disposal methods. Due to its very high buoyancy, they could be recovered from water surface for disposal. Their low cost also makes them cost effective for the purpose [4].

This study tried to compare the adsorption of crude oil, diesel and kerosine using Rice husk as sorbent with that of a standard synthetic sorbent. The sorption process was studied using Langmuir, Freundlich, Elovich, Temkin and Dublin-Radushkevich adsorption models. The kinetics was studied using Lagergren pseudo-first-order, pseudo-second-order intraparticle diffusion and liquid film diffusion models.

MATERIALS AND METHODS

Capacity for Oil Sorption Test Methods

The fresh water environment used for this research was tested for the reason of ensuring that all leaked oil is removed from the exterior of agro-based material used, crude oil and water. The formulated

adsorbents were observed to be in contact with the fresh water environment contaminated with PMS and AGO and the performance of the adsorbent up take of the PMS and AGO monitored with respect to changes in Total Petroleum Hydrocarbon (TPH), ammonia, nitrogen, iron, sulphur copper etc. The performance by the formulated adsorbent in terms of its potential to withstand the water environment without dissolving was monitored and the suitability of the adsorbents in the mitigation or reduction of contaminants concentration (pollutants) in each batch process was investigated.

Pollutants of PMS and AGO of 200 ml was introduced into plastic containers of 700 mL of water each and formulated adsorbents of sample range of weight of 15.3 g to 32.3 g of different mix ratios of particle size were immersed into the different experimental set up. The plastic containers contained the either PMS or AGO, fresh water and formulated adsorbent and the mixture is always stirred and the experiment-maintained room temperature. The formulated adsorbent remains the plastic container whereas small quality of the sample is collected for analysis at interval of one hour for a period of six hours. The adsorbent samples were removed from each plastic container after intervals of one hour for analysis and concentration of medium determined as well as the weight of the adsorbent. The weighed adsorbent is returned to each plastic container and samples analysed. The collected samples are stored in cool environment of temperature of 4 °C to avoid change in composition. The initial weight of the plastic containers was measured plus the weight of pollutant and adsorbent immersed to achieve the total weight of the process.

The samplings at each one hour involve the process of stirring before collection of each sample for gas chromatography (GC) analysis. Before than initial weight of the mixture after removing the adsorbent is measured as well as the weight of the adsorbent and final weight of contaminated container. As the removed mixture of the contaminated fresh water is returned the weight of each container is reweighed.

The application of established procedure for calculating the oil sorption capacity (q) was determined using.

$$q = \frac{M_p - (M_i - M_f)}{M_i} \quad (1)$$

where,

M_p = mass of the adsorbent after draining (g)

M_i = initial mass of the adsorbent (g)

M_f = mass of the water in the adsorbent (g)

For the (PMS and AGO) sorption capacity q_{TPH} can be calculated by using the formula of

$$q_{TPH} = \frac{\left[\begin{array}{l} \text{Mass of PMS or AGO added in each container} \\ \text{Mass of water added into contain-} \\ \text{Mass of adsorbent} \end{array} \right]}{\left[\begin{array}{l} \text{Mass of mixture remaining inside the container} \\ \text{af terth eadsorbed and moved} \end{array} \right]} \quad (2)$$

The amount of AGO or PMS adsorbed (g) per g of the sorbent (s) can be evaluated as

$$S = \frac{W_{SS} - W_{SF}}{W_{SF}} = \frac{W_{TPH}}{W_{SF}} \quad (3)$$

Where, W_{SF} = weight of salt or fresh sorbent sample (g), W_{SS} = weight of sorbent saturated with PMS or AGO (g) and W_{TPH} = weight of AGO or PMS reserved in sorbent matrix (g)

Conceptual Computational Approaches

The conceptual approaches for quantity evaluation of the AGO and PMS adsorbed and percentage of AGO and PMS removed due to the adsorbent performance were calculated using the model or equation stated below

$$q = \frac{C_0 - C_e}{m} \times V \quad (4)$$

$$\%removal = \frac{C_0 - C_e}{C_0} \times 100 \quad (5)$$

where, q = sum of either AGO or PMS adsorbed by the sorbent (mg/g), C_0 = initial AGO or PMS concentration in interaction with the adsorbent (mg/l), C_e = final concentration (mg/l) after the container adsorption procedure, V = volume of aqueous solution (L) that the sum of volume of AGO or PMS and water in the plastic container (L) put in contact with the adsorbent and M = mass (g) of the adsorbent

Adsorption Isotherm Models

The adsorption isotherm models considered in this research work are Langmuir, Freundlich, and Temkin for the determination of the rate of adsorption of petroleum products in polluted water.

