

Investigation of the Adsorption of Arsenic by Acid Modified Cassava Peels (*Manihot esculenta*)

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ABSTRACT

Removal of heavy metal ions present in water has been a challenge owing to the high cost of existing technologies. In this study, natural material (cassava peel) modified with acid was used to determine the feasibility of the removal of arsenic. Cassava peel (*Manihot esculenta*) is considered as a waste which in turn is seen as a form of environmental nuisance in many countries of the world. Arsenic is a naturally occurring heavy metal considered to be very toxic and poisonous to man and his environment. In this study, the adsorption of arsenic by acid modified cassava peel was carried out using the batch adsorption method. The effects of pH, contact time, adsorbent dosage and concentration on the adsorption of arsenic from an aqueous solution were investigated. The results showed that the highest adsorption efficiency was obtained at pH 4.0 with 0.2 g adsorbent dosage at 10ppm initial concentration with a contact time of 60 minutes. Adsorption of arsenic from the solution by the acid modified cassava peels decreased with increase in pH and adsorbent dosage, but it increased with increase in contact time. The results also showed that as the adsorbate concentration increased, the rate of adsorption of the arsenic metal ion decreased. These observations are attributed to the presence of active binding sites or functional groups in the cassava peel. The isotherm data were fitted into the Langmuir model. It can therefore be said that acid modified cassava peels can be used as an adsorbent for the removal of arsenic compounds in an aqueous solution.

Key Words: Adsorption, Arsenic, pH, dosage, cassava peels, adsorbent, isotherm

INTRODUCTION

Water is a very important substance used for various industrial and domestic purposes. Water is absolutely essential for the existence of man and all forms of life, but anthropogenic wastes from domestic and industrial sources have rendered many waters toxic and hazardous to man and other living organisms (Ball *et al.*, 2001). Industrial activities have caused massive contamination and distribution of heavy metals in water causing an increase in the exposure of humans to heavy metals (Kumar *et al.*, 2016). These heavy metal contaminants in drinking water poses a serious

threat to human life because of their toxicity, bio-accumulative nature, and persistence in the environment. The heavy metals contaminate the groundwater and surface water through natural processes and anthropogenic activities like Industrial, agricultural, mining, and domestic activities (Bilgin and Konanc., 2016; Cao *et al.* , 2019).

Arsenic (As) is a type of heavy metal that occurs naturally in soil and rock. It is present worldwide in trace quantities in groundwater, soil and plants. It is a toxic element that is categorized as class (1) human carcinogen and it occurs in the environment in both organic and inorganic forms (Hughes *et al.*, 2011). According to WHO (2011), the maximum concentration level of arsenic in drinking water is 0.01 mg/l, levels higher than this is considered harmful to human's health. There are two major pathways through which As enters the human body;

- Food chain pathway (Arsenic above permissible soil limit is absorbed by the plants and these plants consumed by human in form of food and by drinking of arsenic contaminated groundwater)
- Occupational exposure (industrial exposure of high concentration of As), Rahman *et al.* (2009).

According to WHO (2011) and United States environmental protection agency (USEPA, 2019), high concentration of As in drinking and domestic water can cause skin lesion and cancer, exposure to cardiovascular diseases, respiratory problems, neurological problems, reproductive and human development challenges in both children and adults. In recent studies, there has been an association between chronic arsenic poisoning and various effects, such as DNA impairment, inhibition of enzymatic activities, generation of reactive oxygen species, and tumor promotion (Sarkar and Paul, 2016).

Cassava (*Manihot esculenta*) is an edible perennial woody shrub that originally originated from South America and was later introduced to Nigeria during the sixteenth century (Ikuemonisan *et al.*, 2020). Cassava peel is the fiber part of the tuber that contains a little starch. Cassava processing generates lot of waste, which if not properly disposed can create sanitation and environmental problems (Olukanni and Olatunji, 2018). A survey conducted on cassava processing revealed that about 28% of the cassava peels generated during garri production has no use, hence the peels are either heaped in refuse dump or abandoned (Gin *et al.*, 2014).

