Comparative Studies of the Biosorption of Heavy Metals (Zinc and Lead) using Tea Leaves (Camellia Sinensis) and Tea Fibre as Adsorbents

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Abstract: This research has helped us to ascend from the initial two exploratory studies to a more advanced general conclusion and theory that tea leaves (Camellia sinensis) has a great potential for Pb(ii) and Zn uptake, as such, a more suitable Low-cost adsorbent for the effective removal of Pb (II) and Zn from industrial effluents waste water than tea fibre. This is coined from the results of the different biosorption studies carried out as a function of contact time, initial metal ion concentration, biosorbent dosage and Pseudo first and second order models separately carried out on tea leaves and tea fibres

Keywords: biosorption; spectroscopy; physisorption; chemisorption; adsorption; dosage; kinetics; Camellia sinensis.

1. INTRODUCTION

Heavy metals as described by Barrera et al., 2006 are elements whose density is equal to or greater than 6.0 g/cm³ e.g Lead (Pb), mercury (Hg), cadmium (Cd) and arsenic (As). They have for long been used by man in building materials, as medicine, as pigments or as additives for petrol (Hylander and Meili, 2003), (Järup, 2003). Researchers later proved that heavy metals generally poses a great deal of problems to mankind due to its presence in the environment at concentrations above threshold (Seker et al., 2008). Anthropogenic activities such as industrial effluents, mining, smelting, manufacture of explosives, metal plating, domestic effluents, leaching and run-offs from garbage are responsible for the risen cases of these toxic elements in the environment (Trueby, 2003). And the danger associated to the presence of these heavy metals have been attributed to bioconcentration and bioaccumulation in the food web in different locations (Nussey, 1998) enhanced by their ability to be transported to a distance usually by water (Bradl, 2005). Heavy metal pollution of the environment is now one of the most serious environmental problems worldwide which calls for ways of remediation because they are acutely and exceedingly toxic, indestructible (non-biodegradable) and they bioaccumulates thereby causing different health complications e.g. lead poisoning damages the kidney, liver, brain, reproductive systems and even the nervous systems (Naiya et al., 2009). Also, excess ingestion of zinc can lead to microcytosis, impaired immune response, neutropenia and hypocupremia, (Appelo and Postma, 2007).

Thus, the removal of heavy metals including lead and zinc is therefore justified; this study evaluates the performance of Camellia sinensisbiomass as an adsorbent in the removal of lead and zinc from a contaminated aqueous phase or solution. The paper presents a momentous topic ‘comparative studies of the biosorption of heavy metals (zinc and lead) using tea leaves (cammelia sinensis) and tea fibre as adsorbents’

The work is devised to enhance the use of natural available adsorbents such as Camellia sinensisbiomass instead of chemicals might give advantages such as lesser cost of production, source of income to the producer, source of revenue to the government as taxes from the sells, less sludge production and readily available materials and enhances sustainable development of agro-based waste.
2. MATERIALS AND METHODS

2.1. Materials

Apparatus/reagents required are; volumetric flasks, Analytical balance, beakers, conical flasks, pipette, Mortar bland and pestle, centrifuge tubes, Filter papers, Centrifuge, pH meter, Atomic adsorption spectrometer. Lead (II) nitrate salt [Pb(NO3)2], Nitric acid (HNO3) or Hydrochloric acid (HCl) sodium hydroxide salt (NaOH), distilled water.

2.2. Sample Collection

Camellia sinensis (tea leaves and tea fiber), used for this analysis were obtained from Kakara High Land Tea, Sardauna L.G.A. Taraba state, Nigeria on JULY 2018.

2.3. Methods

2.3.1. Preparation of Adsorbent

The tea leaves (Camellia sinensis) obtained were sundried for one week (7 days) after which it was pulverized and sieved using a 150mm sieve size. This sample was stored in an airtight polytene bag until analysis.

