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Adsorption Equilibrium Study of Lead and Zinc on Rice Husk from Aqueous Solution

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ADSORPTION EQUILIBRIUM STUDY OF LEAD AND ZINC ON RICE HUSK FROM AQUEOUS SOLUTION

Strictly as per the compliance and regulations of :

Adsorption Equilibrium Study of Lead and Zinc on Rice Husk from Aqueous Solution

Onipede, O. J.[α], Oshodi, A. A.[σ] & Enahoro, P. O.[ρ]

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Keywords: metal adsorption; synthetic water; rice husk; varied sizes.

I. Introduction

Effective removal of lead and zinc ions in effluents as a result of industrial, agricultural and mining is a problem; because they find their way into water bodies and their activities are persistent in the environment and are very toxic, and usually manage to get into potable water. Lead accumulation in the human body has been the major cause of dysfunction of kidney, liver, central nervous system, anaemia, high blood pressure, depression and reduced intelligence quotient in children [1; 2]. Zinc accumulation in the human body leads to electrolyte imbalance, nausea, anaemia and lethargy [3]. Lead and zinc metal removal in waste water has been effected by several methods in the time past including chemical precipitation [4], filtration [2], ion exchange [5], reverse osmosis [1], coagulation/flocculation [6] and adsorption [7]. All these methods have their demerits which include high cost of operation, which is not sustainable by small scale industries and also clean up of chemical treatment is difficult if not almost impossible. However out of the all the available methods of effluent treatment, biosorption seems to be the cheapest, fastest and most effective method of remediation of waste water.

Recently some materials have been used for biosorption in waste water remediation which include sugar cane bagasse [8], maize tassel [9], brick [10], clay [7], alumina [11], bark [12] zeolite tuff [13]. Others include walnut shell [6], pineapple peel [14], wheat bran, corn cob and human hair [4], coconut husk, bean chaffs [2], coal [15], kaolinite, illite [16], egg shell and activated carbon [17]. However much attention is focused on bio--adsorbent as a standard. Adsorbents have the advantage of being biodegradable, thus are environmentally friendly and remove toxicants by adsorption, ion exchange and metabolic reaction but a little attention is paid on rice husk which is a universal waste product in zinc and lead remediation of waste water. The few researchers that have examined the potential of rice husk did not examine the effect of variation of particle sizes on the adsorption process.

Hence this research examines the effect of varied particle sizes on adsorption of lead and zinc on using various particle sizes of the rice husk and also the efficiency in remediating waste water, also to examine the effect of change of pH, temperature, adsorbate concentration and time on adsorption capacity of rice husk.

II. Materials and Methods

The rice husk was purchased in the market in Abeokuta in Ogun state south west Nigeria and was washed in tap water, distilled water and de-ionized water respectively after which it was spread on polythene materials in the laboratory to air dry and later oven dried for 24 hours at 105°C, it was then ground and sieved into various particle sizes viz; 150 and 500 micron sizes. All reagents and standards were purchased from Sigma Aldrich Germany. Lead chloride and zinc chloride were used to prepare the lead and zinc standard solutions respectively. The 1000 mg/L standards were prepared by first preparing 0.1 molar of sodium acetate solution. The 0.1 molar sodium acetate solution was used to prepare the 1000 mg/L of the stock standard solution. The 100 mg/L solution was produced from the 1000 mg/L stock by dilution with de-ionized water. 0.5 g of the 500 micron size rice husk was placed in a 100 ml screw cap bottle and 50 mL of the 100 mg/L standard of the lead solution was added to it and was then place in temperature controlled water bath with shaker and was shaken for 30 minutes. It was then filtered and the filtrate was analysed on the atomic absorption spectrometer (AAS). The procedure was done in triplicates and the mean concentration was obtained. The procedure was repeated for 150 micron size and analysed by AAS.

The extent of metal ion uptake on 0.5 g of biosorbent of both micron sizes was examined with 50

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mL test solution of lead and zinc respectively, with concentrations between 100-250 mg/L for 30 minutes. They were filtered with whatman filter paper and the filtrate was analysed with AAS.

Effect of temperature was examined on 0.5 g of biosorbent of both micron sizes with 50 mL test solution of lead and zinc respectively, between 40- 80°C for 30 minutes, after which they were filtered with whatman filter paper and the filtrate was analysed with AAS.

Time variation effect on biosorption was examined on 0.5 g of biosorbent for both micron sizes with 50 mL test solution of lead and zinc respectively, between 30 – 150 minutes and were filtered with whatman filter paper and the filtrate was analysed with AAS.

Effect of pH was examined on biosorption on 0.5 g of biosorbent of both micron sizes with 100 mg/L of 50 mL test solution of lead and zinc respectively, between pH 3 - 9, after which they were filtered with whatman filter paper and the filtrate was analysed with AAS.

