THE EFFECT OF ACID MODIFICATION OF COCONUT FIBRE BIOMASS ON THE ADSORPTION CAPACITY OF ZINC ION FROM AQUEOUS SOLUTION

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Abstract

The removal of Zinc (II) ion from aqueous solution using phosphoric acid modified coconut fibre was investigated. The study revealed that phosphoric acid modified coconut fibre could adsorbed Zn^{2+} ion from aqueous solution over a wide concentration range of up to 76.02 % from an initial 60 mg/L concentration. Langmuir and Freundlich adsorption isotherms were applied in this study and the data fitted more into Langmuir Isotherm model with R^2 value (0.918), maximum monolayer coverage capacity (Qm) (-25.64) mg/g and K_L (Langmuir isotherm constant) is (0.004) L/mg. The sorption study showed that phosphoric acid modified coconut fibre has potentials in the removal of heavy metal from wastewater and can be utilized as low cost adsorbent.

Keywords: Adsorbent, adsorption, fibre, Freundlich, Langmuir, isotherm, modified.

L'EFFET DE LA MODIFICATION ACIDE DE LA BIOMASSE DE FIBRE DE COCO SUR LA CAPACITÉ D'ADSORPTION DE L'ION ZINC D'UNE SOLUTION AQUEUSE

Resume

L'élimination de l'ion zinc (II) d'une solution aqueuse à l'aide de fibres de noix de coco modifiées à l'acide phosphorique a été étudiée. L'étude a révélé que la fibre de coco modifiée à l'acide phosphorique pouvait adsorber Zn^2+ ion d'une solution aqueuse sur une large plage de concentration allant jusqu'à 76,02 % à partir d'une concentration initiale de 60 mg/L. Les isothermes d'adsorption de Langmuir et Freundlich ont été appliquées dans cette étude et les données correspondaient davantage au modèle isotherme de Langmuir avec la valeur R^2 (0,918), la capacité de couverture maximale de la monocouche (Qm) (-25,64) mg/g et K_L (constante d'isotherme de Langmuir) est de (0,004) L/mg. L'étude de sorption a montré que la fibre de coco modifiée à l'acide phosphorique a un potentiel dans l'élimination des métaux lourds des eaux usées et peut être utilisée comme adsorbant à faible coût.

Mots-clés : Adsorbant, adsorption, fibre, Freundlich, Langmuir, isotherme, modifié.

تأثير تعديل الحمض في الكتلة الحيوية لألياف جوز الهند على قدرة امتصاص الزنك أيون من محلول مائي

إيرابور ، جودوين إيروموسيل

نبذة مختصرة

تمت دراسة إزالة أيون الزنك (II) من المحلول المائي باستخدام ألياف جوز الهند المعدلة بحمض الفوسفوريك. كشفت الدراسة أن ألياف جوز الهند المعدلة بحمض الفوسفوريك يمكن أن تمتص أيون Zn2 + من محلول مائي على مدى تركيز واسع يصل إلى 76.02٪ من تركيز أولي 60 مجم / لتر. تم تطبيق متساوي حرارة الامتزاز لانجموير وفريوندليش في هذه الدراسة وتوافقت البيانات بشكل أكبر في نموذج لانجموير ايزوثرم بقيمة R2 (0.918) ، وسعة تغطية أحادية الطبقة القصوى (Qm) (-25.64) مجم / جم و KL (ثابت متساوي الحرارة لانجموير) هي (0.004)) لتر / ملغ. أظهرت دراسة الامتصاص أن ألياف جوز الهند

المعدلة بحمض الفوسفوريك لها إمكانات في إزالة المعادن الثقيلة من مياه الصرف الصحي ويمكن استخدامها كممتاز منخفض التكلفة.

الكلمات الرئيسية: الممتزات ، الامتزاز ، الألياف ، Freundlich ،Langmuir ،isotherm ، معدل.

Introduction

The quest for a cost-effective, high-efficiency adsorbent to counteract the high increase in metal toxicity and its negative environmental impact is a technical and scientific priority. Organic (polymers/polymeric resins) and inorganic (sand, zeolites, alumina, etc.) adsorbents are used in adsorption. The selectivity of an adsorbent decides its effectiveness.