2.3.2. Preparation of Stock Solution

0.1M of lead (II) nitrate Pb(NO3)2 (R & M marketing Essex U.K. with MW=331.20g/mol). It was prepared for use throughout the experimental work. 33.12 g of lead (II) nitrate powder was dissolved in 500 mL of stock solution.

0.1M of Zn (Mw=287.38g/mol). It was prepared for use throughout the experimental work. 28.738g of zinc sulphide powder was dissolved in 500mL of stock solution. (Etim et al., 2019)

2.3.3. Preparation of different Concentrations of Metal Solutions

In this study, a total of 5 different concentrations of Pb2+ and Zn solutions were prepared: 0.02M, 0.04M, 0.06M, 0.08M and 0.1M. Subsequently, the lead and Zn solutions with different concentrations and biosorbent materials were required to be put into the orbital shaker (SSL1; Stuart®) at different temperatures (between 30ºc – 60ºc). The rotational speed of shaker, in all the experiments, was kept constant at 220 rpm. This experiment was performed in duplicate and the best results were used. Lastly, the solution was filtered to prepare samples for the measurements of the metal ion concentration (Etim et al., 2019) and (Asuquo et al., 2019a,b,c)

2.4. Effect of Initial Concentration

50mL of each metal solution, containing different concentrations ;0.02 M, 0.04 M, 0.06 M, and 0.08 M were measured into different conical flasks. 5 g of the biosorbent was dispersed in each of them, the flasks were corked and the mixture agitated with the aid of a shaker for 1 hour to attain equilibrium, the slurries were then filtered using Whatman filter paper and a plastic funnel, the filtrate was kept in well labelled containers and thereafter the concentrations of the resulting filtrate was determined using Atomic absorption spectrometer. (Reddad et al., 2002) and (Entezari et al., 2009).

2.5. Effect of Biosorbent Dosage

2 g, 4 g, 6 g, and 8 g of the adsorbent were weigh into different conical flasks. 50 ml of each metal solution were measured into each of the conical flasks and labelled. The flasks were corked and the mixture agitated with the aid of a shaker for 1 hour to attain equilibrium, the slurries were then filtered using Whatman filter paper and a plastic funnel, the filtrate was kept in well labelled containers and thereafter the concentrations of the resulting filtrate was determined using Atomic absorption spectrometer. (Reddad et al., 2002) and (Entezari et al., 2009).

2.6. Effect of Time (Time dependence)

5g of biosorbent were suspended into different conical flasks containing 50mL of metal solution. Each beaker was agitated on an electrical shaker/rotatory mixer at 30rpm with the time difference between each beaker were 10 min, 20 min, 30 min and 40 min. Maintaining these parameters; temperature of 25oc, pH of 6, concentration of metal 0.1M Constant. Once the spinning is complete, the solute is extracted and placed into plastic centrifuge tubes after which it was centrifuged for 3min at 6000rpm. This enabled the separation of the biosorbent from the solution.
Finally, the solution is extracted from the centrifuge tube using a dropper and it is placed in clean airtight bottles prior to analysis using atomic adsorption spectrometer (AAS) (Reddad et al., 2002), (Entezari et al., 2009) and (Etim et al., 2019).

### 2.7. Estimation of Metal Uptake

The metal uptake, \( q_e \), was determined using the following equation (Madhavi et al., 2011):

\[
q_t = \frac{(C_0 - C_f)V}{m}
\]

Where

- \( q_e \) = metal ions per dry biosorbent (mg/g)
- \( V \) = volume of solution (L)
- \( C_0 \) = initial concentration of metal in solution (mg/L)
- \( C_f \) = final concentration of metal in solution (mg/L)
- \( m \) = the mass of biosorbent (g)

### 3. RESULTS AND DISCUSSION

#### 3.1. Comparing the Effect of Adsorbent Dosage (Between Tea Leaves and Tea Fiber) for the Removal of Pb and Zn from Aqueous Phase

As shown in table 1 and 2, similar dosage measurement was applied in both the assessment, the percentage biosorption was high ranging from 98.81% - 99.95 % sorption, with a constant increase in the dosages. There was a slight decrease in % biosorption of Pb in tea fibre, this confirms (Beatti et al., 2007) statement in their work on heavy metals that it might be as a result of the aggregation of much adsorbents and adsorbates ions. These findings from the two research narrows our understanding to an assumption that an increase in the amount of adsorbent dosage leads to a gradual increase in the amount or percentage biosorption both in tea leaves and tea fibers.

