International Journal of Advanced Research in Chemical Science (IJARCS)
Volume 6, Issue 9, 2019, PP 20-27
ISSN No. (Online) 2349-0403
DOI: http://dx.doi.org/10.20431/2349-0403.0609003
www.arcjournals.org

Comparative Studies of the Biosorption of Heavy Metals (Zinc and Lead) using Tea Leaves (Cammelia 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; chemisorptions; 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/cm^3 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 metalsgenerally 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 sinensis*biomass 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 sinensis*biomass 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. Prepaaration 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 Pb^{2+} 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, qe, was determined using the following equation (Madhavi et al., 2011):

$$qt = \frac{(Co - Cf)V}{m} \quad (1)$$

Where

q_e = metal ions per dry biosorbent (mg/g)

V = volume of solution (L)

C_o= 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.

Table1. *Adsorbent dose data for removal of Lead and Zinc from aqueous phase using Camellia Sinensis (tea leaves)*

Heavy Metals							
Lead				*Zinc*			
Initial Conc.	*Final conc Mg/L*	*% Biosorption*	*Metal uptake*	*Initial Conc.*	*Final conc. Mg/L*	*% Biosorption*	*Metal uptake*
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

Table2. *Adsorbent dose data for removal of Lead and Zinc from aqueous phase using Camellia Sinensis (Tea fibre)*

Heavy Metals							
Lead				*Zinc*			
Adsorbent dosage(g)	*Final conc Mg/L*	*% Biosorption*	*Metal uptake*	*Adsorbent dosage(g)*	*Final conc. Mg/L*	*% Biosorption*	*Metal uptake*
500	240.392	98.84	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	98.81	682.4186	1500	7.545	99.89	326.5228
2000	242.151	98.83	511.9462	2000	7.602	99.88	326.5199

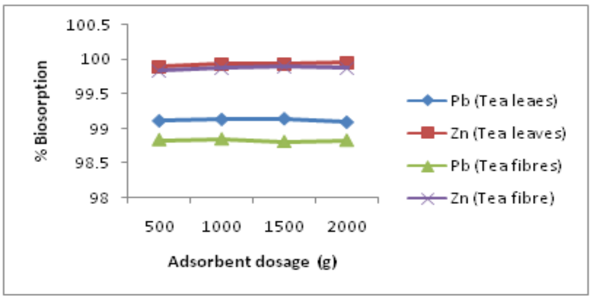

Figure1. *Adsorbent dose chart comparing the removal of Lead and Zinc from aqueous phase using Camellia Sinensis Tea leaves and Tea fibre*

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

Table3. *Effect of Initial concentration data for the removal of lead and zinc from aqueous phase using Camellia Sinensis.(tea leaves)*

Heavy Metals							
Lead				*Zinc*			
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

Table4. *Effect of Initial concentration data for the removal of lead and zinc from aqueous phase using Camellia Sinensis (tea fibre)*

Heavy Metals							
Lead				*Zinc*			
Initial Conc.	*Final conc Mg/L*	*% Biosorption*	*Metal uptake*	*Initial Conc.*	*Final conc. Mg/L*	*% Biosorption*	*Metal uptake*
4144	200.612	95.60	197.1694	1307.6	6.887	99.47	65.0357
8288	237.023	97.14	402.5488	2615.2	7.224	99.72	130.3988
12432	231.163	98.14	610.0418	3922.8	7.487	99.80	195.748
16576	245.681	98.52	816.5159	5230.4	7.754	99.85	261.323