Kinetic of Langmuir Model

The concept of Langmuir Adsorption Isotherm Model was used in evaluating the adsorption process. The concept of Langmuir model expressed the adsorption at the stage of isothermal state and the expression is given as:

$$\frac{C_e}{q_e} = \frac{1}{K_L} + \frac{a_L C_e}{K_1} \quad (6)$$

where,

C_e = concentration of absorbate at equilibrium (mg/l),

q_e = Amount adsorbed per unit mass of the adsorbent (mg/g),

K_1 = Affinity between adsorbent and adsorbate and a_L = Langmuir constant related to the energy of adsorption

Kinetic of Freundlich Model

The concept of kinetic model of Freundlich adsorption was considered and the expression is

$$\text{Log}q_e = \text{Log}K_f + \frac{1}{n}C_e \quad (7)$$

where, K_f = Freundlich constant linked to the adsorption capacity, and $\frac{1}{n}$ = Freundlich constant corrected to the adsorption intensity

The Weber et al (1974) concept was used in expressing the Freundlich adsorption model kinetic

Kinetic of Temkin Model

The concept of the Temkin model equation is commonly used in describing the equilibrium adsorptive characteristics between binary phases comprising the adsorption system. The concept of Temkin isotherm model was demonstrated (Surya-Dash *et al.*, 2020) as given below.

$$q_e = a + b \ln C_e \quad (8)$$

Where, a and b are constant interrelated to energy and capacity of adsorption.

RESULTS AND DISCUSSION

Result of Rate of Adsorption of Isotherms of Langmuir, Freundlich, Temkin and other Rate Parameters

The rate of adsorption of the isotherm of Langmuir, Freundlich and Temkin and other rate parameters was presented in Figure 1 to 12. The slope and the intercept values from the regression equation of the linear curve was obtained and the result obeyed the Langmuir concepts and application in terms of rate of adsorption principle. However, the Freundlich and Temkin isotherms, all follows the same trend revealing the acceptability of the developed adsorbent formulation for treatment of AGO and PMS in fresh and salt water medium in stagnant condition.

Figure 1 shows the relationship of C_e/Q_e and C_e as a plot for the determination of the rate of adsorption of isotherm of Langmuir model in terms of R1(150 μ m) 1:9 mix rate of clay soil with PA1 for pollutant of AGO. The regression equation revealed $Y = 0.0055X + 44.388$ and R^2 value of 0.9894 demonstrating the reliability value of 98.94% and the slope value of 0.0055 and intercept value of 44.388 for salt water environment. The result revealed a good match as related to the equation of the line as well as the variation in C_e/Q_e could be attributed to the variation in C_e .

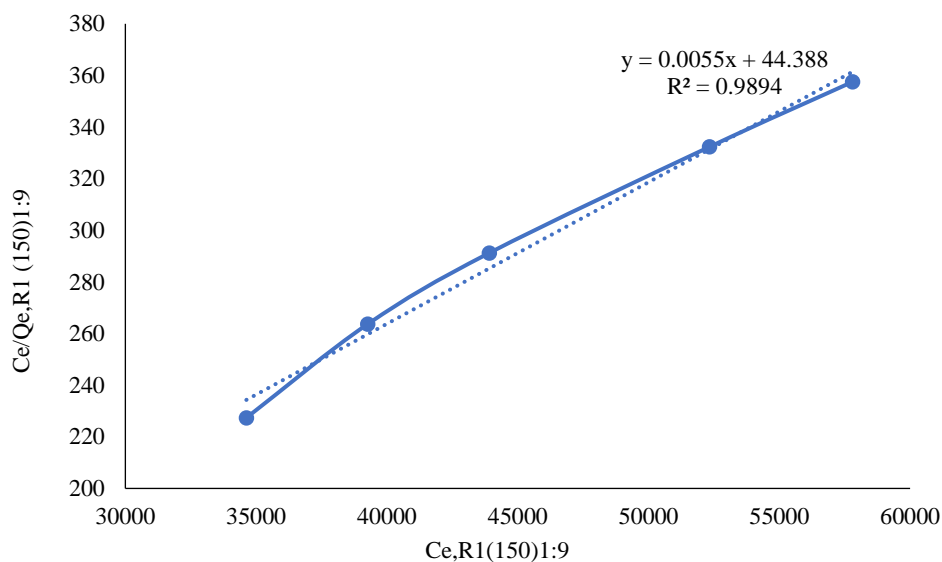


Figure 1. Plot of Langmuir Isotherm of C_e/Q_e versus C_e for R1(150 m) 1:9 Mix Ratio of Clay Soil with PA1 of Salt Water Environment Polluted with AGO.

Figure 2 shows the relationship of $\log Q_e$ and $\ln C_e$ as a plot for the determination of the rate of adsorption of isotherm of Freundlich model in terms of R1(150 μ m) 1:9 mix rate of clay soil with PA1 for pollutant of AGO. The regression equation revealed $Y = 2E-06X + 2.0935$ and R^2 value of 0.9906 demonstrating the reliability value of 99.06% and the slope value of 2E-06 and intercept value of 2.0935 for salt water environment. The result revealed a good match as related to the equation of the line as well as the variation in $\log Q_e$ could be attributed to the variation in $\ln C_e$.