**Table 1. Adsorbent dose data for removal of Lead and Zinc from aqueous phase using Camellia Sinensis (tea leaves)**

| Initial Conc (Mg/L) | Final conc (Mg/L) | % Biosorption | Metal uptake (mg/L) | Initial Conc (Mg/L) | Final conc (Mg/L) | % Biosorption | Metal uptake (mg/L) |
|---------------------|------------------|---------------|---------------------|---------------------|------------------|---------------|---------------------|
| 500                 | 177.8            | 99.11         | 513.0               | 500                 | 1018.7          | 99.90         | 518.0               |
| 1000                | 180.5            | 99.13         | 684.0               | 1000                | 1020.3          | 99.94         | 690.0               |
| 1500                | 183.9            | 99.14         | 102.6               | 1500                | 1026.1          | 99.94         | 103.5               |
| 2000                | 187.3            | 99.09         | 205.4               | 2000                | 1069.7          | 99.95         | 207.0               |

**Table 2. Adsorbent dose data for removal of Lead and Zinc from aqueous phase using Camellia Sinensis (Tea fibre)**

| Adsorbent dosage (g) | Final conc (Mg/L) | % Biosorption | Metal uptake (mg/L) | Adsorbent dosage (g) | Final conc (Mg/L) | % Biosorption | Metal uptake (mg/L) |
|----------------------|------------------|---------------|---------------------|----------------------|------------------|---------------|---------------------|
| 500                  | 240.3392         | 98.88         | 2047.9608           | 500                  | 10.456          | 99.84         | 326.3772            |
| 1000                 | 238.020          | 98.85         | 1024.0090           | 1000                 | 7.802           | 99.88         | 326.5099            |
| 1500                 | 247.442          | 98.88         | 682.4186            | 1500                 | 7.545           | 99.89         | 326.5228            |
| 2000                 | 242.151          | 98.88         | 511.9462            | 2000                 | 7.602           | 99.88         | 326.5199            |

**Figure 1.** Adsorbent dose chart comparing the removal of Lead and Zinc from aqueous phase using Camellia Sinensis Tea leaves and Tea fibre.
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Also, more elucidation carried out as shown in figure 1 below revealed that tea leaves are better adsorbents than tea fibre with an increasing dosage especially in the removal of Zinc, when all other factors are kept constant. This finding supports (Etim et al., 2019) work on tea leaves

3.2. Comparing the Effect of Initial Ion Concentration on Tea Leaves and Tea Fibre

Table 3. Effect of Initial concentration data for the removal of lead and zinc from aqueous phase using Camellia Sinensis (tea leaves)

| Initial Conc. | Final conc. Mg/L | % Biosorption | Metal uptake | Initial Conc. | Final conc. Mg/L | % Biosorption | Metal uptake |
|---------------|------------------|---------------|--------------|---------------|------------------|---------------|--------------|
| 13076         | 144.8            | 98.89         | 646          | 13076         | 958.1            | 99.93         | 653.0        |
| 26152         | 169.5            | 99.35         | 1299         | 26152         | 996.2            | 99.94         | 130.7        |
| 39228         | 179.1            | 99.54         | 1952         | 39228         | 1026.4           | 99.95         | 196.0        |
| 52304         | 183.9            | 99.64         | 2606         | 52304         | 1069.7           | 99.95         | 261.5        |