III. Result and Discussion

a) Effect of Concentration

The adsorption of zinc ion in synthetic solution was examined with increase in zinc concentration on rice husk of 150 and 500 micron sizes. it was observed that the amount of ions adsorbed increased with increase in concentration though not stepwisely as the concentration increased from 150 – 300 mg/L, which seems to suggest that the adsorption increased as the concentration increased, also the adsorption per unit mass was maximum at the highest concentration for both particle sizes. The adsorption in 150 and 500 micron sizes showed similar trends and the t-test showed that there was no significant deference between the adsorption by the two particle sizes. The effect of varied concentration of zinc on rice husk varied particle sizes is as shown in figure 1.

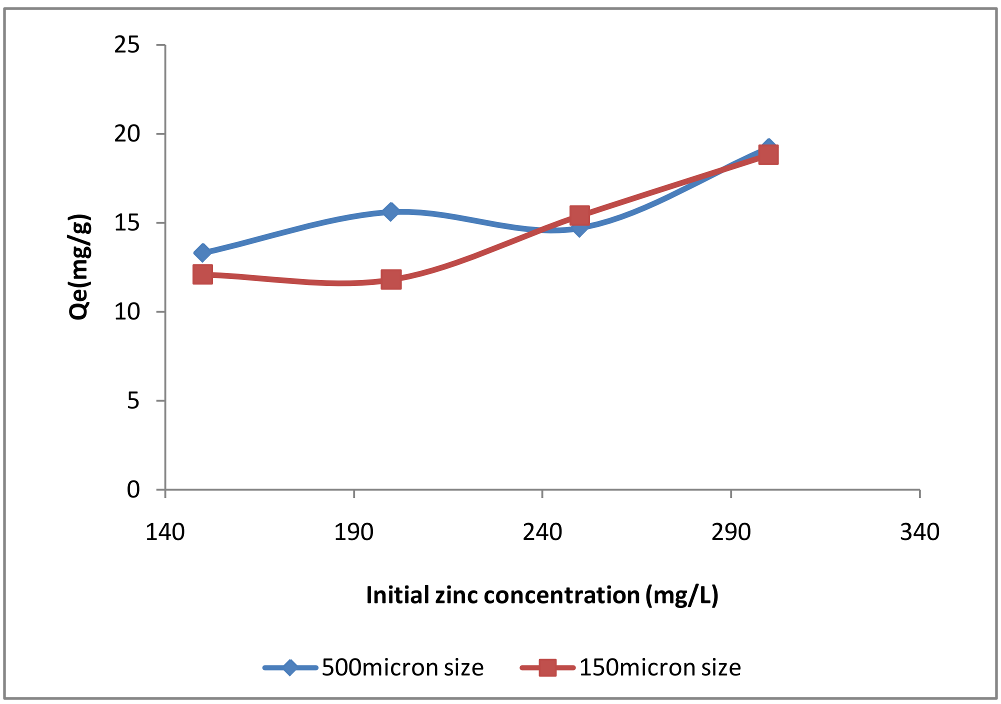

Figure 1 : Showing effect of varied concentration of zinc on rice husk of varied particle sizes

The effect of increased concentration was examined on lead biosorption, on both particle sizes. As the concentration increased, it was observed that the adsorption increased with increase in lead concentration though not stepwisely in 500 micron size, but was stepwise on 150 micron size rice husk. The adsorption per unit mass was maximum at the 300 mg/L concentration for both particle sizes. Nevertheless the paired t-test of adsorption of both particle sizes showed that there was no significant difference between the adsorption of both particle sizes. The effect of varied initial lead concentration on rice husk of varied particle sizes is as shown in figure 2.

