

Article

## Kinetic Studies of the Biosorption of Zn and Pb (ii) from Solution Using Tea Fibre

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**Abstract:** This paper presents a biobiosorption study on Zn and Pb(II) from dilute aqueous solution using tea fiber as the biosorbent. The biosorption study was carried out as a function of contact time, initial metal ion concentration and biosorbent dosage. The residual Zn and Pb(II) in solution were determined using atomic absorption spectrophotometer. The biosorption was also time dependent as maximum biosorption was obtained after 10 and 20 min, for both lead and zinc respectively. Kinetic studies show that the biosorption of both Zn and Pb(II) can be described by pseudo-second-order kinetics while that of Zn in addition, also fitted more to pseudo first order than that of Pb. For the metal ions, the biosorption efficiency increases with increase in initial metal ion concentration. These results indicate that tea fiber has a high potential for the uptake of Zn and Pb (II) from industrial effluents waste water and therefore, present yet another bright future in the treatment of industrial effluents containing these metals with a merit of the tea fiber been safe, readily available and cost effective.

**Keywords:** Biosorption, spectrophotometry, contact time.

### 1. Introduction

The search for new technology is strongly required as the conventional methods do not provide great solutions towards heavy metal pollution issue. In past decades, biosorption technique has emerged as a useful technique along with other conventional methods for metal removal (Panda, 2008).

In general, biosorption can be defined as the ability of biological materials to accumulate heavy metals from wastewater through metabolically mediated or physico-chemical pathways of uptake (Fourest and Roux, 1992). Biosorption is a new technology for removing metal ions from aqueous solutions for replacing conventional methods such as reduction or oxidation, ion exchange, filtration, electrochemical treatment, membrane technology, evaporation recovery, chemical precipitation, chemical lime coagulation and solvent extraction (Anayur, 2009). These conventional Ways has many disadvantages like extremely expensive, incomplete metal removal and generation of toxic compound. Thus, biosorption is a good method compare with others because it has major advantages include low cost, high efficiency, minimization of chemical and or biological sludge, no additional nutrient requirement, regeneration of biosorbent and possibility of metal recovery (Kratochvil and Volesky, 1998).

A number of studies were reported in literature for the removal of heavy metals from wastewater by biosorption. Major studies may be summarized as follows:

Su *et al.*, (1995) compared the ability of metal removal using an aerobic selector activated sludge system. Heavy metals studied in this research were zinc, cadmium and nickel. Results of experimental data revealed that metal biosorption by activated sludge was rapid and about 70% of the soluble metals in solution was removed during the first 30 min.

Byerley and Scharer (1997) examined Uranium(VI) biosorption from process liquor by *Streptomyces levoris*, *Rhizopus arrhizus*, mixed culture (activated sludge), *Saccharomyces cerevisiae* and *Chlorella vulgaris*. Thermal inactivation had only a marginal effect on the sorption equilibrium.

Yetiset *al.*, (1998) determined heavy metal biosorption potentials of two white rot fungi, *Polyporus versicolor* and *Phanerochaete chrysosporium*. It was found that both *P. versicolor* and *P. chrysosporium* were the most effective in removing Pb(II) ion

Bux *et al.*, (1991) investigated the biosorptive capacity of two waste products of the wastewater treatment industry Activated sludge was found to be more effective than digested sludge for removal of zinc from a metal plating effluent,

Gabrielet *al.* (2001) investigated the biosorption of copper to the pellets of different wood-rotting fungal species. The results of column experiments showed that mycelial pellets of wood-rotting fungi can be used as promising biosorbent material for copper removal (Gabriel *et al.*, 2001).