Table 4. Effect of Initial concentration data for the removal of lead and zinc from aqueous phase using Camellia Sinensis (tea fibre)

| Initial Conc. | Final conc. Mg/L | % Biosorption | Metal uptake | Initial Conc. | Final conc. Mg/L | % Biosorption | Metal uptake |
|---------------|------------------|---------------|--------------|---------------|------------------|---------------|--------------|
| 4144          | 200.6            | 95.60         | 197.1694     | 13076         | 6.887            | 99.47         | 650.357      |
| 8288          | 237.0            | 97.14         | 402.5488     | 26152         | 7.224            | 99.72         | 130.3988     |
| 12432         | 231.1            | 98.14         | 610.0418     | 39228         | 7.487            | 99.80         | 195.748      |
| 16576         | 245.6            | 98.52         | 816.3159     | 52304         | 7.754            | 99.85         | 261.323      |

Figure 2. Comparing the effect of Initial ion concentration on tea leaves and tea fibre.

Considering the initial ion concentration of adsorbent data obtained from the analysis of the ability of tea leaves and tea fibers to remove Pb and Zn from an aqueous media with initial concentration of the ions known are presented in table 3 and table 4 respectively, as incorporated in figure 2 for a better understanding, the role of tea leaves in the removal of Zn was highest with a high % biosorption (up to 99.95%) followed bytea fibre.

More so, tea leaves functioned more in the removal or % biosorption of Pb than tea fibres at the same initial concentrations of the metal ion, but it can be seen that presence of more Pb ions or increase in the initial concentration does not affect the uptake of Zn ions, rather more Pb ions were left in the solution with tea leaves adsorbent. Therefore, the effect of initial ion concentration Pb and Zn places preference on tea fiber over tea leaves since it showed a higher % biosorption for the heavy metals under study.
3.3. Comparing the Effect of Contact Time (With Tea Leaves and Tea Fiber) for The Removal of Pb and Zn from Aqueous Phase

Table 5. Contact time for removal of Lead and Zinc from aqueous phase using Camellia Sinensis (tea leaves)

| Time(min) | Final conc (Mg/L) | % Biosorption | Metal uptake | Time(min) | Final conc. (Mg/L) | % Biosorption | Metal uptake |
|-----------|-------------------|---------------|--------------|-----------|-------------------|---------------|--------------|
| 10        | 175.8             | 99.15         | 102.7        | 10        | 716.1             | 99.96         | 103.6        |
| 20        | 164.9             | 99.11         | 262.6        | 20        | 618.3             | 99.97         | 202.0        |
| 30        | 159.4             | 99.18         | 302.8        | 30        | 318.3             | 99.98         | 249.2        |
| 40        | 140.3             | 99.20         | 502.7        | 40        | 119.6             | 99.99         | 480.9        |

Table 6. Contact time for removal of Lead and Zinc from aqueous phase using Camellia Sinensis (tea fibre)

| Time(min) | Final conc (Mg/L) | % Biosorption | Metal uptake | Time(min) | Final conc. (Mg/L) | % Biosorption | Metal uptake |
|-----------|-------------------|---------------|--------------|-----------|-------------------|---------------|--------------|
| 10        | 35.347            | 99.83         | 1634.2326    | 10        | 6.2480            | 99.90         | 326.5876     |
| 20        | 10.637            | 99.95         | 1634.2326    | 20        | 10.247            | 99.84         | 326.3877     |
| 30        | 3.012             | 99.98         | 1035.8494    | 30        | 10.464            | 99.84         | 326.3768     |
| 40        | 2.738             | 99.99         | 1035.8631    | 40        | 10.772            | 99.84         | 326.3614     |

From the observation and then in general, we can resolve that, For a prolonged exposure, the % biosorption of Pb and Zn increases using both tea leaves and tea fibre. In a prolonged exposure, tea leaf is more efficient in the removal of Zn than Pb, as shown from the data obtained during the analysis and presented in table 5, while table 6 shows that at a longer time, the % biosorption of Pb increases gradually. This finding is also in line with (Hanif and Akhtar, 2007) who stated that adsorption increases initially at increase in contact time because initially, all the binding sites are available and so the adsorbate ion easily becomes bonded to the sites. It’s shown in figure 3 below. Comparatively, the performance of tea leaves in the biosorption is higher than that of tea fibre over time.