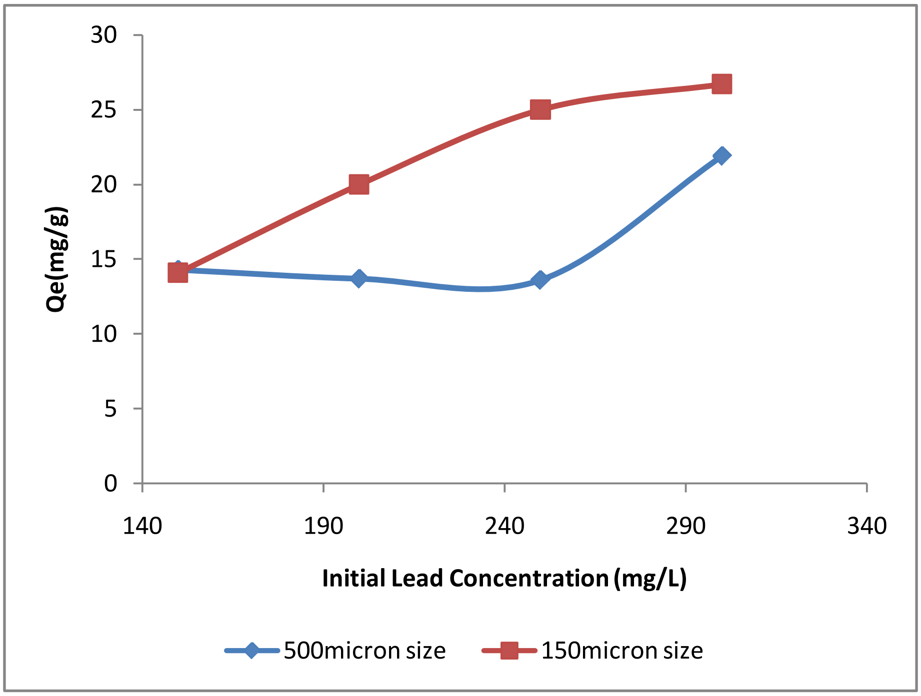

Figure 2 : showing effect of varied concentration of lead on rice husk of varied particle sizes

The Langmuir adsorption isotherm was examined on zinc solution on the 150 and 500 micron sizes of rice husk respectively, the Langmuir model assumes the adsorption of an ideal gas on the ideal surface occurs only on a fixed number of sites, each molecule forms a monolayer sorption on each site and no interaction is between adjacent sites. The Langmuir adsorption is based on the equation.

$$\frac{Ce}{qe} = \frac{1}{KL} - \frac{aL}{KL}Ce$$

Where Ce is the concentration of the adsorbate solution at equilibrium (unit in mg/L) and qe is the mass of adsorbate adsorbed per unit mass of the adsorbent at equilibrium (unit in mg/g) [9]. When $\frac{Ce}{qe}$ is plotted against Ce the slope is $\frac{aL}{KL}$ which is the theoretical saturation capacity (unit in mg/g) and the intercept of the slope is $\frac{1}{KL}$. KL is the equilibrium constant. The result is as tabulated below.

Table 1: Langmuir adsorption isotherm of zinc on rice husk

Rice Husk Size	R^2	$\frac{aL}{KL}$	$\frac{1}{KL}$
500micron size	0.931	0.06	0.42
150micron size	0.737	0.05	1.52

From the table above we could observe that Langmuir adsorption isotherm was obeyed as the correlation coefficients (R^2) were above 0.5, while $\frac{aL}{KL}$ in both cases, the saturation capacity were very low, while the intercept were also low.

The Langmuir adsorption isotherm was also obeyed for lead in 150 and 500 micron sizes of rice husk and the result obtained is as tabulated below.

Table 2 : Showing Langmuir adsorption isotherm of lead on rice husk

Rice Husk Size	R^2	$\frac{aL}{KL}$	$\frac{1}{KL}$
500micron size	0.886	0.07	–0.21
150micron size	0.799	0.04	0.034

From the table above, we could observe that Langmuir adsorption isotherm was obeyed as the regression coefficients (R^2) were above 0.5 while $\frac{aL}{KL}$ the saturation capacity were very low while the intercept was very low.

The Freundlich adsorption isotherm of the adsorption of zinc on rice husk of particle sizes 150 and 500 micron sizes were also examined. The Freundlich model is used for heterogenous surface energy system and with highly interactive species. The Freundlich model is represented by the equation.

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

Where Ce is the concentration of the adsorbate solution at equilibrium (unit in mg/L) and qe is the mass adsorbed per unit mass of the adsorbent at equilibrium (unit in mg/g), K is an indicator of the adsorption capacity, and n is the adsorption intensity which varies with heterogeneity of the material [9: 18]. When qe is plotted against Ce the slope is K and the power of the slope equation gave 1/n.

The result of the Freundlich isotherm is as tabulated in table 3 below.

Table 3 : Showing Freundlich isotherm of Zinc on rice husk

Rice Husk Size	R^2	K	n
500micron size	0.507	9.35	7.83
150micron size	0.449	5.06	4.12

It could be observed from the table above that the regression coefficient (R^2) were very low (<0.5), which is an indication that the Freundlich isotherm was not obeyed, nevertheless n is less than 10 which is an indication that the adsorption was favourable, the K was also low which is an indication of adsorption capacity of the adsorbent is low, but it is quite promising.

The Freundlich isotherm for adsorption of lead on 500 and 150 micron sizes of rice husk, are as tabulated in table 4 below.

Table 4 : Showing Freundlich isotherm of Lead on rice husk

Rice Husk Size	R^2	K	n
500micron size	0.05	13.30	24.51
150micron size	0.002	21.4	−0.005

It could be observed from the table above, that the regression coefficient was very low (<0.5) which is an indication that Freundlich isotherm was not obeyed, however, n was high an indication that the adsorption on the adsorbate was favourable; K was also high which is an indication that adsorption capacity was high.