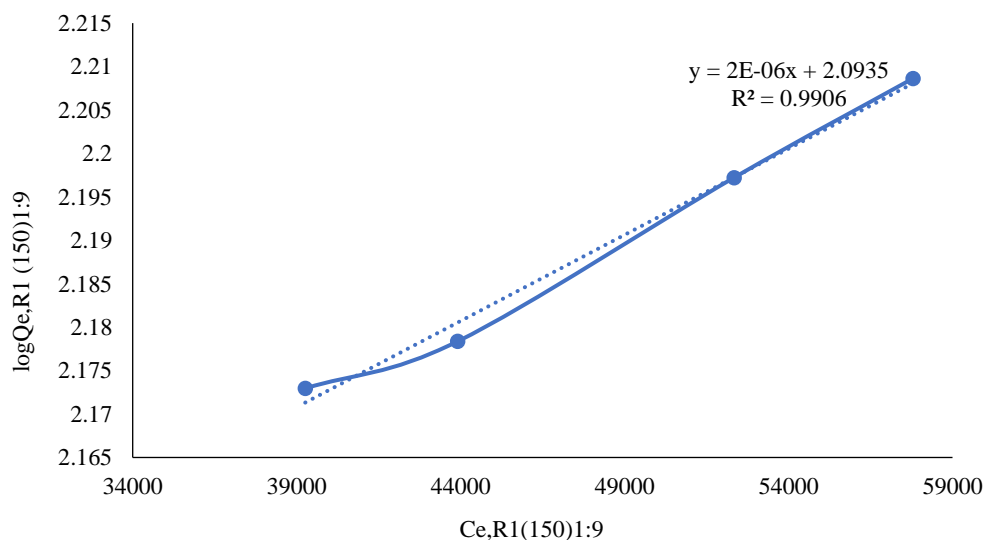


Figure 2. Freundlich Isotherm Plot of Mix Ratio of 1:9 of Particle Size of 150 μ m.

Figure 3 shows the relationship of Q_e and $\ln C_e$ as a plot for the determination of the rate of adsorption of isotherm of Temkin model in terms of R1(150 μm) 1:9 mix rate of clay soil with PA1 for pollutant of AGO. The regression equation revealed $Y = 33.752X + 208.35$ and R^2 value of 0.9787 demonstrating the reliability value of 97.87% and the slope value of 33.752 and intercept value of 208.35 for salt water environment. The result revealed a good match as related to the equation of the line as well as the variation in Q_e could be attributed to the variation in $\ln C_e$.

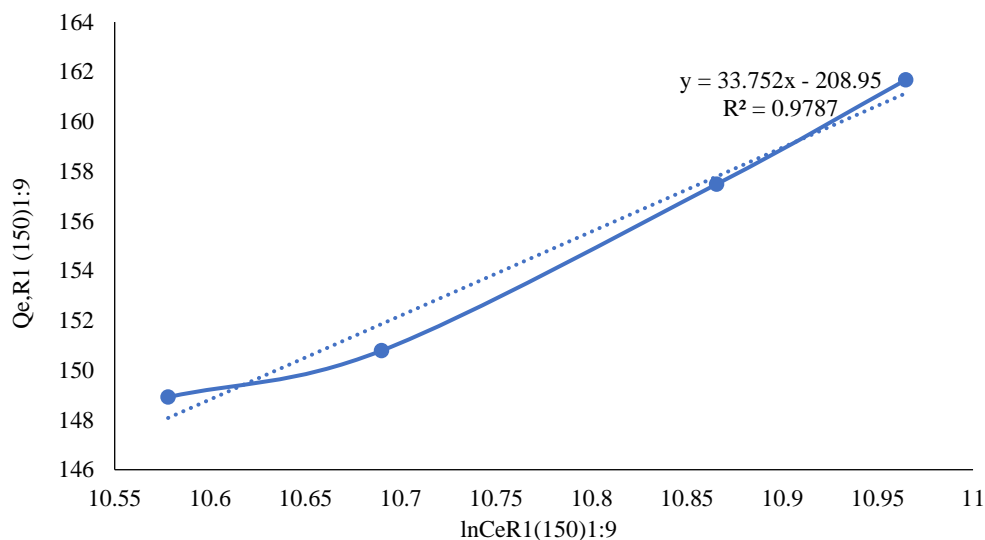


Figure 3. Temkin Isotherm Plot of Mix Ratio of 1:9 of Particle Size of 150 μm .

Figure 4 shows the Langmuir plot of C_e/Q_e against C_e for R2 (300 μm) 6:4 mix ratio of clay soil against C_e for R² (300 μm) 6:4 mix ratio of clay soil with PA1 for salt water environment contaminated with AGO. The regression equation of line obtained in this case is expressed as $Y = 0.0047X + 116.23$ with $R^2 = 0.9924$ demonstrating the reliability value of 99.24%. The result further revealed a good match of the linear equation obtained as well as showcases the slope of 0.0047 and intercept value of 116.23.