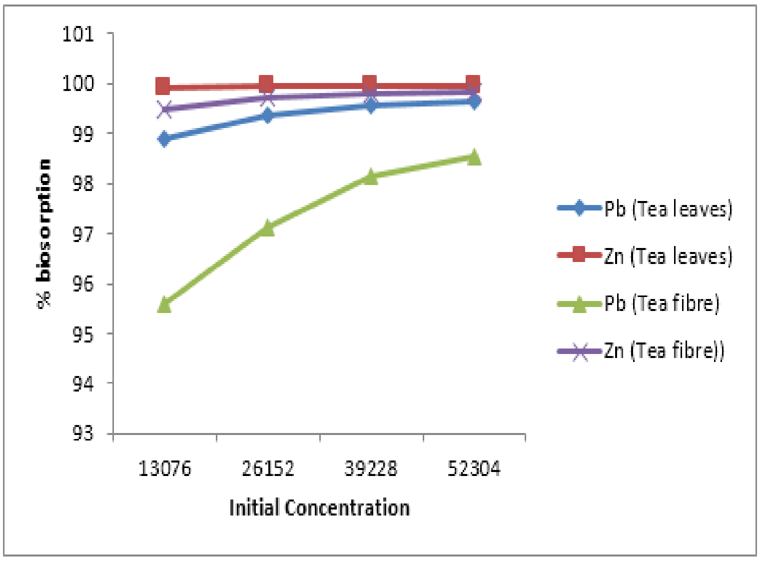

Figure2. *Comparing the effect of Initial ion concentration on tea leaves and tea fibre.*

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 leavesr 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 o of Pb and Zn from Aqueous Phase

Table5. *Contact time for removal of Lead and Zinc from aqueous phase using Camellia Sinensis (tea leaves)*

Heavy Metals							
Lead				*Zinc*			
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

Table6. *Contact time for removal of Lead and Zinc from aqueous phase using Camellia Sinensis (tea fibre)*

Heavy Metals							
Lead				*Zinc*			
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 adsorb ate 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

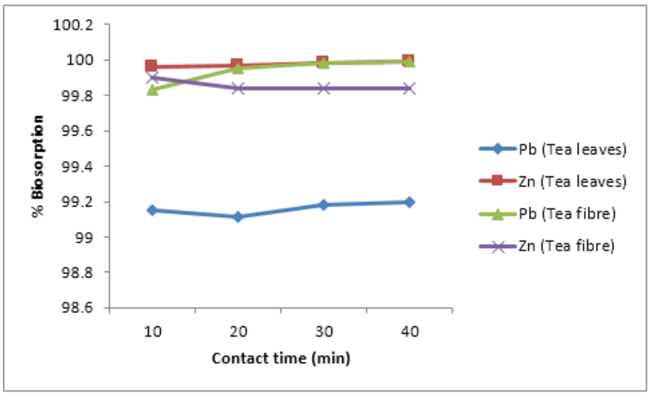

Figure3. *Comparing the effect of contact time (with tea leaves and tea fiber) for the removal of Pb and zn from aqueous phase*

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

$$\text{Log}\ (qe - qt) = logqe - \frac{K1t}{2.303} \quad (2)$$

Where qe (mg/g), qt (mg/g) are adsorption capacity at equilibrium and at time t, respectively. K1 is the rate constant of pseudo first-order adsorption (L/min). The value of the constants qe, k1 and R2 obtained from the linear plot of log (qe-qt) vs t

Table7. *First order kinetic of biosorption of Lead and Zinc using tea leaves*

Heavy Metals									
Lead					Zinc				
Time(min)	Final conc Mg/L	% Biosorption qe	Metal uptake qt	Log (qt-qe)	Time(min)	Final conc Mg/L	% Biosorption qe	Metal uptake qt	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

Table8. *First order kinetic of biosorption of Lead and Zinc using tea fibre*

Heavy Metals									
Lead					Zinc				
Time(min)	Final conc Mg/L	% Biosorption qe	Metal uptake qt	Log (qt-qe)	Time(min)	Final conc Mg/L	% Biosorption qe	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	99.84	326.3614	2.3551

The data used for the Pseudo first-oder 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-oder kinetics best fitted adsorption of Zn on tea leaves with regression coefficient $R^2 = 0.833$, 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 nd 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.