Heavy metals are metals with densities of more than 5 g/cm^3. These metals are high contaminants. Among the classes of contaminants, heavy metals deserve greater concern because of their high toxicity, accumulation and retention in the human body (Araújo *et al*., 2013). Moreover, heavy metals do not degrade to harmless end products (Nadeem *et al*., 2006; Obuseng *et al*., 2012). Heavy metals arise from different sources/activities carried out by industries (Sharma *et al*., 2014), which neglect the implementation of environmentally friendly industries waste disposal protocols tend to be the main agents for the contamination of water they discharge the waste with disregard to the future consequences there of (Obuseng *et al*., 2014). It is well established that the presence of heavy metals in the environment, even in moderate concentrations, is responsible for producing a variety of illnesses of the central nervous system (manganese, mercury, lead, arsenic), the kidneys or liver (mercury, lead, cadmium, copper) and skin, bones, or teeth (nickel, cadmium, copper, chromium) (Araújo *et al*., 2013; Zevenhoven and Kilpinen, 2001).

Contamination of water supplies due to reckless handling of heavy metals has been a cause of concern around the world for decades (Reddy et al., 2014). Heavy metals, unlike organic compounds, do not degrade to form harmless end products and are harmful to marine flora and fauna even at low concentrations (Shanker *et al*., 2009). Heavy metals that are significantly toxic to humans and ecological environments include Cadmium (Cd), Arsenic (AS), Chromium (Cr), Manganese (Mn), Copper (Cu), Mercury (Hg), Nickel (Ni), Iron (Fe), Zinc (Zn), and Lead (Pb), among others (Jarup, 2003). Some of these have the potential to be assimilated, stored, and concentrated throughout the human body, causing several health challenges like chronic, pulmonary problems, erythrocyte destruction, nausa, diarrhea, muscular cramps, and renal degradation (Jarup, 2002).

Adsorbents extracted from agricultural byproducts such as maize cob, onion peel, cassava waste, banana pith, coconut fiber, palm kernel fiber, and sugar cane baggase are examples of biosorbents (Igwe *et al*., 2005; Kinshnami *et al*., 2004). Biosorption is a mechanism in which inactive dead biomass binds and concentrates heavy metals from aqueous solution, and it is seen as an alternative technology for removing heavy metals and other contaminants from wastewater and industrial effluents (Naja *et al*., 2002). Because of the inclusion of polar functional groups such as aldehyde, ketones, phenolic acid, and carboxylic acid in a few molecular structures, these low-cost agricultural products are used (Qin *et al*., 2015). These promising agricultural waste materials are used in the removal of metal ions either in their natural form of after some physical or chemical modification (Okon *et al*., 2012).

Among several agricultural wastes studies as biosorbent for water treatment, coconut has been of great important as various parts of this tree (e,g fibre, shell etc) have been extensively studies as biosrobent for the removal of diverse type of pollutants from water. Coconut palm (*Cocos nucifera*) is a member of the family Arecaceae (palm family) the coconut palm is grown throughout the tropical world. It sustains the livelihood of millions of people in coastal regions of tropics. Because of their various applications, coconut palms have been nicknamed the "tree of life."

The husks, shells, leaves, and stem of the coconut palm tree, in addition to the useful material of the nuts, have a variety of uses. Because of its high potash content, the husk of the tree that covers the coconut is a source of coir fiber, which is called waste and leaves behind or even used as fertilizer. The raw coir fibre consists of 35.0% cellulose, 25.2% lignin, 7.5 % pentosans, 1.8% fats and resin, 8.7% ash contents, 11.9% moisture content and 10.6% other substances (Dan *et al*., 1993). This study investigates the effect of acid modification on the adsorption capacity of coconut coir fibre in the removal of zinc ion from aqueous solution

Plate 1: Coconut (Cocos nucifera L.) tree with fruits (Okon et al., 2012)