Adsorption is defined as the adhesion (ability of a substance to stick) of a liquid or gas on the surface of a solid material forming a thin film on the surface. It can also be defined as the deposition of molecular species onto a surface (Clark *et al.*, 2000). Factors such as pH, adsorbate dosage, adsorbent and contact time of the adsorbent in the adsorption solution affect the rate of removal of adsorbates from solution (Wang *et al.*, 2010). Modification of adsorbents with acid or base also affects the rate of removal of adsorbates from solution (Stawinski *et al.*, 2017)

Materials And Methods

Materials: Distilled water, H₂SO₄, NaOH, Na₂HAsO₄.7H₂O, and cassava peels (*Manihot esculenta*) were used for this study.

Methods

Preparation of acid modified cassava peels (Adsorbent preparation)

The cassava peels (*Manihot esculenta*) were bought from mile 3 market in Port Harcourt, Rivers state, Nigeria. The cassava peel samples were washed, air dried and placed in oven at a temperature of 85 °C until they became very dried and crispy. The peels were ground into fine powder using a mechanical grinder and half of the sample was modified chemically with 0.1M of H₂SO₄ until pH 7 was obtained. It was oven dried again at 50 °C to remove physical characteristics.

Adsorbate preparation: A stock solution with concentration 1000ppm of the arsenic metal ion was prepared by dissolving 1.04 g of the metal salt (disodium hydrogen arsenate) into a 250 ml volumetric flask and filling the flask to its maximum volume with distilled water. Various adsorbate concentrations were obtained from the stock solution (10ppm-50ppm) by appropriate dilution of the stock with distilled water using the formula $C_1V_1 = C_2V_2$.

Where; C_1 = concentration of stock solution in ppm, C_2 = concentration of the working solution, V_1 = volume of working solution taken from the stock solution, V_2 = volume of measuring cylinder used in ml.

Batch Adsorption studies

Batch adsorption studies were conducted using 100 ml beakers into which the arsenic solution was poured in and the weighed ground cassava peel powder were added. The beakers were agitated on an electric orbital shaker at a constant speed of 200rpm at a shaking time of 60 mins, the samples were withdrawn and filtered using Whatman No. 1 filter paper, the filtrates were analyzed for residual arsenic concentrations using an Atomic Absorption Spectrophotometer (AAS). The percentage of arsenic removed by the cassava peels was calculated from the difference between the initial arsenic concentration (C_o) and the final arsenic concentration (C_f) with formula,

$$\% \text{ Removal} = \frac{C_o - C_f}{C_o} \times 100. \quad 1$$

The amount of arsenic ions adsorbed (q_e) was calculated with the formula; $\frac{v(C_o - C_f)}{m}$ 2

where; m = mass of the adsorbent v = volume of adsorbate

Effect of pH

The effect of pH was determined by weighing a constant adsorbent dosage of 0.2 g into a beaker there after measuring 20 ml from 10ppm of arsenic solution adding it into the beaker. The pH of the 10ppm arsenic solution was adjusted from pH 4 up to pH 9. This adjustment was done by adding few drops of 0.1M of H₂SO₄ and 0.1M NaOH solution. The mixture of the adsorbent and adsorbate in the beaker was covered and agitated on an electrical orbital shaker at 200rpm for 60 mins. The mixture was then filtered and the concentration of arsenic was determined using AAS.

Effect of adsorbent dose

The effect of adsorbent dose was determined by weighing various dosages from 0.2, 0.4, 0.6, 0.8, and 1.0 g of the powder cassava peel adsorbent into different beakers containing 20 ml of 10ppm arsenic solution. The pH was kept at a neutral pH 7, the beakers were agitated at a constant speed of 200rpm for 60 mins. There after, the mixture was filtered and the arsenic filtrate concentration was analyzed using AAS.

Effect of contact time

The effect of contact time was determined by weighing a constant adsorbent dose of 0.2 g, 20 ml from 10ppm arsenic solution at a neutral pH 7, the mixture was agitated at a constant speed of 200rpm at different contact time of 30 mins, 60 mins, and 90 mins respectively. There after, the mixture was filtered and the filtrate concentration of the arsenic solution was analyzed using AAS.

Effect of adsorbate concentration

The effect of concentration was determined by weighing a constant adsorbent dose of 0.2 g into a beaker containing 20 ml of different adsorbate concentrations ranging from 10ppm – 50ppm respectively. The mixture was covered and agitated at a speed of 200rpm at a constant time of 60 mins, after which the mixture was the filtered and the concentration of the arsenic filtrate was analyzed using AAS.

