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Removal of Zinc from Aqueous Solution by Adsorption using Coir Dust (Residue and Extract)

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Abstract
The adsorption behaviour of Zn (II) ions onto unmodified coconut coir dust residue and NaCl treated coconut coir dust extract (UCCR, UCCE, NaCCE and NaCCR) was investigated at 30^{o}C and pH of 7.0. The trend in the percentage adsorption of the adsorbate on the adsorbents decreased in the following descending: UCCR ≈ NaCCE>NaCCR> UCCE. There was justification in modifying the coir dust extract since this enhanced its adsorptive capacity. However, modification of the coir residue decreased its adsorptive capacity and not justifiable. The sorption data were also subjected to Langmuir, Freundlich and Flory-Huggins adsorption isotherm models. The adsorption of Zn(II) ions on NaCl treated coir residue (NaCCR) conform to the Langmuir isotherm model with high correlation value $R^2 > 0.90$. This indicated monolayer surface coverage of the Zn(II) ions on these adsorbents. The data for the adsorption on NaCl treated coir residue (NaCCR) and NaCl treated coir residue (NaCCR) at 50^{o}C fitted well to the Freundlich isotherm model implying a heterogeneous surface coverage of the adsorbents. All the adsorptions of Zn(II) ions on these adsorption did not fit the Flory-Huggins model. The adsorption increased with increase in temperature indicating an endothermic process. The negative value of ΔG^{o} signifies that the adsorption reaction was spontaneous process. The result indicate that the coir dust residue and modified coir dust extract could be employed as low-cost alternative for the removal of Zn(II) ions from solution.
Keywords: Adsorption, coir dust residue, isotherm, NaCl treated coir extract, zinc.

INTRODUCTION

Removal of heavy metals from industrial wastewater is of primary importance because they are not only causing contamination of water bodies but are also toxic to many life forms. Since most of heavy metals are non-biodegradable into non-toxic end products, their concentrations must be reduced to acceptable levels before discharging them into the environment. At least 20 metals are classified as toxic and half of these are emitted into the environment in quantities that pose risks to human health (Kortenkamp *et al.,* 1996). The ability of water body to support aquatic life as well as its suitability for other uses, however depends, on many trace elements.

Trace concentrations of zinc (Zn) are important for the physiological functions of living tissue and regulate many biochemical processes. However, just like other heavy metals, when Zn is discharged into natural waters at increased concentrations in sewage, industrial wastewater or from mining operation it can have severe toxicological effects on humans and aquatic ecosystems (Aremu *et al.,* 2002). A similar poisoning episode in Japan (Nitta, 1970).

Hence, it is essential to remove Zn from industrial wastewaters before transport and cycling into the natural environment. A number of technologies have been developed over the years to remove heavy metals from aqueous solution and industrial wastewaters. The most important technology includes precipitation, adsorption, ion exchange, reverse osmosis, electrochemical processes and membrane technology. However, these methods are uneconomical and inefficient, especially at metal concentrations in range from 10 to 100 mg/l (Basci *et al.,* 2004). Biosorption has been found to be economically feasible, rapid, reversible, and ecologically friendly in the removal of heavy metals from aqueous solutions, especially when used in the treatment of high volumes and low concentrations of wastewater Akar, containing heavy metals (Tunali and 2006, Yazici *et al.,* 2008, Melckova and Ruzorix, 2010). Agricultural byproducts which are available in large quantities may have potential to be used as low cost adsorbents. They represent unused resources that are widely available and environmentally friendly (Jeans and Dixon, 1992). Literature has shown that some agricultural wastes such as peanut skin (Randall *et al.,* 1978), onion skin (Kumar and Dara, 1982), maize cob (Odozi *et al.,* 1985) sugarcane bagasse (Krishanmi *et al.,* 2004). Coconut fibre (Bhatnager *et al.,* 2010).

Among, all these forms of agricultural by-products as adsorbent, coconut coir based adsorbents has proved to be very promising in recent years. The sorption properties of coconut shell are due to the presence of some functional groups, such as carboxylic hydroxyl and lactone which have high affinity for metal ions (Tan *et al.,* 1993). The use of coconut shell with surface modification to improve its metal removal performance would add to its economic valve, help reduce the cost of waste disposal and most importantly, provide a potentially in expensive alternative to existing commercial adsorbents such as activated carbon and synthetic resins (Babel and Kurniawan, 2004, Ng *et al.,* 2000, Amuda and Brahrin, 2006). The objective of this study was to investigate

Zn^{2+} ions biosorption by the residue and aqueous extract of coconut coir dust.