3.4. Comparing the Kinetics of Biosorption of Lead and Zinc Using Tea Leaves and Tea Fiber

3.4.1. Lagergren Pseudo First-Order

The lagergren Pseudo first-order reaction was used to describe the kinetics with the linear equation form shown below

\[
\log (q_e - q_t) = \log q_e - \frac{K_1 t}{2.303}
\]

Where \( q_e \) (mg/g), \( q_t \) (mg/g) are adsorption capacity at equilibrium and at time \( t \), respectively. \( K_1 \) is the rate constant of pseudo first-order adsorption (L/min). The value of the constants \( q_e, K_1 \) and \( R^2 \) obtained from the linear plot of \( \log (q_e-q_t) \) vs \( t \)
Table 7. First order kinetic of biosorption of Lead and Zinc using tea leaves

| Time(min) | Final conc Mg/L | % Biosorption | Metal uptake | Log (qt-qe) | Time(min) | Final conc Mg/L | % Biosorption | Metal uptake | Log (qt-qe) |
|-----------|-----------------|---------------|--------------|-------------|-----------|-----------------|---------------|--------------|-------------|
| 10        | 175.8           | 99.15         | 102.7        | 0.55022     | 10        | 716.1          | 99.96         | 103.6        | 0.5599      |
| 20        | 164.9           | 99.11         | 262.6        | 2.2134      | 20        | 618.3          | 99.97         | 202.0        | 2.0087      |
| 30        | 159.4           | 99.18         | 302.8        | 2.3088      | 30        | 318.3          | 99.98         | 249.2        | 2.1738      |
| 40        | 140.3           | 99.20         | 502.7        | 2.6058      | 40        | 119.6          | 99.99         | 480.9        | 2.5808      |

Table 8. First order kinetic of biosorption of Lead and Zinc using tea fibre

| Time(min) | Final conc Mg/L | % Biosorption | Metal uptake | Log (qt-qe) | Time(min) | Final conc Mg/L | % Biosorption | Metal uptake | Log (qt-qe) |
|-----------|-----------------|---------------|--------------|-------------|-----------|-----------------|---------------|--------------|-------------|
| 10        | 35.347          | 99.83         | 1634.2326    | 3.1859      | 10        | 6.2480         | 99.90         | 326.5876     | 2.3554      |
| 20        | 10.637          | 99.95         | 1634.2326    | 3.1859      | 20        | 10.247         | 99.84         | 326.3877     | 2.3551      |
| 30        | 3.012           | 99.98         | 1035.8494    | 2.9712      | 30        | 10.464         | 99.84         | 326.3768     | 2.3551      |
| 40        | 2.738           | 99.99         | 1035.8631    | 2.9712      | 40        | 10.772         | 99.99         | 326.3614     | 2.3551      |

The data used for the Pseudo first-order kinetics are contained in table 7 and 8 above, and the linear plot is shown in figure 4 below comparing tea leaves and tea fibers on the adsorption of Pb and Zn. The result of the Pseudo first-order kinetics best fitted adsorption of Zn on tea leaves with regression coefficient R² = 0.833, followed by the adsorption of Pb in tea fibre with a regression coefficient of R² = 0.8 and moderately fitted the adsorption of Pb in tea leaves with a regression coefficient of R² = 0.758 and Zn in tea leaves with R² = 0.6. This implies that the reaction involving tea fiber favors (more inclined) towards physisorption than tea leaves since it values are close to 1.