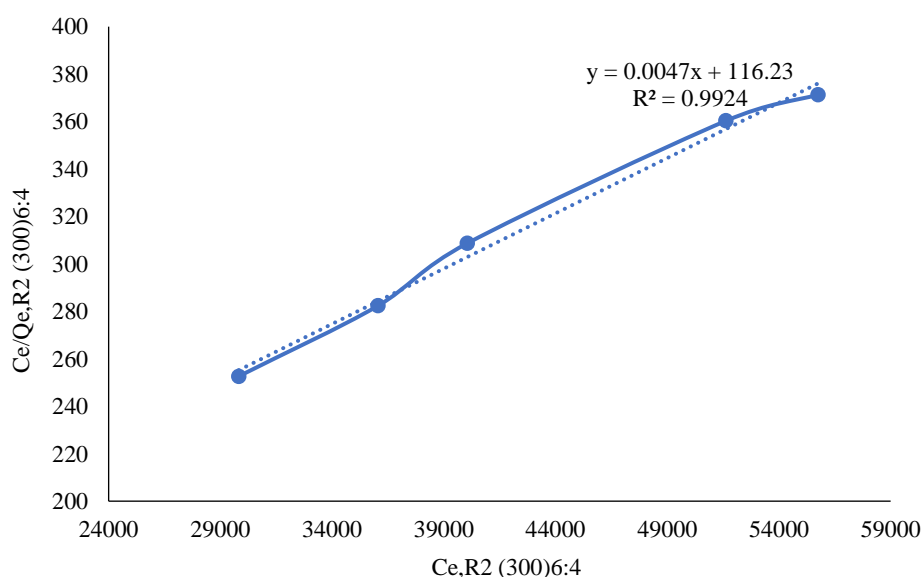


Figure 4. Plot of Langmuir Isotherm of C_e/Q_e versus C_e for R2(300 μm) 6:4 Mix Ratio of Clay Soil with PA1 of Salt Water Environment Polluted with AGO.

Figure 5 shows the Temkin isotherm plot of Q_e against $\ln C_e$ for R2 (300 μm) 6:4 mix ratio of clay soil against C_e for R² (300 μm) 6:4 mix ratio of clay soil with PA1 for salt water environment contaminated with AGO. The regression equation of line obtained in this case is expressed as $Y = 49.435X + 391.94$ with $R^2 = 0.9833$ demonstrating the reliability value of 98.3%. The result further revealed a good match of the linear equation obtained as well as showcases the slope of 49.435 and intercept value of 391.94.

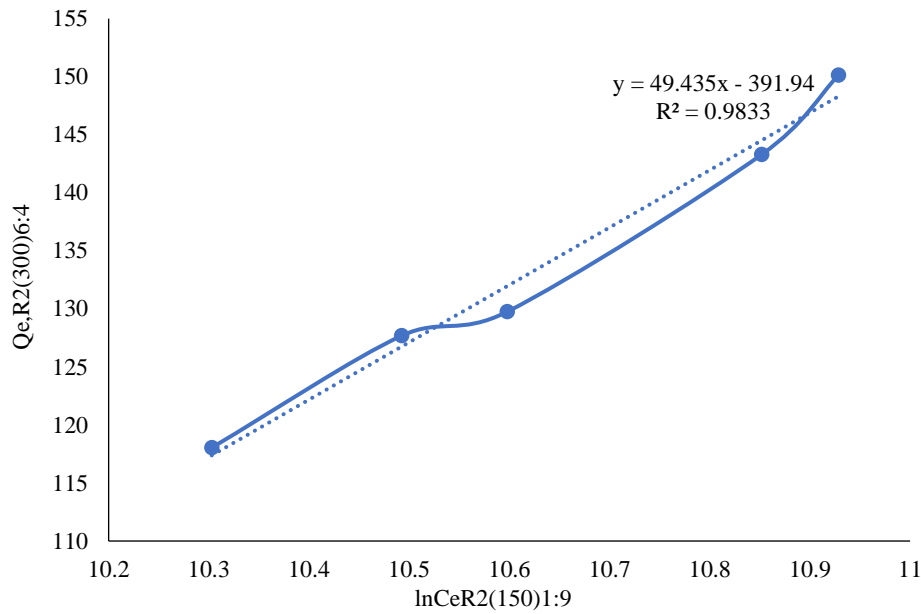


Figure 5. Temkin Isotherm Plot of Mix Ratio of 6:4 of Particle Size of 300 μm .

Figure 6 shows the Freundlich plot of $\log Q_e$ against C_e for R2 (300 μm) 6:4 mix ratio of clay soil against C_e for R² (300 μm) 6:4 mix ratio of clay soil with PA1 for salt water environment contaminated with AGO. The regression equation of line obtained in this case is expressed as $Y = 4E-06X + 1.9614$ with $R^2 = 0.9897$ demonstrating the reliability value of 98.97%. The result further revealed a good match of the linear equation obtained as well as showcases the slope of 4E-06 and intercept value of 1.9641.