Diaset *al.*, (2002) investigated the biosorption of chromium, nickel and iron from metallurgical effluents, produced by a steel foundry, using a strain of *Aspergillus terreus* immobilized in polyurethane foam. *A. terreus* UFMG-F01 was immobilized in polyurethane foam and subjected to biosorption tests with metallurgical effluents. Maximal metal uptake values of 164.5 mg g<sup>-1</sup> iron, 96.5 mg g<sup>-1</sup> chromium and 19.6 mg g<sup>-1</sup> nickel were obtained in a culture medium containing 100% of effluent stream

supplemented with 1% of glucose, after 6 d of incubation. In this work, a strain of *A. terreus* was successfully used as a metal biosorbent for the treatment of metallurgical effluents

Ozdemir *et al.*, (2003) investigated the removal of chromium, cadmium and copper ions applying a dead exopolysaccharide producing bacterium, *Ochrobactrum anthropi*, isolated from activated sludge. Optimum pH values of Chromium(VI), cadmium(II) and copper(II) were 2.0, 8.0 and 3.0 respectively. Both the Freundlich and Langmuir adsorption models were suitable for describing the short-term biosorption of Chromium(VI), cadmium(II) and copper(II) by *O. anthropi*.

Galli *et al.*, (2003) determined the optimum conditions for copper (Cu) biosorption by *Auricularia polytricha* mycelium in view of its immobilization in polyvinyl alcohol (PVA). The adsorption of Cu(II) onto *A. polytricha* was studied in batch with respect to initial pH, temperature, adsorption time, initial metal ion and biomass concentration. Ca(II), Mg(II), and Mn(II) were the major ions released from the algal biomass during the sorption which revealed that ion exchange was one of the main sorption mechanisms (Apiratikul and Pavasant 2008).

### 1.1. Biosorbent

Dead and metabolically inactive cells can serve as a basis for the progress to treat and removing heavy metals, nuclear fuel and radioactive elements. They can be highly selective, cheap and efficient compare to activated carbon that used in classical methods. Some types of biosorbents would be broad range, binding and collecting the majority of heavy metals with no specific activity, while others are specific from certain metals. Recent biosorption experiments have focused attention on waste materials, which are byproducts or the waste materials from large-scale industrial operations. For example, the waste mycelia available from fermentation processes, olive mill solid residues (Pagnanelli *et al.*, 2002), activated sludge from sewage treatment plants and biosolids (Pamukoglu and Kargi, 2006) aquatic macrophytes (modak 1995)

### 1.2. Factors Affecting Biosorption

The metal uptake is usually measured by the parameter 'q' which indicates the milligrams of metal accumulated per gram of biosorbent material and 'qH' is reported as a function of metal accumulated, sorbent material used and operating conditions. The following have been reported as factors that affect the biosorption processes:

**Effect of pH** was reported by (Friis and Myers-Keith, 1986). Friis observed that the pH of an adsorbent decreases with increase in acidic groups on the surface of the adsorbent. Metal ions show lesser adsorption at very high and very low pH value (Saeed *et al.*, 2005).

**Temperature** seems to affect biosorption to a lesser extent within the range from 20°C to 35°C (Veglio and Beolchini, 1997). Biosorption removal of most adsorptive pollutants is endothermic, thus higher temperature usually enhances biosorption removal of the adsorbate through increases in its surface activity and kinetic energy (Vijayaraghavan and Yun, 2008). However, higher temperature can also cause physical damage to the biosorbent. Thus, room temperature is usually desirable for the biosorption processes.

**Metal ion concentration:** At very low concentrations of adsorbate, the ratio of available binding sites and adsorbate ions is high, so there are more chances to get adsorbed. Therefore, at low concentrations, adsorption capacity is high. On the other hand, when adsorbate concentration increases, binding sites become occupied more quickly because the amount of adsorbent is limited (Sakaguchi and Nakajima, 1991).