Coconut coir dust is the name given to millions of micro-spongy materials that constitute. The thick mesocarp or husk of the coconut (*Cocos nucifera L)* fruits. These are the fluffy materials that falls off when coconut husk is shredded in coir processing mills and are left as wastes of no industrial value. The present investigation is to extract coconut coir dust with water and therefore use the residue and extract as adsorbents for removing Zn^{2+} from aqueous solution studying the adsorptive capacity of the residue and extract by varying the agitation time, initial ion concentration.

Materials and Method

Coconut (*Cocos nucifera L.)* coir dust was procured from a local coconut processing mill in Uyo, Nigeria. It was air dried for 24 hours and in an oven at 60^{o}C to constant weight.

Extraction of Coconut Coir Dust with Hot Water

Coconut coir dust (10g) was boiled in 250ml of water in a beaker placed on hot plate. It was stirred continuously using magnetic stirrer for 12 hours to ensure complete extraction. After the coir dust residue was separated from the extract by filtration using filter paper Whatman No. 41. The filtrate and residue were dryness and the extract obtained was dried in an oven kept at 60^{o}C to constant weight.

Activation of Coir Dust Residue and Extract using NaCl

Five grammes (5g) of the dried residue was mixed with 20ml of 2 NaCl and the mixture kept for 24 hours. The mixture was filtered and the coir residue sodium chloride was air dried for 24 hours and in oven at 60^{o}C. The sample procedure was repeated for the coir extract. These adsorbents were stored in airtight hid containers.

Adsorption Kinetics of Zn(II) Ions on Adsorbents

A Zn^{2+} ion solution was prepared by dissolving solid Zinc nitrate hexahydrate $Zn(NO_3).6H_20$ in deionised water. The biosorption experiment was conducted in 250ml Erlenmeyer flasks containing 200mg of Na-activated coir residue with of Zn^{2+} ion 25ml solution with initial concentration of 10 mg/l. the flasks were agitated on a shaker at 140rpm at 30^{o}C for 0.5, 5, 10, 30, 60 and 90 mins respectively. the pH of the Cu^{2+} ion solution was maintained at pH 7. At the end of each time interval, each solution was filtered and the filtrate was analysed for the residual Zn^{2+} ion concentration. This procedure was repeated for the Na-activated coir extract. The removal efficiency (E) of adsorbent on Zn(II) is calculated as

$$\% \text{ Efficiency} = \frac{C_o - C_t}{C_o} \times \frac{100}{1} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots$$

Equilibrium Studies of Adsorption of Zn(II) on Adsorbents

The equilibrium isotherm was determined by mixing 0.2g of NaCl-treated coir residue in the concentration of 2, 6, 10 ppm of 25ml of Zn^{2+} ion solution. The different flasks containing the different concentrations of Zn(II) ion solution were shaken for 90 mins, sufficient to reach equilibrium with constant agitation speed of 140rpm at 30^{o}C.

The amounts of Zn^{2+} ions biosorbed by untreated coir residue/extract and residue/extract pre-treated with NaCl were calculated by the application of the equation

$$q = \frac{C_o - C_e}{m} \quad \frac{V}{1000} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots$$

in which C_o, C_e are the initial concentration (mg/l), C_e the concentration at equilibrium (mg/l), m is the dry weight of biomass (g), V is the volume of Zn(II) solution (ml), q is the quantity of metal ion bioadsorbed per of biomass.

Results and Discussion

Equilibrium time Study

Table 1 show increase in the percentage uptake of Zn(II) ions by the untreated coir residue/extract and NaCl-treated coir residue and extract with time. The untreated coir residue is the UCCR while the coir extract is UCCE while the NaCl-treated coir residue and extract are Na-CCR and Na-CCE respectively. It was observed that the biosorption on untreated and NaCl-treated coir residue and extract was rapid and attains equilibrium at 60 mins for the four adsorbents. The untreated coir dust residue removed more of Zn(II) ions than the NaCl-treated coir residue and became almost the same between 60-90 minutes when the sorption is already at equilibrium.