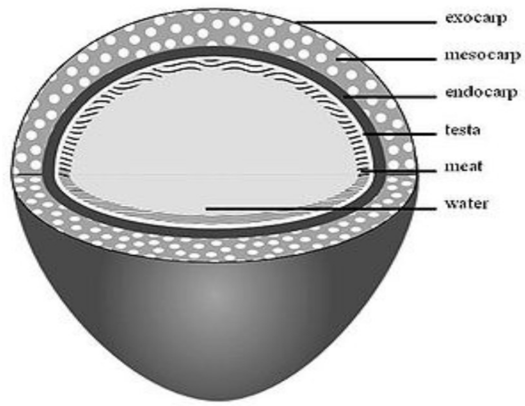

Plate 2: A Cross-sectional area of a Coconut fruit (An annotated diagram showing the mesocarp where the coir dust is generated) (Okon et al., 2012).

Materials and Methods

Samples Collection and Preparation

The coconut coir fibre was gotten from a coconut fruit obtained from a garden in Ekpoma, Esan West Local Government Area of Edo State. The coconut coir fibre adsorbent was prepared as described by Augustine *et al*., (2005). The raw fibre was dried in an oven at 80°C for 6 hours, pulverized and screened through a set of sieves to obtain particles of size 250 µm.

Modification of agro waste adsorbent was done so as to improve the adsorption affinity of adsorbate to the adsorbent. Acid modification of the biomass was done according to Ideriah *et al*. (2012), with slight modification. The finely sieved biomass was treated with 0.1M H_3PO_4 solution for 24 hours, followed by washing with deionized water until pH of 7.2 was achieved and oven dried at 60°C with constant mixing. The prepared biomass was stored in desiccators.

Synthetic wastewater preparation.

Synthetic wastewater was prepared according to Dada *et al*. (2012). The Preparation of adsorbate was carried out by preparing stock solution containing 1000 mg/L of Zn by dissolving 1.68g of $ZnSO_4$ in 1000cm^3 of distilled water. Working concentrations in the range of 25, 50, 75, 100, and 125 mg/L were prepared by serial dilution from stock solution.

Isotherm experiment

The equilibrium sorption of the Zinc (Zn) ion onto coconut coil fibre was carried out by contacting 0.5 g of the absorbent with 100 cm^3 of different concentrations of the adsorbate of 20 mg/L – 100mg/L in 250cm^3 Pyrex conical flasks, shaking intermittently for 60 minutes on the orbital shaker. The mixture was filtered and the residual concentration of the filtrate was analyzed using Atomic Absorption Spectrophotometer (2380 UNICAM AAS). The amount of adsorbed (mg/g) was calculated using the formulae reported by Dada *et al*., (2012).

$$q_e = \frac{V\,(C_i - C_e)}{W}$$

Where; q_e = the amount of solute adsorbed from the solution, V = Volume of the adsorbate, C_i = the concentration before adsorption, C_e = the concentration after adsorption and W = the weight in gram of the adsorbent.

The percentage sorption efficiency was determined by computing the percentage sorption using the formulae;

$$\%\,Sorption = \frac{(C_i - C_e)}{C_i}\,x\,100$$

Isotherm modeling

Two different models are considered in this study; Freundlich and Langmuir.

***Freundlich isotherm*:** Freundlich isotherm is one of the empirical formula, it takes into account that the surface is heterogeneous in nature and adsorption is multilayer to the binding sites located on the sorbent surface. The model of Freundlich is expressed as:

$$qe = KCe^{1/n}$$

K = mg/g or l/mg; 1/n or n = Freundlich constant related to adsorption capacity; n = Freundlich constant related to adsorption intensity.

Langmuir model: Langmuir model is assumed to be a monolayer adsorption of solutes on to the surface which is comprised of finite number of similar sites with the homogeneity in adsorption energy. The model of Langmuir is expressed as

$$q_e = \frac{q_{max} b C_e}{1 + Kb}$$

q_e = Amount of metal ion removed (mg/g); Ce = Equilibrium concentration (mg/L); *b* = Langmuir constant related to afinity; qmax = maximum metal uptake (mg/g) under the given conditions.

n, K = Freundlich and Langmuir constants (n value greater than 1.0 shows that sorption is favorable physical process) (El-Sikaily *et al*., 2011; Abdi and Kazemi, 2015).