The Temkin adsorption isotherm of the adsorption of zinc on rice husk of particle sizes 150 and 500 micron sizes were examined. The Temkin model is represented by the equation.

$$q_e = \left(\frac{RT}{b}\right) lnA + \left(\frac{RT}{b}\right) lnCe$$

where

q_e = equilibrium mass adsorbed per unit mass

R = gas constant = 8.314 J $K^{-1}mol^{-1}$

T = Temperature in Kelvin = 303K

Ce = equilibrium concentration in mg/L

B = Temkin isotherm constant (J mol^{-1})

A= Equilibrium binding constant corresponding to the maximum binding energy (L/mg).

The plot of lnCe against qe gives a linear graph whose slope is $\left(\frac{RT}{b}\right)$ and intercept is $\left(\frac{RT}{b}\right) \ln A$ which can be used to obtain A [19].

The Temkin isotherm of zinc on 500 and 150 micron sizes of rice husk is as tabulated in table 5 below.

Table 5 : Showing Temkin isotherm of zinc on rice husk.

Rice Husk Size	R^2	A	b
500micron size	0.480	44.37	1247
150micron size	0.446	0.798	700

It could be observed from the table 5 above that the regression coefficients were low, an indication that the Temkin isotherm was not obeyed; however the isotherm constant in both cases were high.

The Temkin isotherm of lead on 500 and 150 micron sizes of rice husk is as tabulated in table 6 below.

Table 6 : Showing Temkin adsorption isotherm of lead on rice husk

Rice Husk Size	R^2	A	b
500micron size	0.058	1.5E7	3232
150micron size	0.003	4.3E62	16,465

It could be observed from the table 6 above that the regression coefficients were low, an indication that the Temkin isotherm was not obeyed; however the isotherm constant in both cases were high, an indication that lead interaction with adsorption sites at that temperature were high.

b) Effect of pH

The effect of varied pH (3, 5, 7, 8 and 9) was also examined on the adsorption of zinc on rice husk of both micron sizes, the initial concentration at various pH was 100 mg/L. It was observed that the zinc adsorption per unit mass on adsorbent was low in acidic pH but increased stepwisely to the neutral pH on both micron sizes, but maintained a crest to the basic pH examined in this research. Which seems to suggest that acidic pH does not favour adsorption of zinc on rice husk, whereas it is more favourable at neutral and basic pH. The neutral pH seems to be the optimum pH for the adsorption. The t-test for paired result was conducted on the mass adsorbed per unit mass of adsorbent of the two particle sizes indicated no significant difference. The effect of varied pH on the adsorption of zinc on rice husk is as shown in Figure 3 below.

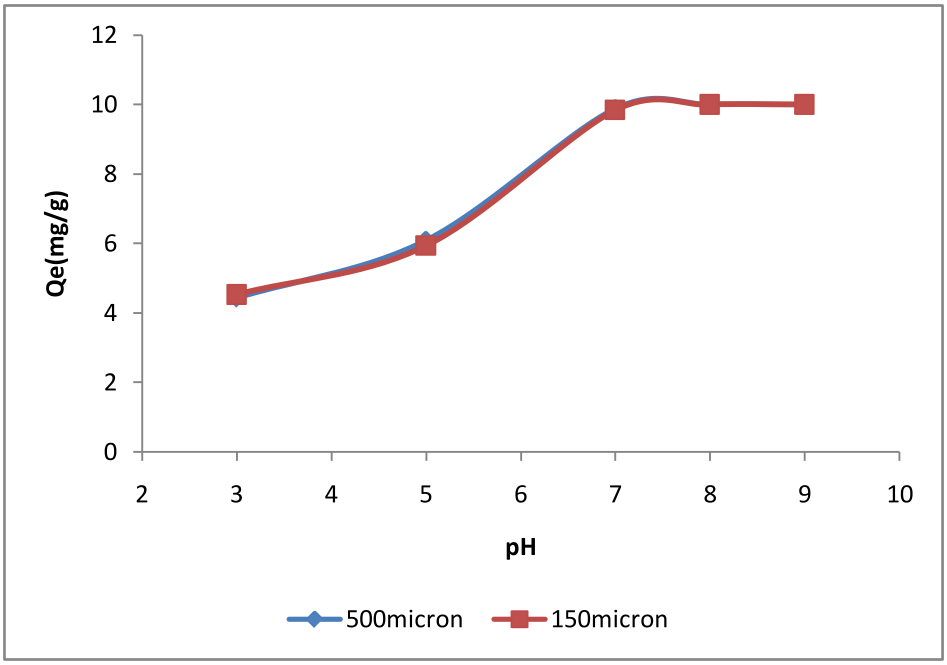

Figure 3 : Showing the effect of varied pH on the adsorption of zinc on rice husk