Results and Discussion

Table 1. represents obtained results at different pH with a constant dose of 0.2 g and constant time of 60 mins.

Table 1: pH variation and % Removal of the arsenic

Ph	Co	Ce	Co – Ce(x)	% Removal	Qe (F)
4	10ppm	5.040ppm	4.960	49.60%	0.496
5	10ppm	6.980ppm	3.020	30.20%	0.302
6	10ppm	7.823ppm	2.177	21.77%	0.218
7	10ppm	8.262ppm	1.738	17.38%	0.174
8	10ppm	8.757ppm	1.243	12.43%	0.124
9	10ppm	9.023ppm	0.977	9.77%	0.098

Where; Co = initial concentration of adsorbate, Ce = final concentration of adsorbate

Co - Ce(x) = initial minus final concentration of adsorbate

Qe(F) = amount of arsenic ions adsorbed.

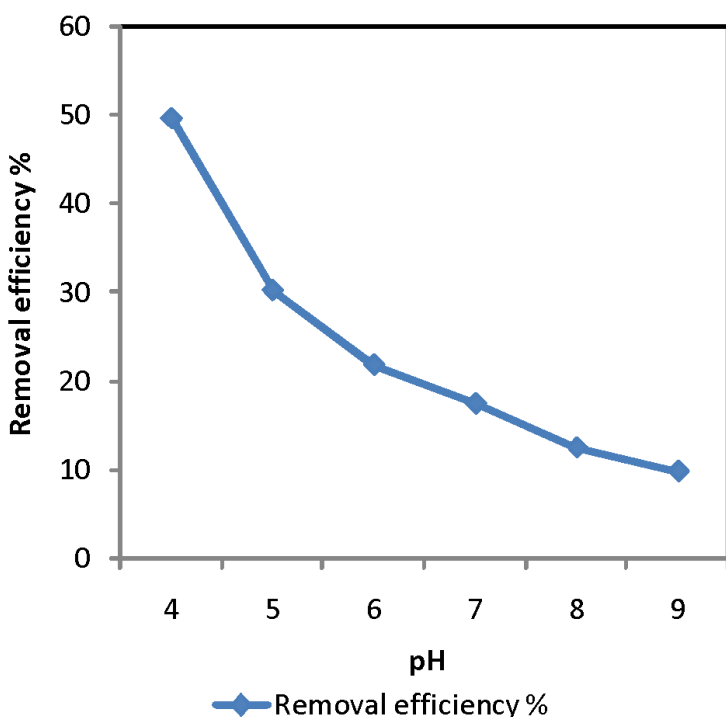


Figure 1: Plot of % Removal efficiency varying pH .

Table 2 represents obtained result at different adsorbent dosages at a constant concentration 10ppm, pH 7 and contact time of 60 mins.

Table 2: Adsorbent dose variation and % Removal of arsenic

Dosage	Co	Ce	Co – Ce(x)	% Removal	Qe (F)
0.2g	10ppm	4.934ppm	5.066	50.66%	0.5066
0.4g	10ppm	5.734ppm	4.266	42.66%	0.2133
0.6g	10ppm	6.723ppm	3.277	32.77%	0.1092
0.8g	10ppm	7.845ppm	2.155	21.55%	0.0538
1.0g	10ppm	8.545ppm	1.455	14.55%	0.0291