Results and Discussion

Many authors have reported the application of unmodified agrowastes as adsorbent, however has been established that the use of unmodified agrowaste adsorbent has been proven to have low adsorption affinity and selectivity (Ibrahim *et al*., 2010; Johari *et al*., 2016). Raw agrowastes has shown to contain a large number hydroxyl (-OH) groups which has made the agrowastes to be readily functionalized with various chemical functional groups and reagents (Kumar *et al*., 2014). Functionalization or modification of agrowaste adsorbents have improve their adsorption affinity towards specific pollutants (Fu and Wang, 2011).

The adsorption of zinc ions from aqueous solution onto phosphoric acid modified coconut fibre was studied. Table 1 shows result of the concentrations of the Zn^{2+} ions before and after treatment with phosphoric acid modification of the agrowaste material (coconut fibre). The percentage sorption for Zn^{2+} ion onto the phosphoric acid modified and unmodified coconut fibre adsorbent is represented in figure 1, while figures 2 and 3 shows the graph of Langmuir and Freundlich Adsorption Isotherms respectively for Zn^{2+} ion.

Table 1: Results for the removal Zn2+ ions from aqueous solution using phosphoric acid modified/unmodified coconut fibre

S/NO	C_o (mg/L)	Modified		Unmodified	
		C_e (mg/L)	% Sorption	C_e (mg/L)	% Sorption
1	20	6.80	66.00	10.57	47.15
2	40	10.44	73.90	12.63	68.43
3	60	14.39	76.02	18.50	69.17
4	80	21.67	72.91	38.97	51.29
5	100	32.88	67.12	54.32	45.68

Phosphoric acid modified coconut fibre was found to have better adsorption of Zn^{2+} ion from solution over a wide concentration range than unmodified coconut fibre as shown in table 1. The effectiveness of acid modified sorbent removal up to about 76.02 % from an initial 60 mg/L. The sorbent was very effective in removing ions even at high concentrations of up to 80 mg/L. As the concentration is increased the percentage removal of curve decreased as showed in figure 1. The accessible binding sites are constant for a given amount of biomass added, so the same amount of metal ions would be bound. As the initial metal ion concentration increases, the percentage elimination of metal ions decreases (Mataka *et al*., 2010). Chukwu *et al*. (2017), reported a higher and relatively optimum percentage removal using kola-nut testa modified with formaldehyde as adsorbent when compared to the unmodified kola-nut testa adsorbent in the adsorption of Cu^{2+} and Fe^{2+} from single metal ion solution.

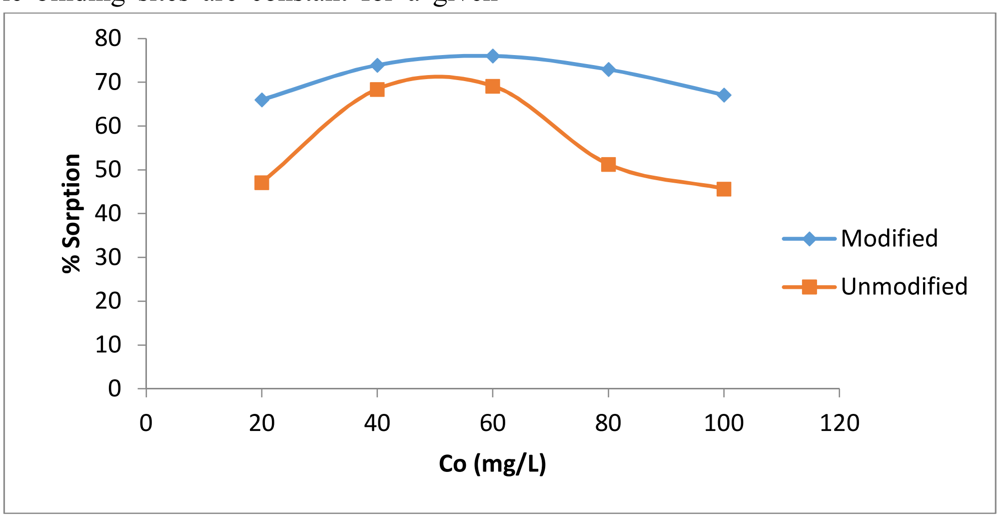

Figure 1: % sorption for Zn^{2+} ion onto phosphoric acid modified and unmodified coconut fibre