**Sorbent Dose:** The effect of adsorbent dosage on adsorption follows the same pattern where the rate of adsorption increases proportionally to the amount of adsorbent. Nevertheless, there is no significant increase in adsorption capacity beyond the optimal mass (Liu 2002). (modak and Natarajan 1995) reported that Metal ion removal capacity of any adsorbent is directly related to the number of available binding sites. Therefore, greater available surface area and more binding sites can be attributed when the adsorbent dose increases

**Effect of contact time** is an important parameter because this factor determines the adsorption kinetics of an adsorbate at a given initial concentration of the adsorbate. Initially adsorption increases when the contact time between two phases is higher. Reaching the optimal value (specific in every case), maximum adsorption occurs, followed by an equilibrium state when there is no considerable increase or decrease in adsorption by increasing time span (Hanif and Akhtar, 2007).

**Pb and Zn:** Lead (II) is a well-known highly toxic substance and is a cumulative poison, exposure to which can produce a wide range of adverse health effects. Both adults and children can suffer from the effects of lead poisoning, but childhood lead poisoning is much more frequent (Russell 1989). Humans are exposed to lead, for example, through deteriorating paint, household dust, bare soil, air, drinking water, food, ceramics, home remedies, hair dyes and other cosmetics as reported by Russell 1989 and (Hanif and Akhtar, 2007).

Zinc, an essential trace mineral, is required for the metabolic activity of 300 of the body's enzymes, critical to tissue growth, wound healing, taste acuity, connective tissue growth and maintenance, immune system function, prostaglandin production, (Russell 1989). Zinc is found in meat, liver, kidney, fish, chicken and cereals, eaten alongside vegetables to enhance zinc absorption. Fats and various non-nutritive foods, Fish, fruits, cakes, Poultry, pork, dairy products and whole grains. Zinc toxicity due to acute or chronic ingestion of high quantities of zinc supplements can also occur and lead

to impaired immune response, hypocupremia, microcytosis, and neutropenia. Consumption of excess zinc can cause ataxia, lethargy.

### 1.3. Tea Leaves (*camellia sinensis*)

Camellia is evergreen or small tree that is usually trimmed to below 2m (6.6 ft) when cultivated for its leaves used in the production of tea. It has strong taproot. The flower is yellow-white, 2.5-4 cm in diameter (Mondelet *al.*, (2007). In the production of tea (black tea), the leave passed through different sub-unit under the production line ranging from tea reception , withering , cutting , fermentation drying and sorting (Fourest, 1992). After production, different grade are obtained based on their sizes. At this point, tea fibers are obtained which do not have any significant use (Fourest, 1992).

The purpose of this experimental work is to understand and analyze result to access the ability of Tea fiber to remove Pb(II) and zinc from aqueous solution and to study the effect of biosorbent dosage, contact time, in removal of Lead using biosorption method.

## 2. Materials and Methods

### 2.1. Sample Collection

Tea fiber for the analysis was obtained from kakara High land tea, Sardauna Local government area which is located at the extreme southeast of Taraba state, Nigeria.

### 2.2. Preparation of Stock Solution

0.1M of lead (II) nitrate  $Pb(NO_3)_2$  (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 1000 mL of distilled water.

0.1M of  $ZnSO_4$ : It was prepared for use throughout the experimental work. 16.14g of zinc sulphate powder was dissolved in 1000mL distilled water.

### 2.3. Estimation of Metal Uptake

The metal uptake,  $q_e$ , was determined using the following equation (O'neil *et al.*, 2011):

$$q_e = \frac{(c_e - c_i)V}{M}$$

Where

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

V = volume of solution (L)

$C_i$  = initial concentration of metal in solution (mg/L)

$C_e$  = final concentration of metal in solution (mg/L)

M= the mass of biosorbent (g)

#### 2.4. Effect of Pb Concentration

Effect of  $Pb^{2+}$  and Zn concentrations were evaluated in this study. Five beakers, each filled with 50 mL of metal solution, containing different concentrations ;0.02 M ,0.04 M, 0.06 M, 0.08 M and 0.1 M . 1 g of the biosorbent was added to each beaker keeping the pH6; Time 1 hour and biosorbent dose 1 g constant. It was agitated for 1 hour on an electric shaker. After that, centrifuge for 3 min to separate the solvent for further analysis by AAS.(Aksuet *al.*, 1992).