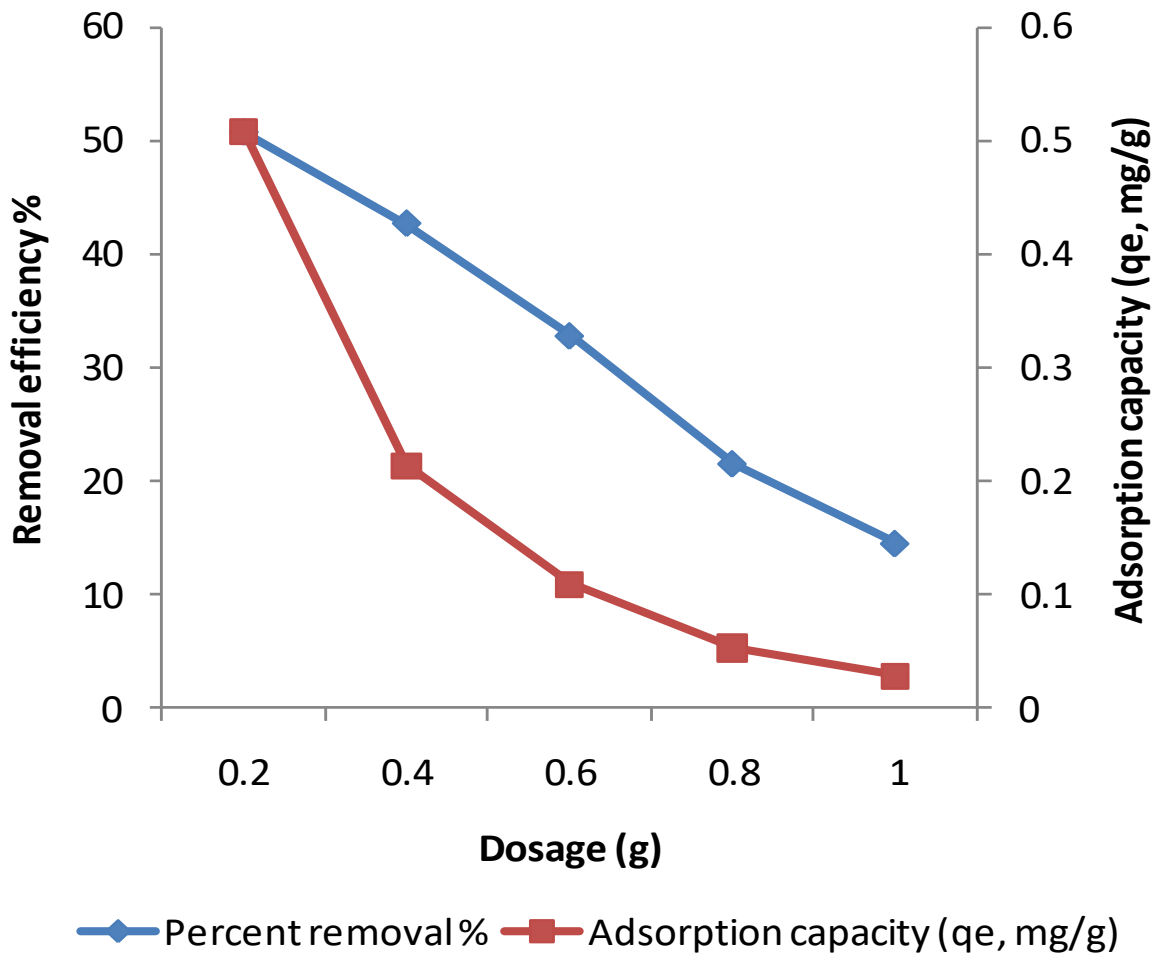


Figure 2: Plots of % Removal versus Adsorption efficiency varying adsorbent dosage.

Table 3 represents obtained results at different contact time and adsorbent dose of 0.2 g and concentration of 10ppm.

Table 3: Contact time variation and % Removal at 0.2 g dose of adsorbent and 10ppm solution.

Time	Co	Ce	Co – Ce(x)	% Removal	Qe (F)
30mins	10ppm	9.065	0.935	9.35%	0.0935
60mins	10ppm	6.416	3.584	35.84%	0.3584
90mins	10ppm	4.913	5.087	50.87%	0.5087

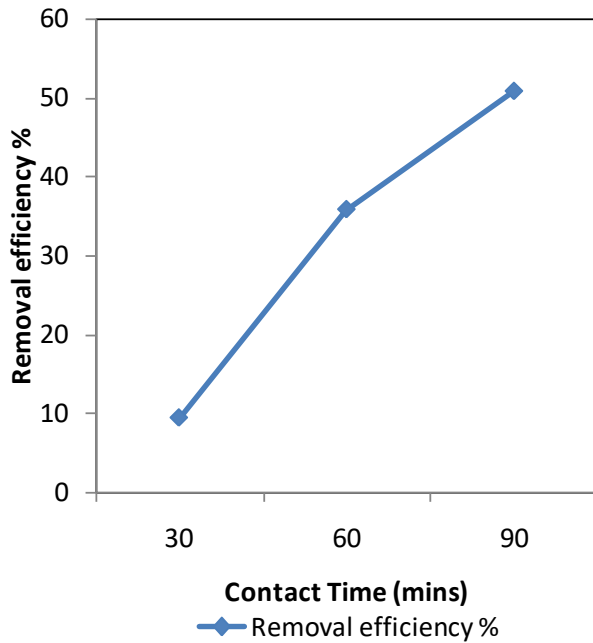


Figure 3: Plot of % Removal efficiency varying contact time.