Adsorption isotherm describes the equilibrium relationships between adsorbent and adsorbate. The Langmuir (Hossain *et al*., 2012) and Freundlich (Ho and Mckay, 1999) adsorption isotherms were used to fit the equilibrium data. As a result, using theoretical or analytical equations to analyze experimentally collected equilibrium data is useful for functional adsorption device design and operation. In this analysis, the Langmuir and Freundlich adsorption isotherms were used. Its applicability was judged with the correlation coefficient (R^2). Table 3 shows the Langmuir and

Freundlich Isotherm constants for the adsorption of Zn^{2+} ion unto the acid modified coconut fibre.

Table 2 shows the parameters for the isotherms adsorption studies of Zn^{2+} ion onto the modified coconut fibre adsorbent.

Table 2: Parameters for plotting Langmuir and Freundlich Adsdorption Isotherms of Zn^{2+} ion onto phosphoric acid modified coconut fibre

S/NO	C_o (mg/L)	C_e (mg/L)	$1/C_e$	Log C_e	q_e (mg/g)	$1/q_e$	Log q_e
1	20	6.80	0.147	0.833	2.640	0.379	0.422
2	40	10.44	0.096	1.019	5.912	0.169	0.772
3	60	14.39	0.069	1.158	9.122	0.110	0.960
4	80	21.67	0.046	1.336	11.666	0.086	1.067
5	100	32.88	0.030	1.517	13.424	0.074	1.128

Table 3: Langmuir and Freundlich Isotherm constants for the adsorption of Zn^{2+} ion unto phosphoric acid modified coconut fibre.

Metal ion	Langmuir Isotherm			Freundlich Isotherm		
Zn^{2+} ion	Q_m (mg/g)	K_L (L/mg)	R^2	1/n	n	R^2
	-25.64	0.004	0.918	1.008	0.992	0.893

The Langmuir equation relates the coverage of particles on a solid surface to the concentration of a medium above the solid surface at a fixed temperature (Ayawei *et al*., 2017). This isotherm founded on the following assumptions, that is to say, adsorption is limited to monolayer coverage, all surface adsorption sites are the same with each site accommodating one adsorbed particle and the propensity of a particle to be adsorbed on a given site is independent of its neighboring sites occupancy. The basic assumption of Langmuir adsorption isotherm is based on monolayer coverage of the adsorbate on the surface of adsorbent (Wang *et al*., 2014) which is an indication of the fact that the adsorption of Zn^{2+} ions onto the modified coconut fibre generates monolayer formation with maximum monolayer coverage capacity (Qm) (-25.64) mg/g, K_L (Langmuir isotherm constant) is (0.004) L/mg, and the R^2 value (0.918) proving that the sorption data fitted well to Langmuir Isotherm model.

The Freundlich isotherm is able to describe the surface assimilation of organic and inorganic compounds on widely diverse adsorbents including biosorbent. Thus a favourable adsorption tends to have Freundlich constant *n* between 1 and 10. According to Yongde *et al*. (2016), larger value of *n* (smaller value of 1/*n*) implies stronger interaction between biosorbent and heavy metal while 1/*n* equal to 1 indicates linear adsorption leading to identical adsorption energies for all sites. This isotherm attempts to incorporate the role of substrate-substrate interactions on the surface (Febrianto *et al*., 2014; Reddy *et al*., 2014). Therefore the adsorption of Zn^{2+} ion on to adsorbent was favourable since 1/n value 1.008 while n was 0.992 indicating that the sorption of nickel ion unto the biomass is favourable and the R^2 value was 0.893.