Figure 4. First order kinetic comparing the biosorption of Pb and Zn using tea leaves and tea fibre.

3.4.2. Pseudo Second-Order Model

Table 9. Second order kinetic of biosorption of Lead and Zinc using tea leaves

| Time(min) | Final conc Mg/L | % Biosorption | Metal uptake | t/qt | Time(min) | Final conc Mg/L | % Biosorption | Metal uptake | t/qt |
|-----------|-----------------|---------------|--------------|------|-----------|-----------------|---------------|--------------|------|
| 10        | 175.8           | 99.15         | 102.7        | 0.0973| 10        | 716.1          | 99.96         | 103.6        | 0.0965|
| 20        | 164.9           | 99.11         | 262.6        | 0.0761| 20        | 618.3          | 99.97         | 202.0        | 0.0990|
| 30        | 159.4           | 99.18         | 302.8        | 0.0990| 30        | 318.3          | 99.98         | 249.2        | 0.1203|
| 40        | 140.3           | 99.20         | 502.7        | 0.0795| 40        | 119.6          | 99.99         | 480.9        | 0.0831|
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Table 10. Second order kinetic of biosorption of Lead and Zinc using tea fibre

| Time(min) | Final conc Mg/L | % Biosorption | Metal uptake qt | t/qt | Final conc Mg/L | % Biosorption | Metal uptake qt | t/qt |
|-----------|-----------------|----------------|-----------------|-------|-----------------|----------------|-----------------|-------|
| 10        | 35.347          | 99.83          | 1634.2326       | 0.0061| 10              | 6.248          | 99.90           | 326.5876 | 0.0306 |
| 20        | 10.637          | 99.95          | 1634.2326       | 0.0122| 20              | 10.247         | 99.84           | 326.3877 | 0.0612 |
| 30        | 3.012           | 99.98          | 1035.8494       | 0.0289| 30              | 10.464         | 99.84           | 326.3768 | 0.0919 |
| 40        | 0.738           | 99.99          | 1035.8631       | 0.0386| 40              | 10.772         | 99.84           | 326.3614 | 0.1225 |

The Pseudo second order equation is shown below and the model is based on the assumption that the rate determining step is chemisorptions (Das and mondal, 2011).

\[
\frac{t}{q_t} = \frac{1}{K_2q_e^2} + \frac{t}{q_e}(3)
\]

Where \(K_2\) = Rate constant (g/mg/min) of pseudo second- order adsorption rate constant. The values of \(k_2\) and \(q_e\) were obtained from the plots of \(t/q_t\) versus \(t\) of lead and zinc (Table 9 and 10) as shown in figure 5 below. The correlation coefficient of \(R^2 = 1\) and \(R^2 = 0.964\) for the adsorption of Zn and Pb respectively from tea fibre indicates that the reaction is more inclined towards chemisorptions and Pseudo second order kinetics provides the best fit for the adsorption of Pb and Zn in tea fibre than Pb and Zn in tea leaves whose correlation coefficients \(R^2 = 0.025\) and \(0.110\) respectively.

![Figure 5. Second order kinetics](image)

4. CONCLUSION

Comparatively, The finding showed that Tea leaves are very effective and better biosorbent for the clearing and removal of Pb and Zn ions from aqueous phase compared to tea leaves all of Camellia sinensis. This conclusion was derived from the comparative analysis of the percentage biosorption of the two Camellia sinensis in the form of tea leaves and tea fibers that were examined, as a function of adsorbents dosage, initial metal ion concentration and contact time. kinetics of the biosorption showed that the processes of the adsorption was feasible as the adsorption data fits more into Langergren Pseudo first-order kinetics for Zn and Pb in tea leaves and tea fibres respectively while a good fit of the adsorption data of tea fibre was found only to fit more into Pseudo second-order kinetics for both Pb and Zn. The research therefore presents tea leaves which is a very cheap and low cost by-product of tea processing as an effective biosorbent.

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