Table 4 represents obtained results at different concentrations with a constant time of 60 mins and a constant dosage of 0.2 g.

Table 4: Adsorbate concentration variation and % Removal of arsenic

Concentration	Co	Ce	Co – Ce(x)	% Removal	Qe (f)
10ppm	10ppm	4.395ppm	5.605	56.05%	0.5605
20ppm	20ppm`	13.950ppm	6.050	30.25%	0.6050
30ppm	30ppm	23.183ppm	6.817	22.1%	0.6817
40ppm	40ppm	32.979ppm	7.021	17.552%	0.7021
50ppm	50ppm	42.979ppm	7.081	14.16%	0.7081

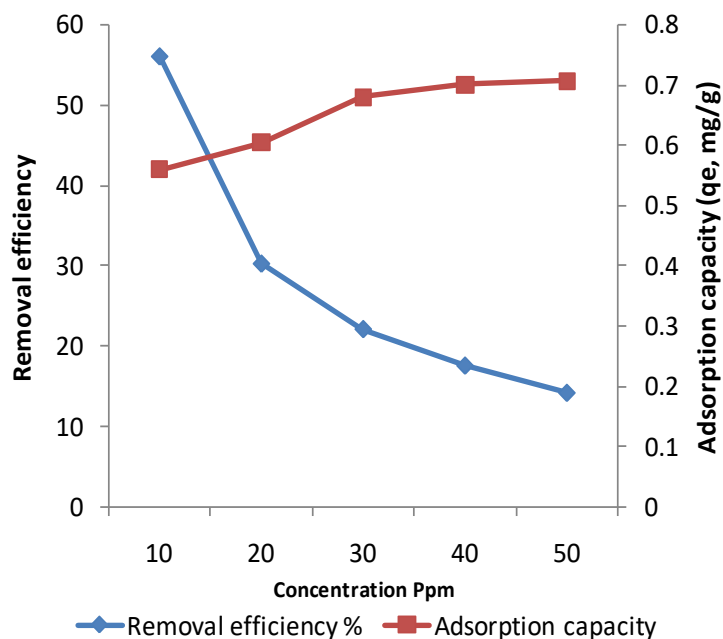


Figure 4: Plots of % Removal versus adsorption efficiency varying concentrations

Discussion

Effect of pH

The effect of pH was studied using 0.2 g of the adsorbent with pH values ranging from 4 – 9 and a concentration of 10ppm. Arsenic occur in both organic and inorganic forms, with the inorganic variants exhibiting various oxidation states (-3, 0, +3, +5) in response to specific redox conditions present in the environment. Nevertheless, the prevalent occurrences involve trivalent arsenite [As(III)] and pentavalent arsenate [As(V)] oxyanions (Singh and Pant, 2004). In disodium hydrogen arsenate, As exist as the pentavalent arsenate and +5 oxidation state. From figure 1, the percentage removal of ions was high at a low pH of 4, 5 and 6 which is the acidic medium. High removal efficiency at low pH is generally attributed to high electrostatic force of attraction between adsorbent surface and the adsorbate medium. In an acidic medium, Na_2HAsO_4 exist as AsO_4^{3-} , this will suggest that the already acidic adsorbent surface will be readily attract the oppositely charged arsenate by strong electrostatic force of attraction between unlike charges. A similar observation has be reported by Dittua *et al.* (2004) and they attribute this observation to arsenate acting like strong base in acidic medium. A decrease in adsorption efficiency is observed on increasing pH value, this sharp and steady decrease reported can be attributed to decrease in $[\text{H}^+]$ of the medium which may have led to progressively weakened electrostatic force of attraction between the adsorbent and the adsorbate. This observation is agreement with Babaeiveli and Khodadoust (2013) result.