However, the amount of Zn(II) removed by the NaCl-treated coir extract Na-CCE were greater than that removed by the untreated coir extract (UCCE). This result justify the reason for the modification of the coir dust extract. It showed that it was not necessary to modify the coir dust residue. The unmodified coir dust residue

(UCCR) is a better adsorbent for the removal Zn(II) ions from aqueous than the unmodified coir dust extract (UCCE).

The coir dust residue (UCCR) is composed of lignin and cellulose while the coir extract is composed of tannins which is a flavonoid substrate. It has been reported that lignocellulosic materials remove more metal ions from aqueous solution than the extractive which is a non structural wood compound (Israel, 2012).

Adsorption Isotherms

Adsorption isotherm is the curve which relates the amount of adsorbate adsorbed per unit mass of adsorbent to the amount of unabsorbed adsorbate remaining in solution at equilibrium time. Several models have been developed to describe adsorption system behaviours. The result obtained on the adsorption of Zn(II) on unmodified and modified coir dust residue and extract were analyzed by the well known models given by Langmuir, Freundlich and Flory-Huggins the Langmuir model has been successfully applied to many pollutant adsorption processes and it is most commonly used adsorption isotherm for the adsorption of a solute from a liquid solution (Langmuir, 1916). The linear form of Langmuir isotherm is given by the following equation

$$\frac{C_e}{q_e} = \frac{1}{Q_o b} + \frac{C_e}{Q_o} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots$$

where Q_o and b are Langmuir constants related to adsorption capacity and energy of adsorption respectively. C_e and q_e are the equilibrium concentration and quantity absorbed respectively. Table 2 shows the Langmuir parameter's and correlation coefficients evaluated from the model at the temperatures of 30°C, 50°C and 70°C respectively. The values of the correlation coefficients indicate favourable conditions for adsorption of Zn(II) ions on unmodified coir extract (UCCE) at 70°C, NaCl modified coir extract (NaCCE), unmodified coir residue (UCCR) at 70°C and NaCl modified coir residue (NaCCR).

The results should that the adsorption of Zn(II) ions on unmodified coir extract (UCCE) was favoured at higher temperatures than lower one. It again demonstrate that modified coir dust residue (UCCR) was a better adsorbent for the removal of Zn(II) ions from aqueous solution than the coir dust extract whether treated or not.

According to Hall *et al.,* (1966), the essential features of the Langmuir isotherm can be expressed in terms of a dimension less constant separation factor or equilibrium parameter K_L which is defined by the following relationship:

$$K_L = \frac{1}{1+bc_o} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots$$

where K_L is a dimensionless separation factor, c_o is the initial concentration (mg/l), b the Langmuir constant (L/mg). The parameter K_L indicates the shape of the isotherm and nature of the adsorption process ($K_L > l$, unfavourable, $K_L = 1$, linear; $0 < K_L < 1$, favourable; $K_L = 0$: irreversible). The value of K_L obtained were in between 0 and 1 indicating the favourable adsorption of Zn(II) onto modified coconut coir extract (UCCE) and modified coir residue (NaCCR).

The adsorption data obtained were also analysed with the Freundlich isotherm model. The Freundlich isotherm (Freundlich, 1906) is the earliest known relationship describing the sorption equation. This fairly satisfactory empirical isotherm can be used for non-ideal sorption that involves heterogeneous sorption. The linear form of Freundlich isotherm is given by the following equation:

$$\log q_e = \frac{1}{n}\log + \log K_F \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots$$

where q_e is the amount adsorbed per unit mass of adsorbent (mg/g), C_e the equilibrium concentration of the adsorbate (mg/l), K_F and n are Fruendlich equilibrium coefficients. The plot of $\log q_e$ versus $\log C_e$ enables the constant K_F and $\frac{1}{n}$ to be determined from the slope and intercept of the plot. These constants K_F and $\frac{1}{n}$ with correlation constants R^2 for this model are presented in Table 7. The high correlation constants (R^2) for the adsorption of Zn(II) on NaCl treated modified coir dust residue (NaCCR), untreated coir dust extract (UCCE) at 50°C, 70°C, NaCl treated UCCE at 30°C, 70°C and unmodified coir residue (UCCR) at 70°C showed that these adsorptions conforms to this model. These results indicate that adsorption of Zn(II) ions on unmodified coir extract (UCCE) and NaCl modified coir residue (NaCCR) is temperature dependent showing endothermic reaction. Isreal and Eduok (2012) had reported an endothermic adsorption of Zn(II) ions on

unmodified coconut coir dust.