The effect of varied pH on adsorption of lead was also examined on 500 and 150 micron sizes of rice husk, for pH (3 - 9) and the initial concentration was 100 mg/L for all pH. The effect of changes in pH is not as pronounced as in the case of zinc, as the mass adsorbed per unit mass increased only slightly from acid to neutral pH. However pH 7 seems to be the optimum for 500 micron size of rice husk, while the pH 8 seems to be the optimum for the 150 micron size of rice husk; which seems to suggest that the reduced H^+ ion in neutral and basic pH led to increased adsorption but not significantly. The results of the adsorption of 500 and 150 micron sizes were compared and the paired t-test was conducted and it showed that there was no significant difference between the two results. The effect of varied pH on adsorption of lead on rice husk of varied particle sizes is as shown in Figure 4 below.

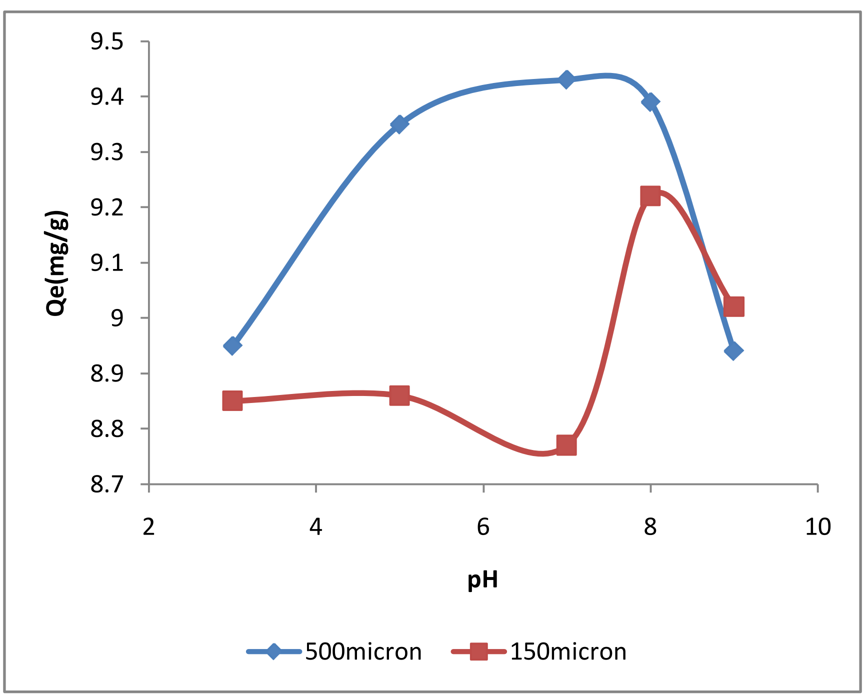

Figure 4 : Showing the effect of varied pH on adsorption of lead on rice husk

c) Effect of time on adsorption

The effect of increase in time was examined on adsorption of lead on rice husk, the effect was examined with 100 mg/L lead solution at 30, 60, 120 and 150 minutes respectively on rice husk of 500 and 150 micron sizes respectively. It was observed that on the 500 micron size of the rice husk, that the mass adsorbed per unit mass increased between 30 and 60 minutes but thereafter decreased at 120 and 150 minutes respectively, this seems to suggest that the optimum time for adsorption of lead on rice husk is the 60 minutes, the mass adsorbed per unit mass was lowest at the 150 minutes, which seems to suggest that longer contact time encouraged desorption of lead on rice husk at the 500 micron size. The trend was different on the 150 micron size of the rice husk, the mass adsorbed per unit mass was about the same for both 30 and 60 minutes of contact, and at 120 minutes the mass adsorbed per unit mass was maximum, however it dropped again at 150 minutes, this seems to suggest that the optimum contact time for the 150 micron size rice husk is 120 minutes, but higher contact time tends to encourage desorption. The graphical representation of effect of increase in contact time on lead adsorption on 500 and 150 micron sizes of rice husk is as shown in figure 5 below.