#### 2.5. Effect of Biosorbent Dosage

Different amount of biosorbent dose 0.5 g, 1 g, 1.5 g, and 2 g, and were added to five different beakers containing 50 ml Pb(II) and another 50 ml of Zn solution. While keeping the pH 6.0, temperature 25, concentration 0.1M and time 60 min constant. The beakers were agitated on an electric shaker for 1 hr. centrifuged for 3 min, and separate the solvent from the biosorbent for further analysis(Aksuet *al.*, 1992).

#### 2.6. Effect of Time (Time dependence)

1 g of biosorbent was suspended into 5 different beakers 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 25°C, 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) (Aksuet *al.*, 1992).

### 3. Results and Discussion

The results obtained from all the analysis carried out in this studies considering the various factors affecting biosorption and every other parameters chosen for this studies are discussed in various sub headings below;

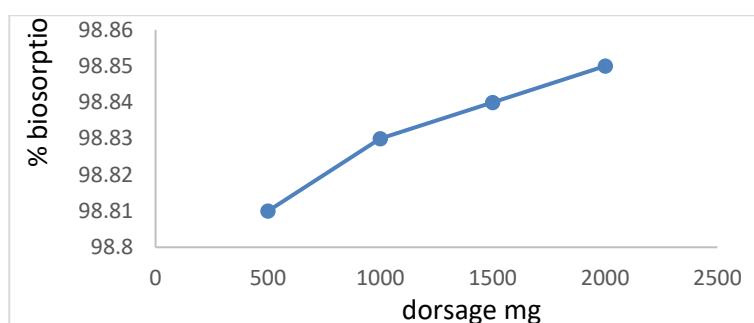
#### 3.1. Effect of Biosorbent Dosage on % Biosorption of Lead

Adsorbent quantity is very imperative feature as it determines the extent of removal of metal and may be used to determine the cost of adsorbent per unit volume of solution to be treated.

The effect of adsorbent dosage on the removal of Pb was investigated and found that the % absorption of lead increase with increase in biosorbent dosage (tea fibre), this is because of the availability of large surface area and adsorption sites with maximum adsorption at 2000 mg as shown in table 1, figure 1 below;

**Table 1.** Effect of biosorption dosage on % biosorption of Lead

Biosorbent Dosage (mg)	Concentration (mg/L)	Biosorption, $q_e$ (%)	Metal Uptake (mg)
500	240.392	98.84	2047.9608
1000	238.020	98.85	1024.0090
1500	247.442	98.81	682.4186
2000	242.151	98.83	511.9462



**Figure 1.** Effect of biosorbent dosage

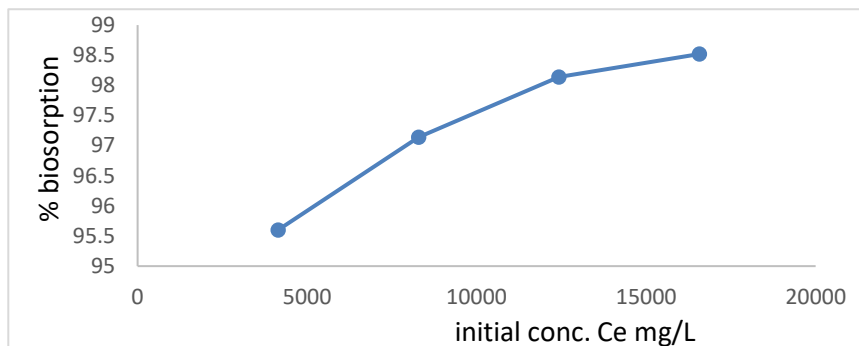
### 3.2. Effect of Initial Concentration of Aqueous Solution on % Biosorption of Lead

The feasibility and efficiency of a biosorption process depends not only on the properties of biosorbents but also on the concentration of the metal ion solution. The initial metal concentration provides an important driving force to overcome all mass transfer resistances of the metal between aqueous and solid phase (Aksu and Akpinar, 2001). The initial concentration of the metal (lead) in the solution remarkably influenced the equilibrium uptake of lead (II).