The Flory-Huggins model has been to describe the sorption of Zn(II) ions on the unmodified and modified coir dust extract and residue. This model which helps to account for the degree of surface coverage characteristic of the adsorbate on the adsorbent is expressed in the linear form as:

$$\log\left(\frac{\theta}{C}\right) = \log K_{FH} + \alpha_{FH} \log(I - \theta) \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots$$

where θ is the surface coverage of the adsorbent by the adsorbate.

$\theta = \frac{(1-C_e)}{C_o}$, where C_o and C_e are the initial and equilibrium Zn(II) concentrations respectively. The plot of $\log\left(\frac{\theta}{C_0}\right)$ versus $\log(1-\theta)$ should be linear if the adsorption conforms to this model and the equilibrium constants K_{FH} and α_{FH} were obtained from the intercept and slope of the plot are shown in Table 4. Furthermore, the equilibrium constant K_{FH}, obtained from the Flory-Huggins models is used to compute the Gibbs free energy for the adsorption process. The Gibbs free energy is related to the equilibrium constant as follows:

$$\Delta G^{\circ} = -RTlnK_{FH} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots$$

where R is the universal gas constant, 8.314J/K/mol, T is absolute temperature (K) and K_{FH} is equilibrium constant from Flory-Huggins isotherm equation. The ΔG° is equilibrium constant from Flory-Huggins isotherm equation. The ΔG° for Zn(II) adsorption onto untreated and treated coir dust extract and residue (UCCE, NaCCE, UCCR and NaCCR) in this study were computed and shown in Table 4. The negative values of ΔG° for the adsorption of Zn(II) ions on untreated and NaCl treated coir extract and residue showed that it is a spontaneous and endothermic process. Karthikeyan *et al.,* 2004, Israel and Eduok, 2012 had reported endothermic adsorption of Zn(II) ions onto chitosan and coconut coir dust, respectively. The increase in the adsorption of Zn(II) ions with increase in temperature might be due to the acceleration of some originally slow adsorption steps or creation of some active sites on the adsorbent surface.

Effect of temperature on adsorption of Zn(II) onto coir dust extract and residue. The effect of temperature on the adsorption of untreated and NaCl treated coir dust extract and residue (UCCE, NaCCE, UCCR and NaCCR) are shown in Table 5.

The percentage adsorption of Zn(II) on these adsorbents were calculated for the temperatures of 30^{o}C, 50^{o}C and 70^{o}C respectively. The result showed that the adsorption of untreated coir dust extract (UCCE) increase with rise in temperature indicating an endothermic adsorption. However for the NaCl treated coir dust residue (NaCCR) the adsorption seemed to remain constant at lower temperatures but slightly decreased as temperature increases indicating exothermic reaction.

The adsorption of NaCl treated coir dust residue (NaCCR) increased with increase in temperature whereas the NaCl treated coir extract show little or no change with temperature. This demonstrate that these two adsorbents untreated and treated coir residue and extract have different constituents. The coir residue (UCCR) being a lignocellulosic material whereas the coir dust extract in composed mainly of tannins a non woody material.

Conclusion

The adsorption of Zn(II) ions on the unmodified and NaCl modified coir dust residue and extract increased with increase in contact time. However, the trend of adsorption decreased in the following descending order unmodified coir residue (UCCR $\approx$ NaCl modified coir extract (NaCCR) > NaCl modified coir residue (NaCCR) > unmodified coir extract UCCE respectively. Hence, there was need to modify coir dust extract (CCE) and not necessary to modify the coir residue.

The adsorption of Zn(II) ions on NaCCE, the modified extract, and NaCCR the modified coir residue confirm to the Langmuir isotherm with high correlation values of R^2 >0.90. Also the adsorption on NaCl treated coir extract (NaCCE) conform to the Freundlich isotherm model with correlation constant R^2 of 0.974 at 50^{o}C while the NaCl treated coir residue (NaCCR) confirm to this model at all temperatures of 30^{o}C, 50^{o}C and 70^{o}C respectively. The adsorption of Zn(II) on these adsorbents increased with increase in temperature indicating an endothermic reaction and with negative value of ΔG, the Gibbs free energy confirming the adsorption to be spontaneous.