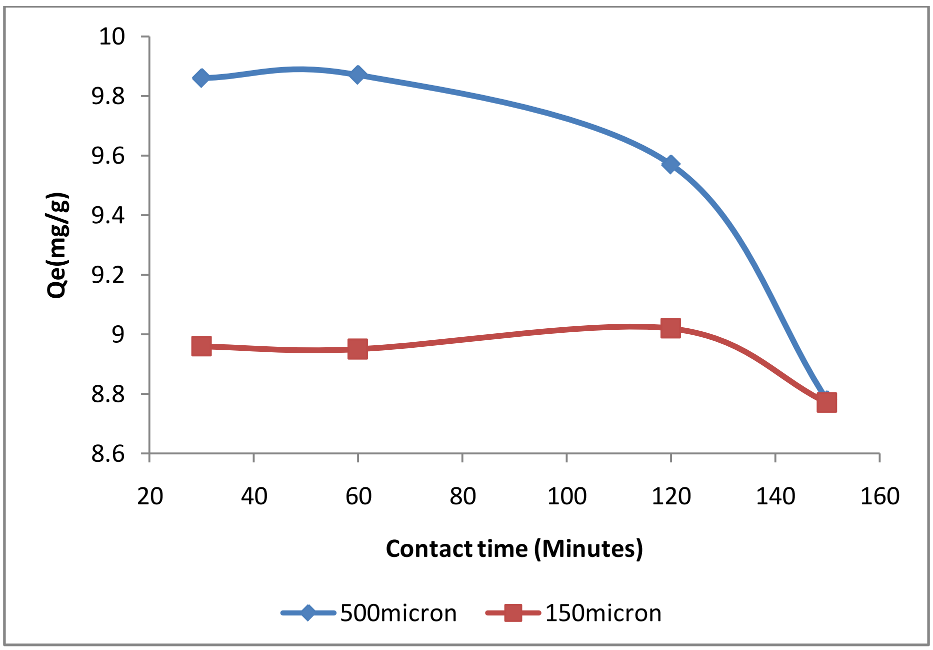

Figure 5 : Showing the effect of varied contact time on adsorption of lead on rice husk of varied particle sizes

Similarly, the effect of increase in time was examined on adsorption of zinc on rice husk of varied particle sizes, the effect was examined with 100 mg/L zinc solution at 30, 60, 120 and 150 minutes respectively on rice husk of 500 and 150 micron sizes respectively. It was observed on the 500 micron size that the mass adsorbed per unit mass increased steadily as the contact time increased from 30 to 120 minutes but dropped at 150 minutes; this seems to suggest that increase in contact time increased adsorption between 30 and 120 minutes. 120 minutes seems to be the optimum contact time but a further increase in time beyond the 120 minutes encouraged desorption of zinc at the 500 micron size of rice husk. But the trend was different on the 150 micron size rice husk as mass adsorbed per unit mass decreased steadily from the 30 to 120 minutes respectively but increased sharply at the 150 minutes contact time, 150 minutes was the optimum for the 150 micron size of rice husk; this seems to suggest that increased contact time does not favour adsorption on the 150 micron size of rice husk between 30 and 120 minutes but was optimum at the 150 minutes. The graphical representation of the effect of contact time on adsorption of zinc on rice husk of varied particle sizes is as shown in figure 6 below.

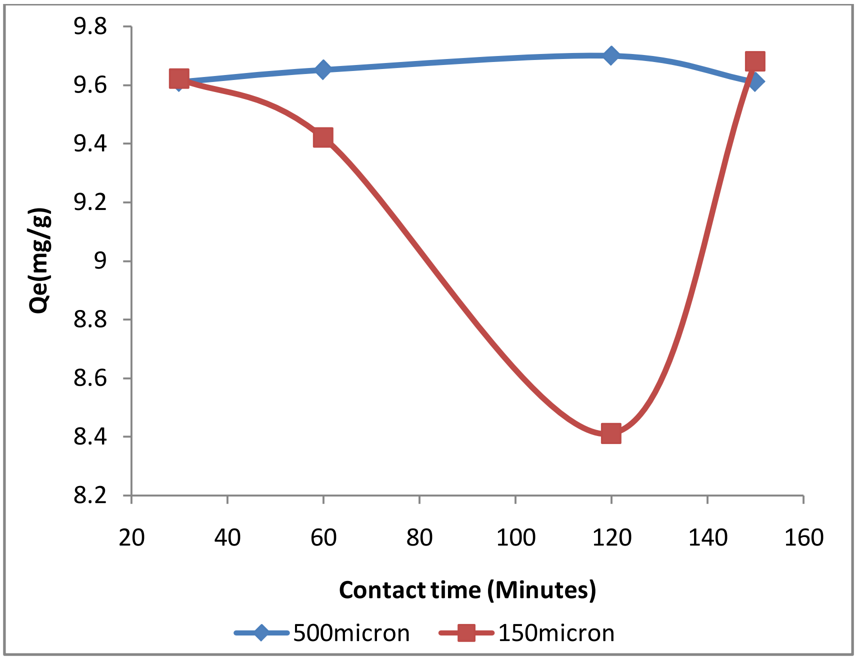

Figure 6 : Showing the effect of varied contact time on adsorption of zinc on rice husk of varied particle sizes

The data of lead adsorption in varied contact time were examined using the Lagergren first order and Ho's second order kinetics; this is an indication of the molecularity of the sorption mechanism and the rate controlling step. The Lagergren model is a pseudo first order kinetic and is guarded by the equation

$$\ln(Co – Ct) = Kt + A$$

where Co = Initial concentration of the adsorbate solution.