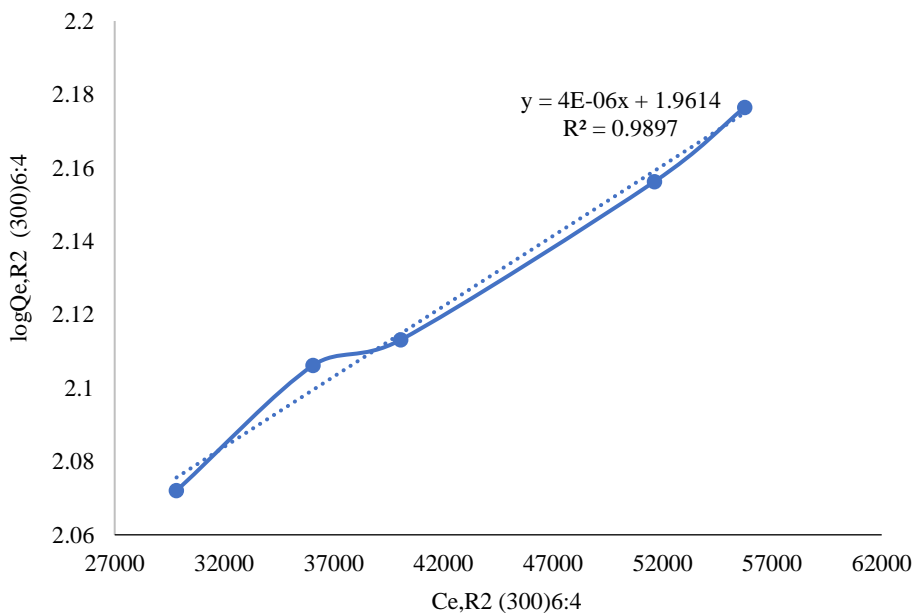


Figure 6. Freundlich Isotherm Plot of Mix Ratio of 6:4 of Particle Size of 300 μm .

Figure 7 shows the concept of Langmuir isotherm of the plot of C_e/Q_e against C_e for R3 (600 μ m) 6:4 mix ratio of clay soil with PA1 for adsorbent immersed in salt water environment which was contaminated with AGO. The line curve equation obtained is given as $Y=0.009 X + 637.6$ with $R^2 = 0.9895$ revealing slope of 0.009 and intercept value of 637.6 as well as reliability value of the Langmuir as 98.95%. Furthermore, this obtained result has shown a good match of fitting and the variation of the Langmuir isotherm of C_e/Q_e could be integrated to the variation in C_e .

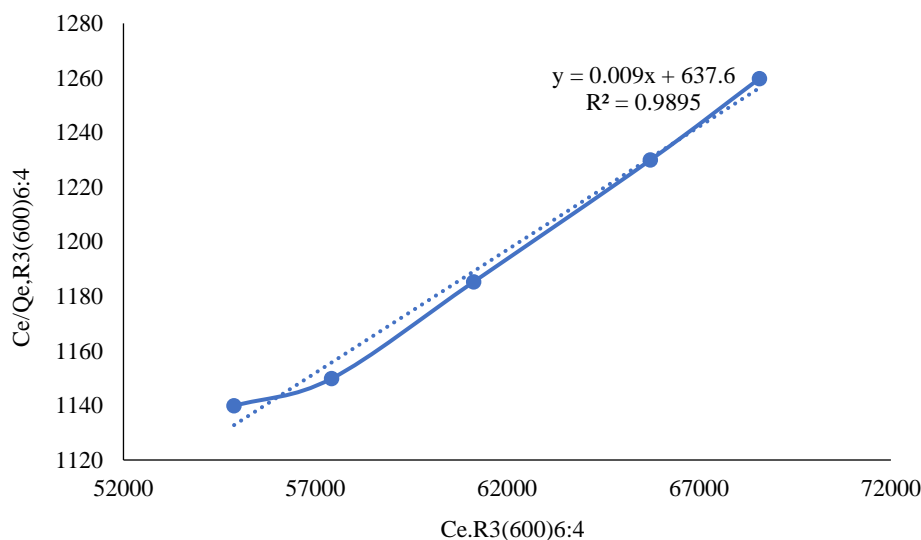


Figure 7. Plot of Langmuir Isotherm of C_e/Q_e versus C_e for R3(600 μ m) 6:4 Mix Ratio of Clay Soil with PA1 of Salt Water Environment Polluted with AGO.

Figure 8 shows the concept of Temkin isotherm of the plot of Q_e against $\ln C_e$ for R3 (600 μ m) 6:4 mix ratio of clay soil with PA1 for adsorbent immersed in salt water environment which was contaminated with AGO. The line curve equation obtained is given as $Y= 27.593 X + 252.68$ with $R^2 = 0.9925$ revealing slope of 27.593 and intercept value of 252.68 as well as reliability value of the Temkin as 99.25%. Furthermore, this obtained result has shown a good match of fitting and the variation of the Langmuir isotherm of Q_e could be integrated to the variation in $\ln C_e$.