**Table 2.** Effect of initial concentration of aqueous solution on % biosorption of Lead

Initial conc., $C_0$ (mg/L)	Final conc., $C_e$ (mg/L)	Biosorption, $q_e$ (%)	Metal Uptake, $q_t$	Log $C_e$	Log $q_t$	$C_e/q_t$	In $C_e$
4144	200.612	95.60	197.1694	2.3023	2.2948	1.0174	5.3013
8288	237.023	97.14	402.5488	2.3748	2.6048	0.5888	5.4682
12432	231.163	98.14	610.0418	2.3639	2.7853	0.3789	5.4431
16576	245.681	98.52	816.5159	2.3904	2.9119	0.3009	5.5040

In table 2, It was noted that initial concentration level affected the sorption of lead (II) as generally expected owing to equilibrium process as shown in figure 2 below .The increase in uptake capacity of the biosorbent with the increase in initial metal concentrations is owing to higher availability of metal ions (lead) for the sorption.



**Figure 2.** Effect of initial concentration

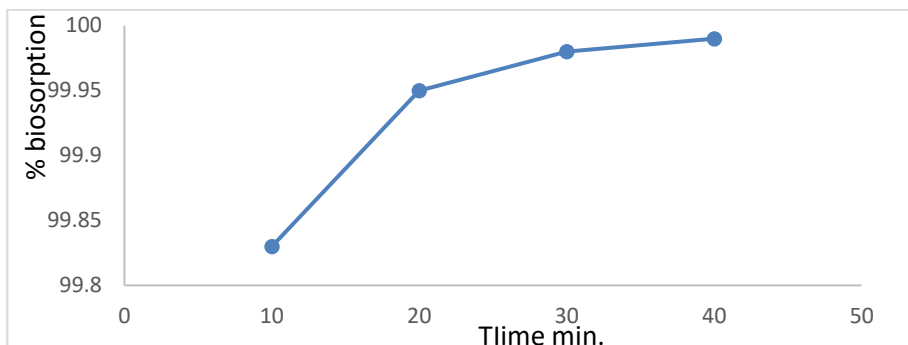
### 3.3. Effect of Agitation Time on % Biosorption of Lead

Contact time plays a vital role in the efficient removal of heavy metals using tea fibre as biosorbent. A series of contact time experiments for lead was carried out and the results are illustrated in Figure 3 below. The figure shows that the adsorption increases with contact time. At first there is a rapid removal efficiency. The changes in the rate of Pb removal might be due to the fact that initially all adsorbent sites were vacant and the solute concentration gradient was high, hence the rate of adsorption was also high. As these sites were exhausted, the uptake rate was controlled by the rate at which the adsorbate is transported from the exterior to the interior sites of the adsorbent particles (Dorris et al., 2000). The lead uptake rate by adsorbent was observed to decrease significantly, due to the decrease in number of adsorption sites as well as lead concentration. The number of sites on the adsorbent filled up by the.