Ct = Concentration of the adsorbate at time t

t = time in minute

K = Sorption rate constant.

A = intercept. [9].

And the Ho's is a pseudo second order kinetic and is guarded by the equation

$$\frac{1}{qe} = Kt + A$$

Where qe = the amount adsorbed by the adsorbent per mass of the adsorbent at equilibrium,

K = Sorption rate constant

A = intercept [9].

The result of Lagergren kinetic of lead on rice husk of 500 and 150 micron sizes is as shown in table 7 below.

Table 7 : Showing Largergren kinetic of lead on rice husk

Rice Husk Size	R^2	K	A
500micron size	0.772	8.0×10^{-4}	4.63
150micron size	0.417	2.0×10^{-4}	4.51

From the table above, we observe that the 500 micron size of rice husk obeyed the Lagergren kinetics, as the correlation coefficient was high (0.772) and the rate constant was very low and the intercept is high. But for 150 micron size was quite different as the correlation coefficient was very low (0.417) an indication that the Lagergren model was not obeyed and the rate constant was very low but the intercept is high.

The result of Ho's kinetic of lead on rice husk of 500 and 150 micron sizes is as shown in table 8 below.

Table 8 : Showing Ho's kinetic of lead on rice husk

Rice Husk Size	R^2	K	A
500micron size	0.798	$1.0 \mathrm{x} 10^{-4}$	$9.62 \mathrm{x} 10^{-2}$
150micron size	0.190	$1.0 \mathrm{x} 10^{-5}$	$1.11 \mathrm{x} 10^{-1}$

From the table above, we observe that the 500 micron size of rice husk obeyed the Ho's kinetics, as The correlation coefficient was high (0.798) and the rate constant was very low and the intercept is very low as well. But for 150 micron size was quite different as the correlation coefficient was very low (0.190), an indication that the Ho's model was not obeyed and the rate constant was very low but the intercept is equally very low.

In the same vein the data of zinc adsorption at varied time was examined on Lagergren pseudo first order model and the result of the model is as tabulated in the table 9 below.

Table 9 : Showing Lagergren kinetic of zinc on rice husk

Rice Husk Size	R^2	K	A
500micron size	0.0364	2.0×10^{-5}	4.567
150micron size	0.4248	6.3×10^{-3}	4.209

We observe from the table above that the Lagergren model was not obeyed on both particle sizes, as the correlation coefficients were very low, and the rate constant in both cases too were very low, but the intercept in both cases were high.

The Ho's model of the data of zinc adsorption on rice husk of varying particle sizes is shown in the table 10 below.

Table 10 : Showing Ho's kinetic of zinc on rice husk

Rice Husk Size	R^2	K	A
500micron size	0.0364	2.0×10^{-6}	1.04×10^{-1}
150micron size	0.0798	4.0×10^{-5}	1.05×10^{-1}

We observe from the table above that the Ho's model of kinetic was not obeyed, as the correlation coefficients were very low and the rate constant were very low as well, the intercept of in both cases were also very low.

d) Effect of temperature on adsorption

The effect of increase in temperature was examined in lead adsorption on rice husk of 500 and 150 micron sizes at 100mg/L of lead solution between 20°C - 80°C; it was observed that for both micron sizes the adsorption per unit mass were higher at lower temperature as compared to higher temperature; which seems to suggest that lower temperature favours adsorption on rice husk, hence the adsorption seems exothermic. The paired t-test of the two data obtained for the two particle sizes showed that there was no significant difference between the two results. The graphical representation of the adsorption of lead on rice husk of 500 and 150 micron sizes are as shown in the figure 7 below.