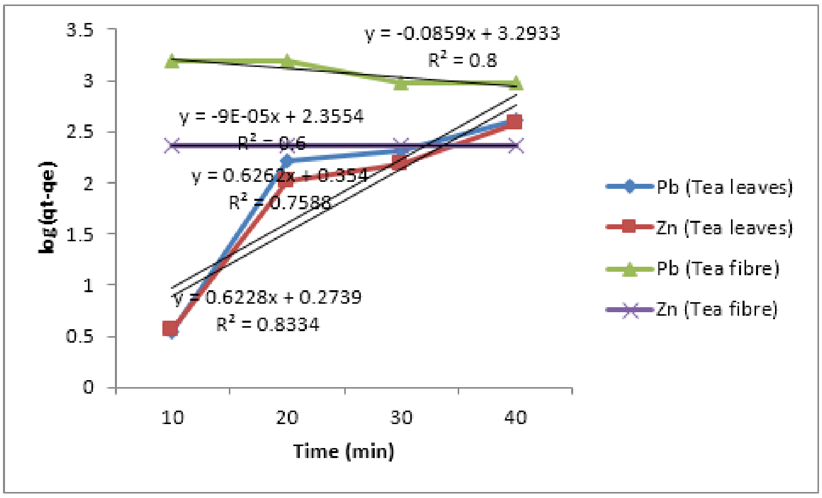

Figure4. *First order kinetic comparing the biosorption of Pb and Zn using tea leaves and tea fibre.*

3.4.2. Pseudo Second-Order Model

Table9. *second order kinetic of biosorption of Lead and Zinc using tea leavs*

Heavy Metals									
Lead					Zinc				
Time(min)	Final conc Mg/L	% Biosorption qe	Metal uptake qt	t/qt	Time(min)	Final conc Mg/L	% Biosorption qe	Metal uptake qt	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

Table10. *second order kinetic of biosorption of Lead and Zinc using tea fibre*

Heavy Metals									
Lead					Zinc				
Time(min)	Final conc Mg/L	% Biosorption qe	Metal uptake qt	t/qt	Time(min)	Final conc Mg/L	% Biosorption qe	Metal uptake	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	2.738	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).

$$\frac{t}{qt} = \frac{1}{K2qe2} + \frac{t}{qe} (3)$$

Where K2 = Rate constant (g/mg/min) of pseudo second- order adsorption rate constant. The values of k2, R2 and qe were obtained from the plots of t/qt versus t of lead and zinc (Table 9 and 10) as shown in figure 5 below. The correlation coefficient of R2 = 1 and R2 = 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 R2 = 0.025 and 0.110 respectively

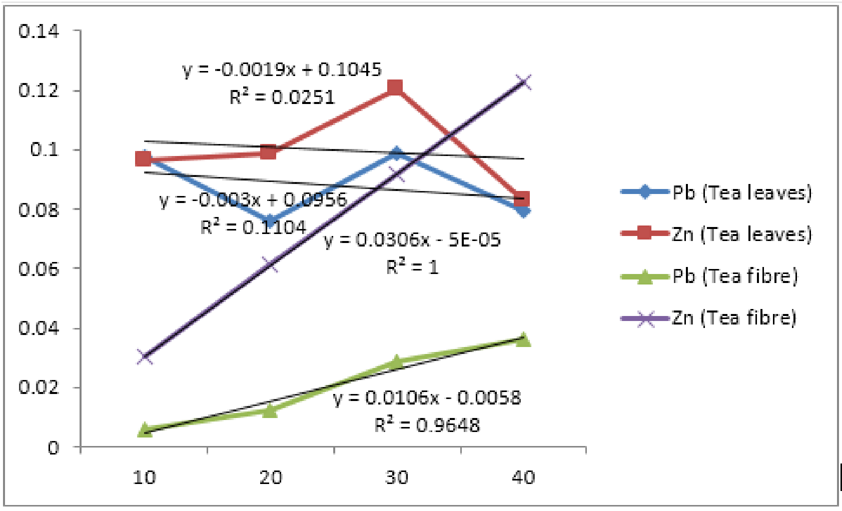

Figure5. *Second order kinetics*

4. Conclusion

Comparatively, The finding showed that Tea leavesare very effective and better biosorbent for the clearing and removal of Pb and Zn ions froman 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 a very cheap and low cost by-product of tea processing as an effective biosorbent.

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Citation: *Emmanuel E. Etim, et.al, "Comparative Studies of the Biosorption of Heavy Metals (Zinc and Lead) using Tea Leaves (Cammelia Sinensis) and Tea Fibre as Adsorbents", International Journal of Advanced Research in Chemical Science, vol. 6, no. 9, p. 20-27, 2019. DOI: http://dx.doi.org/10.20431/2349-0403.0609003*