Effect of adsorbent dosage

The effect of adsorbent dosage was studied using dosages ranging from 0.2 – 1.0 g and a concentration of 10ppm. As seen in figure 2, the result shows that as the adsorbent dosage increased the rate or percentage removal decreased. Similar result was reported by Padmavathy *et al.* (2016). Although, one would intuitively expect increase in adsorbent dose to increase binding sites and directly increase adsorption efficiency, this is rarely the case. Padmavathy *et al.* (2016) explained that increasing the adsorbent dosage leads to aggregation and overlapping of adsorbent, which lead to significant decrease in active binding site. Nduka *et al.* (2019) described and attributed this behaviour to increasing unsaturation resulting from overcrowding in the medium as adsorbent dosage is increased. 0.2 g of the adsorbent had the highest adsorption efficiency as seen on the result. This may also be because with a small surface area, the pores can be of large diameter and there is no undesirable pore blocking effect, Li *et al.*, (2002) made similar observations in their work.

Effect of contact time

The effect of contact time was studied using 0.2 g of the adsorbent and a concentration of 10ppm with different time of contact varying from 30 – 90mins. From figure 3 it can be seen that as the contact time increased the percentage removal of the adsorbate increased. As contact time increased, the adsorbate and adsorbent surface had more time to interact, bind and possibly cover more binding sites, thereby causing an increase in the biosorbent efficiency. Neupane *et al.*,(2015) gave similar report on this study.

Effect of adsorbate concentration

The influence of concentration of adsorbate on adsorption was studied using 0.2 g of the adsorbent at different concentrations ranging from 10ppm – 50ppm. The result from figure 4 shows that increase in adsorbate concentration was associated with decrease in the percentage removal. This observation may be attributed to availability of adsorption sites on the adsorbent; ie at low concentrations of the arsenate solution (adsorbate), there was more adsorption sites available. However, at higher concentrations a decrease in adsorption efficiency was experienced. This may be due to the saturation of arsenate ions at the surface of the adsorbent leading to competition over binding sites, Amuda *et al.*, (2007) and Dehghani *et al.*, (2021) also gave similar report. Inetainbor and Irabor (2021), also carried out a similar study by comparing the use of modified and unmodified cassava peels varying concentrations of nickel metal ion and concluded that at lower concentrations the adsorbent removed more metal ion from the solutions.

Adsorption isotherm

Adsorption isotherms pertain to a sequence of adsorption evaluations carried out at a specific temperature. These evaluations are then graphed to showcase the connection between quantities that have been adsorbed and those that haven't. The configuration of these isotherms contains substantial insights into the characteristics of the adsorption mechanism (Flat and Schober. 2012). Ali Riza *et al.* (2007) explained that the adsorption isotherm illustrates the state of equilibrium

between the dissolved adsorbate in the solution and the adsorbate adhered to the adsorbent, all under a constant temperature. The results obtained from this study were subjected to both Langmuir and Freundlich Isotherms. The Freundlich constants (Q_{max} , K_L) as well as the Langmuir constants (K_F , n) were all calculated from the slope and intercepts plot. The linear best-fit correlation (R^2) values were obtained and presented alongside the Freundlich and Langmuir constants in Table 5.