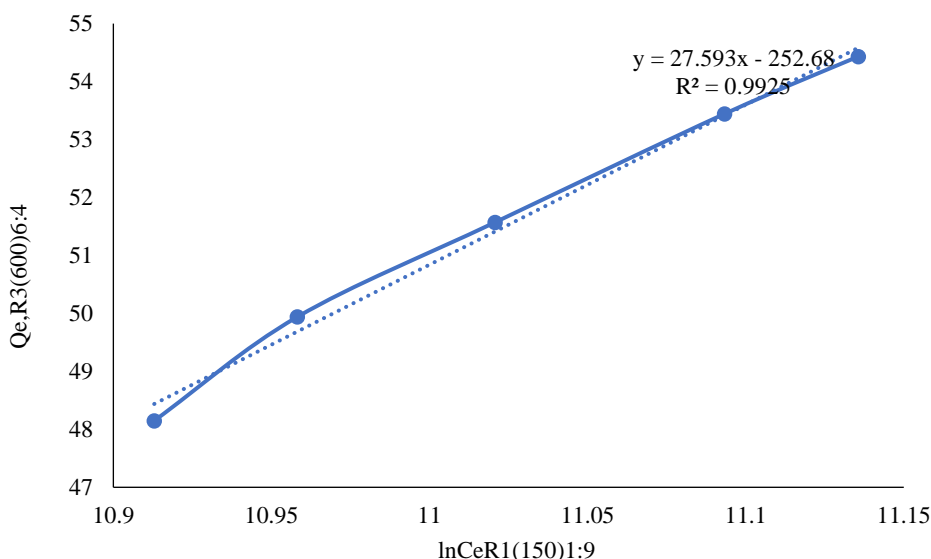


Figure 8. Temkin Isotherm Plot of Mix Ratio of 6:4 of Particle Size of 600 μ m.

Figure 9 shows the concept of Freundlich isotherm of the plot of $\log Q_e$ against C_e for R3 (600 μm) 6:4 mix ratio of clay soil with PA1 for adsorbent immersed in salt water environment which was contaminated with AGO. The line curve equation obtained is given as $Y = 4E-06 X + 1.4787$ with $R^2 = 0.9831$ revealing slope of $4E-06$ and intercept value of 1.4787 as well as reliability value of the Freundlich as 98.31%. Furthermore, this obtained result has shown a good match of fitting and the variation of the Langmuir isotherm of $\log Q_e$ could be integrated to the variation in C_e .

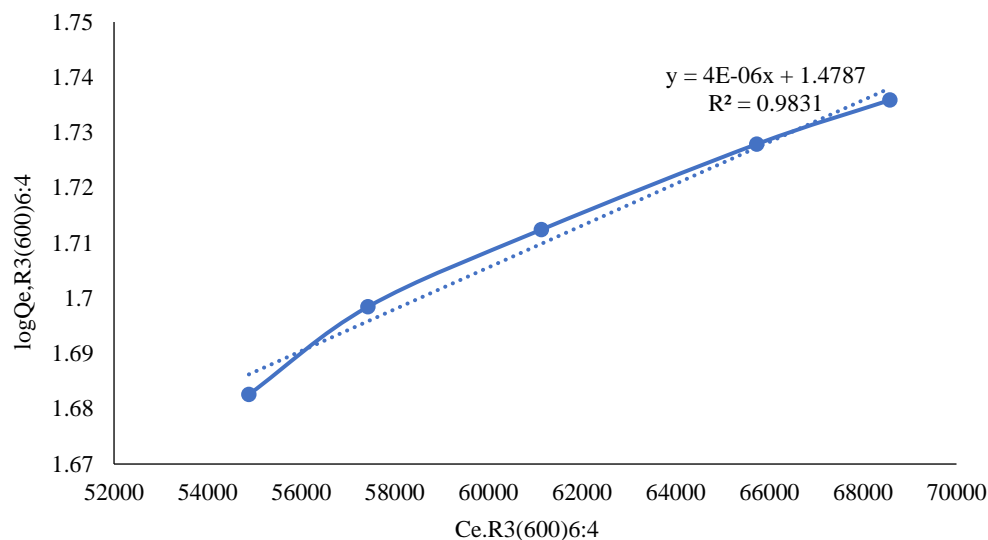


Figure 9. Freundlich Isotherm Plot of Mix Ratio of 6:4 of Particle Size of 600 μm .

Figure 10 shows the relationship between the Langmuir isotherm plot of C_e/Q_e against C_e for R4 (1.18 mm) 6:4 mix ratio of clay soil with PA1 for adsorbent immersed in salt water polluted with AGO. The regression equation of the line is given as $Y = 0.0088 X + 288.56$ with the square root of the best fit $R^2 = 0.995$ showing the reliability value of the Langmuir isotherm of 99.5% as well as the equation of the line illustrating the slope value of 0.0088 and intercept value of 288.56 . The variation in the Langmuir isotherm of C_e/Q_e could be integrated to the variation in C_e .