Dada *et al*. (2012), reported that Langmuir adsorption model was found to be have the highest regression value when compared with Freundlich, Temkin and Dubinin–Radushkevich Isotherms in the equilibrium sorption of Zn^{2+} unto phosphoric acid modified rice husk.

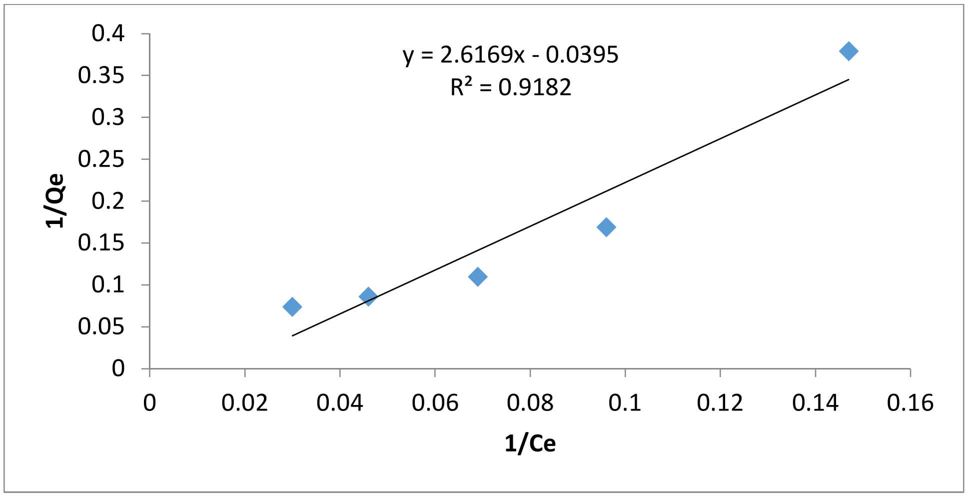

Figure 2: Langmuir Adsorption Isotherm for Zn^{2+} ion

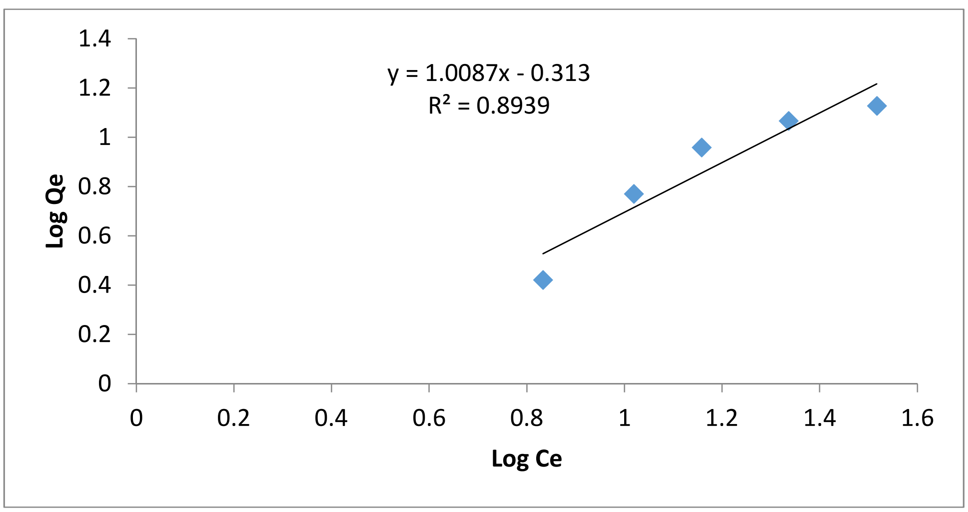

Figure 3: Freundlich Adsorption Isotherm for Zn^{2+} ion

Conclusion

In the investigation of the adsorption of Zn^{2+} ion onto phosphoric acid modified coconut fibre shows that the data fits into Langmuir isotherm more because of it highest regression value when compared with Freundlich isotherms. It could be concluded that this biosorbent has potentials in the removal of heavy metal from wastewater even over a wide range of concentration, hence can serves as suitable cheap and effective adsorbent for wastewater/water treatment.

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