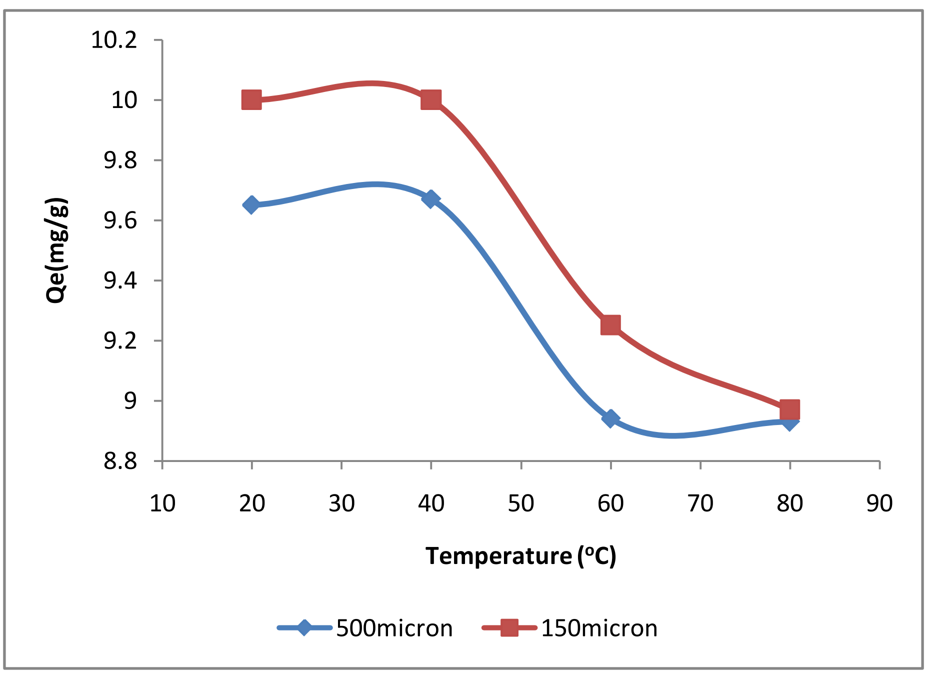

Figure 7 : Showing the effect of varied temperature on adsorption of lead on rice husk of varied particle sizes

The effect of increase in temperature was examined in zinc adsorption on rice husk of 500 and 150 micron sizes at 100 mg/L of lead solution between 40 – 80°C; it was observed that both sizes seems to favour adsorption of lead on rice husk at higher temperature and was optimum at 60°C. the adsorption per unit mass was higher at 60°C, which seems to suggest that the adsorption of lead on rice husk is endothermic. The paired t-test of the two data obtained for the two particle sizes showed that there was no significant difference between the two results. The graphical representation of the effect of temperature on adsorption of lead on rice husk of 500 and 150 micron sizes are as shown in the figure 8 below.

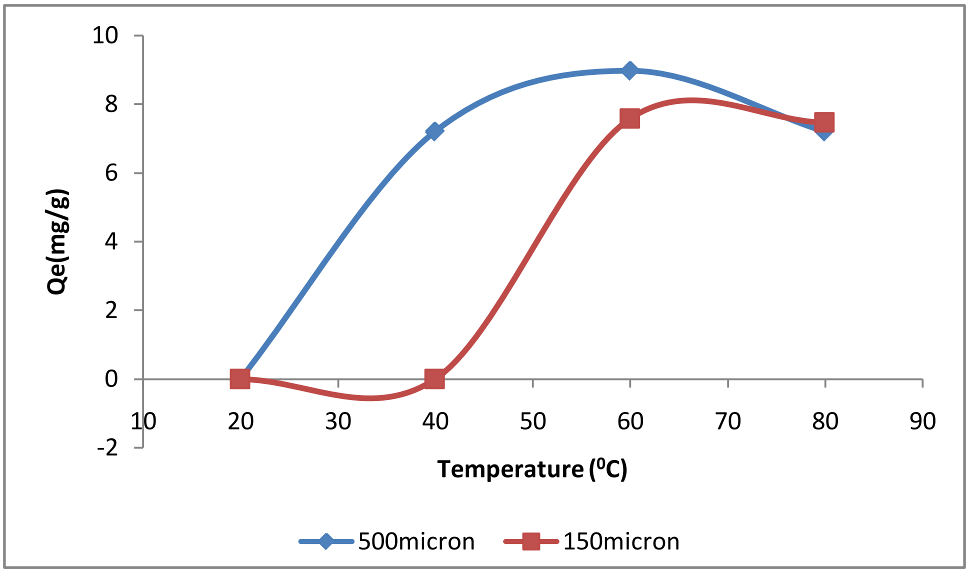

Figure 8 : Showing the effect of varied temperature on adsorption of Zinc on rice husk of varied particle sizes

IV. Conclusion

In general, the result showed that rice husk is a good biosorbent for lead and zinc in waste water remediation, with a very high mass adsorbed per unit mass under atmospheric conditions, albeit rice husk showed greater affinity for the removal of lead than zinc in all the conditions examined and the adsorption seems to be exothermic rather than endothermic as the adsorption was favoured by lower temperature. Neutral and basic pH favoured the adsorption in most of the cases as compared to the acidic pH. Increase in contact time did not have any significant difference in the level of adsorption for all the contact time considered. The smaller particle sizes seems not to obey the Lagergren pseudo first order kinetic nor the Ho's pseudo second order kinetic while the larger particle size tends to obey the Lagergren pseudo first order and Ho's pseudo second order kinetic. Relatively Langmuir and Largergren isotherm were obeyed by both particle sizes at varied concentration and contact time respectively. It is noteworthy to state that rice husk is a good adsorbent for lead and zinc in waste water with good efficiency even at small quantity.

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