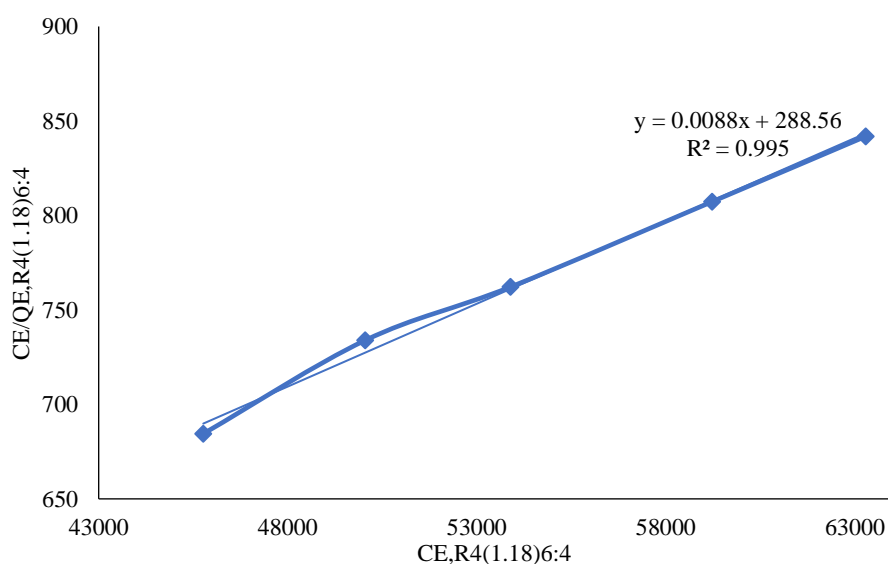


Figure 10. Plot of Langmuir Isotherm of C_e/Q_e versus C_e for R4(1.18 mm) 6:4 Mix Ratio of Clay Soil with PA1 of Salt Water Environment Polluted with AGO

Figure 11 shows the relationship between the Temkin isotherm plot of Q_e against $\ln C_e$ for R4 (1.18 mm) 6:4 mix ratio of clay soil with PA1 for adsorbent immersed in salt water polluted with AGO. The regression equation of the line is given as $Y = 26.638 X + 219.45$ with the square root of the best fit $R^2 = 0.9877$ showing the reliability value of the Temkin isotherm of 98.77% as well as the equation of the line illustrating the slope value of 26.638 and intercept value of 219.45. The variation in the Langmuir isotherm of Q_e could be integrated to the variation in $\ln C_e$.

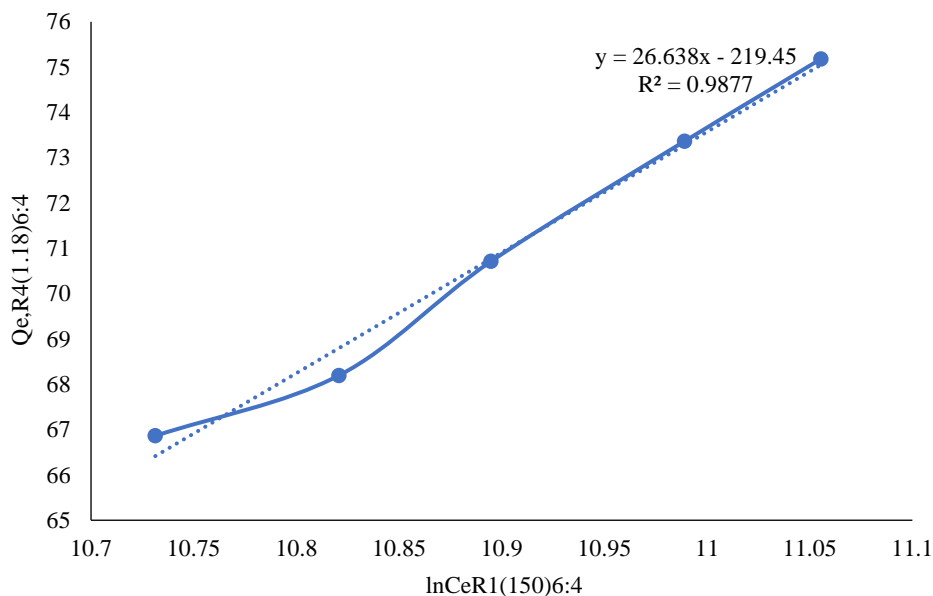


Figure 11. Temkin Isotherm Plot of Mix Ratio of 6:4 of Particle Size of 150 μm .

Figure 12 shows the relationship between the Freundlich isotherm plot of $\log Q_e$ against C_e for R4 (1.18 mm) 6:4 mix ratio of clay soil with PA1 for adsorbent immersed in salt water polluted with AGO. The regression equation of the line is given as $Y = 3E-06X + 1.6856$ with the square root of the best fit $R^2 = 0.9528$ showing the reliability value of the Freundlich isotherm of 95.28% as well as the equation of the line illustrating the slope value of 3E-06 and intercept value of 1.6856. The variation in the Langmuir isotherm of $\log Q_e$ could be integrated to the variation in C_e .