Table 1: Percentage Adsorption of Zn(II) on untreated and Na-treated coir dust residue and extract

Time (mins)	UCCR	Na-CCR	UCCE	Na-CCE
0.5	93.10	87.40	85.40	93.0
5.0	93.90	87.50	88.80	93.40
10.0	94.70	88.50	90.10	94.10
30.0	94.90	93.30	90.40	94.70
60.0	97.20	97.30	91.50	94.90
90.0	97.20	97.30	91.50	94.90

Initial conc. of Zn(II) = 10mg/l, pH = 7.0, dosage = 0.2g, particle size 100-106µm, temp. = 30^{o}C

Table 2: Langmuir isotherm parameters for Zn(II) Ions on unmodified and NaCl modified coir dust residue/extract

Adsorbent	Temp. ^{o}C	Q_o(mg/g)	b(L/mg)	R^2
	30	0.361	0.785	0.329
UCCE	50	0.044	1.568	0.765
	70	2.762	1.016	0.191
	30	7.194	0.222	0.996
NaCCE	50	0.0736	1.347	0.992
	70	0.960	0.5419	1.00
	30	0.205	1.282	0.370
UCCR	50	1.519	0.445	0.170
	70	6.037	1.378	0.961
	30	0.522	0.436	0.998
NaCCR	50	0.674	1.958	0.999
	70	2.392	1.251	0.499

Table 3: Freundlich isotherm parameters for Zn(II) Ions on unmodified and NaCl modified coir dust residue/extract

Adsorbent	Temp. ^{o}C	K_F(mg/g)	$\frac{1}{n}$	R^2
	30	1.492	2.771	0.586
UCCE	50	287.07	11.21	0.964
	70	1.559	0.753	0.845
	30	2.013	0.547	0.816
NaCCE	50	12.330	0.151	0.974
	70	1.352	0.632	0.49
	30	6.886	3.800	0.625
UCCR	50	1.291	1.298	0.584
	70	1.559	0.753	0.954
	30	0.482	0.503	0.924
NaCCR	50	50.699	0.124	0.929
	70	6.591	0.787	0.934

Table 4: Flory-Huggins isotherm parameters for Zn(II) adsorption on unmodified and NaCl modified dust residue/extract

Adsorbent	Temp. ^{o}C	K_F(mg/g)	α_{FH}	R^2	ΔG^{o} (kJ/mol)
	30	0.596	0.041	0.727	-2768.01
UCCE	50	0.289	0.572	0.797	-482.43
	70	1.658	1.690	-	-6017.09
	30	8.729	0.474	0.074	-219.50
NaCCE	50	1.432	0.029	0.952	-16.70
	70	1.476	0.052	0.007	-841.53
	30	0.261	0.052	0.797	-3289.99
UCCR	50	0.336	0.086	0.875	-381.70
	70	0.06	0.033	0.724	-1693.91
	30	2.183	0.671	0.596	-426.88
NaCCR	50	2.158	0.023	0.164	-870.31
	70	93.54	1.464	0.349	-2323.88

Table 5: Percentage adsorption of Zn(II) at equilibrium for unmodified coir residue/extract and modified NaCl modified coir residue and extract at different temperatures

$C_{o(mg/l)}$	30°C				50°C				70°C			
	UCCE	NaCCE	UCCR	NaCCR	UCCE	NaCCE	UCCR	NaCCR	UCCE	NaCCE	UCCR	NaCCR
2.0	71.0	90.50	79.50	67.50	73.50	73.50	79.50	80.50	90.0	74.50	62.0	96.50
6.0	90.0	94.0	91.66	83.66	90.50	89.0	91.66	92.66	91.0	91.16	84.66	96.50
10.0	92.0	96.20	93.50	83.20	94.0	93.0	93.50	95.40	93.60	94.10	88.60	99.10
15.0	92.0	93.40	94.20	88.46	95.73	95.20	94.20	96.60	95.0	91.16	92.06	98.06

Time= 90 mins, dosage = 0.2g, particle size = 100-106μm

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