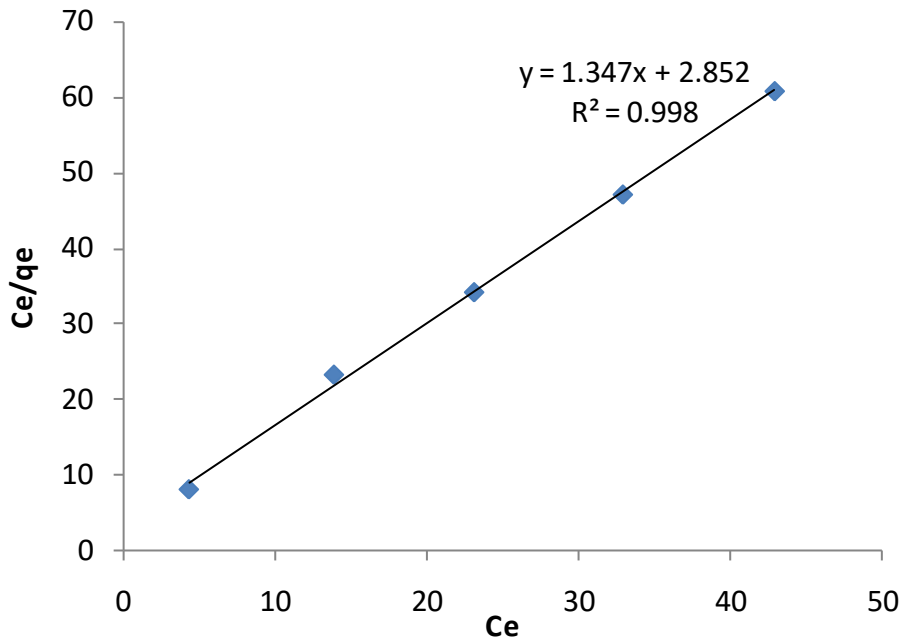


Figure 5: Linearized Langmuir isotherm plot for arsenic onto cassava peel (*Manihot esculenta*).

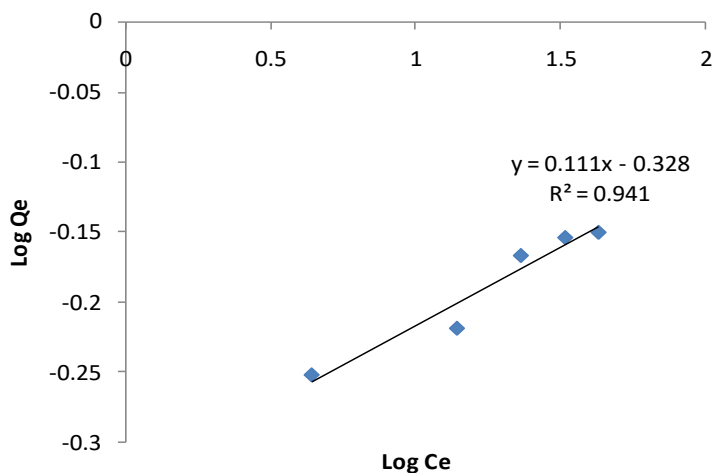


Figure 6: Linearized Freundlich isotherm plot for arsenic onto cassava peel (*Manihot esculenta*)

Table 5: Comparison of isothermal constants for adsorption of Arsenic

Isotherm model	Langmuir	Freundlich
	$Q_{\max} = 0.350$	$K_F \text{ (mg/g) (L mg}^{-1})^{1/n} = 0.720$
	$K_L = 2.12$	$n = 0.111$
	$R^2 = 0.998$	$R^2 = 0.941$

Table 6: Separation Factor(R_L) Values

Concentrations (Ppm)	R_L values
10	0.045
20	0.0231
30	0.0155
40	0.0116
50	0.0093

The results indicate that the coefficient of determination of Langmuir adsorption isotherm ($R^2 = 0.998$) was higher than that of Freundlich ($R^2 = 0.941$). Zahangir *et al.* (2007) in R^2 value of an adsorption isotherm suggest inclination and higher efficiency of the isotherm. The higher R^2 value in Langmuir isotherm indicate heterogeneity and compatibility of the isotherm data with the Langmuir isotherm. The value of R_L in Table 6 indicates that the isotherm plot fitted into Langmuir isotherm reason being ($0 < R_L < 1$) which is favorable. The Freundlich “n” value indicate the interaction and affinity of the adsorbate to the adsorbent. Timbo *et al.* (2007) reported that n values ranging from 1-10 indicates that the isotherm data will favour Freundlich isotherm. The values for n and K_f presented in Table 5 show that the Freundlich adsorption isotherm may not be able to simulate the data, since the value of n is less than 1, $n = 0.111$.

Conclusion: Modified cassava peel adsorbent (*Manihot esculenta*) is an effective adsorbent for the removal of arsenic ions from aqueous solutions. The adsorbent shows selectivity and adsorption efficiency towards the removal of the arsenic ion and it does not produce any toxic sludge. Optimum removal was observed at pH of 4 with an efficiency of 49.6% which maybe as a result of hydrogen proton competition with metal ion. The adsorbent dose affected the rate of adsorption, with an adsorbent dosage of 0.2 g at 10ppm with contact time of 60mins showing optimum efficiency of approximately 50.7%. The adsorption of metal ions onto the surface of the adsorbent increased with an increase in contact time and decreased with an increase in adsorbate concentration. The Langmuir adsorption isotherm was favorable and gave a better conformity to the isotherm data.

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