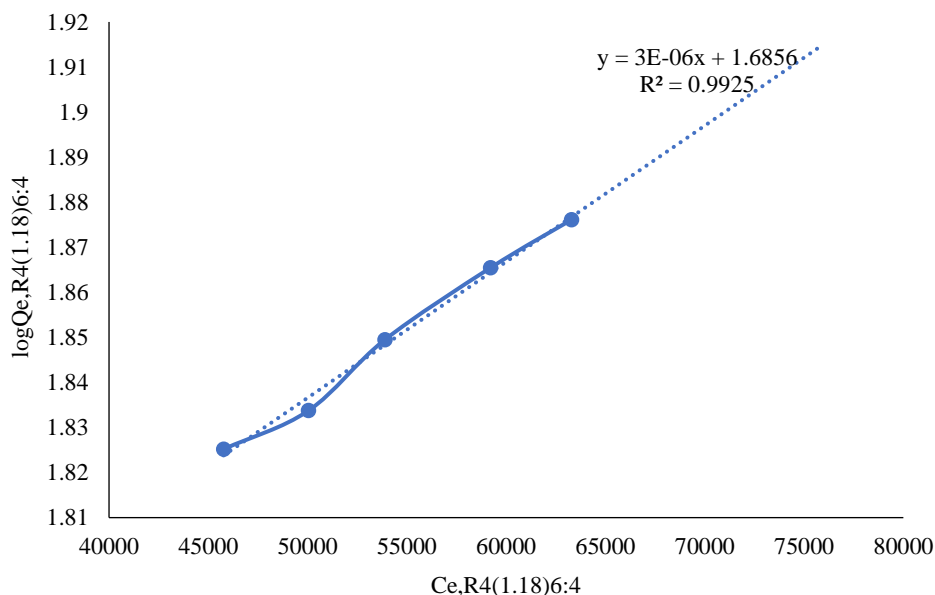


Figure 12. Freundlich Isotherm Plot of Mix Ratio of 6:4 of Particle Size of 600 μm .

CONCLUSION

The research revealed the following conclusion from the outcomes of the study, which include;

- i. The adsorption isotherms of adsorbent mix ratio of R1 PAI AGO 150 μm of R1:9 in contaminated salt water medium shows the Temkin isotherm value of a. as -208.95 and b. as 33.752 , whereas the Langmuir isotherms value of $K_1 = 0.022528611$ and AL as 0.000123907 as well as the Freundlich isotherm value of $K_f = 8.3261945$ and n as $5.00\text{E}+05$.
- ii. For adsorbent mixed ratio of R2 PA1 AGO 300 μm of R6:4 in polluted salt medium revealed that a and b coefficient have the value of $a = -391.94$ and $b = 49.435$ for Temkin isotherm, coefficients of AL and K_f are 0.00860363 and $4.04371\text{E}-05$ for Langmuir isotherm whereas for Freundlich isotherm we have $K_f = 7.109273084$ and $n = 2.50\text{E}+05$.
- iii. The Temkin isotherm value for a and b are $a = -252.68$ and $b = 27.593$, for Langmuir isotherm values are $K_L = 0.001568381$ and $AL = 1.41154\text{E}-05$ whereas for the Freundlich isotherm value calculated are $K_f = 4.387238562$ and $n = 2.50\text{E}+05$. The obtained showcases the adsorbent formulated of particle size and mix ratio of R3 PA1 600 μm , of R6:1 immersed in polluted salt water with AGO.
- iv. The effect of particle size and mix ratio of R4 PA1 AGO 1.18 mm of R6:4 immersed in a contaminated salt water medium revealed the following results as catenated in terms of coefficient of the adsorption rate isotherms value of the three models considered in this research as a and b values as $a = 219.45$ and $b = 26.638$ for Teinkin, $K_L = 0.003465484$ and $AL = 3.04963\text{E} - 05$ for Langmuir isotherm and for Freundlich isotherm value of $K_f = 5.3956874$ and $n = 3.33\text{E} + 05$.
- v. The investigation revealed that the isotherm coefficient of n from the Freundlich model shown the order of magnitude as $R1 \text{ PA1 AGO } 150 \mu\text{m} \text{ of } R1: 9 > R4 \text{ PAN AGO } 1.18 \text{ mm of } R6:4 > R2 \text{ PA1 AGO } 300 \mu\text{m} \text{ R6:4} = R3 \text{ PA1 AGO } 600 \mu\text{m} \text{ 6:4}$ as well as the K_f value of $150\mu\text{m} \text{ R1:9} (8.11326195) > 300 \mu\text{m} \text{ R6:4} (7.109273084) > 1.18 \text{ mm R6:4} (5.395687374) > 600\mu\text{m} \text{ R6:4} (4.387238562)$. From the rating the best performance adsorbent produced based on the Freundlich isotherm in terms of adsorbent of particle size of $150\mu\text{m}$ with mixed ratio of R1:9
- vi. The performance rating of the adsorption rate of isotherm of Temkin revealed and Langmuir isotherm followed the same trend, demonstrating the best fitting of the adopted concepts.

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