**Table 3.** Effect of agitation time

Time, t (min)	Final Concentration Ce (mg/L)	% biosorption $q_e$	metal uptake $q_t$ (mg)
10	35.347	99.83	1634.2326
20	10.637	99.95	1035.4683
30	3.012	99.98	1035.8494
40	2.738	99.99	1035.8631





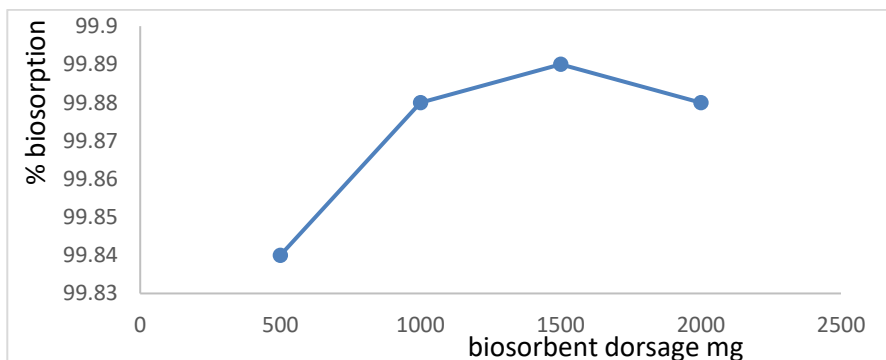
**Figure 3.** Effect of contact time

### 3.4. Effect of Biosorbent Dosage on % Biosorption of Zinc

To determine the optimum biomass dosage for biosorption of Zn, different biomass dosages ranging from 500 mg to 2000 mg were chosen. It was observed that the biosorption of Zinc was higher at biomass dosage of 1500 mg. and it was observed to start to decrease at higher dosage as shown below.

**Table 4.**Effect of biosorbent dosage on % biosorption of Zinc

Biosorbent Dorsage (Mg)	Final Concentration Ce ( mg/L )	% biosorption $q_e$	metal uptake $q_t$ (mg)
500	10.456	99.84	326.3772
1000	7.802	99.88	326.5099
1500	7.545	99.89	326.5228
2000	7.602	99.88	326.5199



**Figure 4.** Effect of biosorbent dosage

### 3.5. Effect of Initial Concentration of Aqueous Solution on % Biosorption of Zinc

The increase in uptake capacity with increase in initial metal concentration has been accounted for in terms of higher availability of metal ions for sorption (Das, 2011). The increase in concentration leads to increase in collision between the metal ions and the sorbent which is a major factor in kinetics for increase in the rate of chemical reactions.

It was observed from this analysis that there was an increase on the biosorption with increase in concentration, as shown in below.

**Table 5.** Effect of initial concentration of aqueous solution on % biosorption of zinc

Initial conc mg/l	Final conc mg/l	% metal uptake $q_e$	Log $C_e$ log $q_t$ mg/g	$C_e/q_t$	ln $C_e$ Conc.	$C_o$ Conc.	$C_e$ sorption $q_t$
1307.6	6.887	99.47	65.0357	0.8380	1.8132	0.1059	1.9296
2615.2	7.224	99.72	130.3988	0.8588	2.1153	0.0554	1.9774
3922.8	7.487	99.80	195.748	0.87470	2.2917	0.0401	2.0601
52304	7.754	99.85	261.323	0.8895	2.4169	0.0297	2.0482

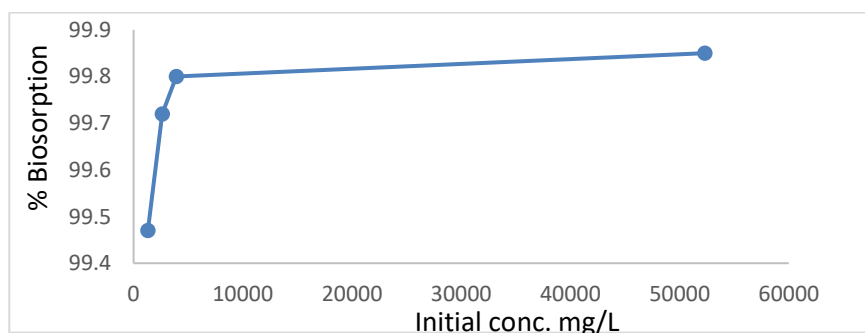


Figure 5. Effect of initial concentration

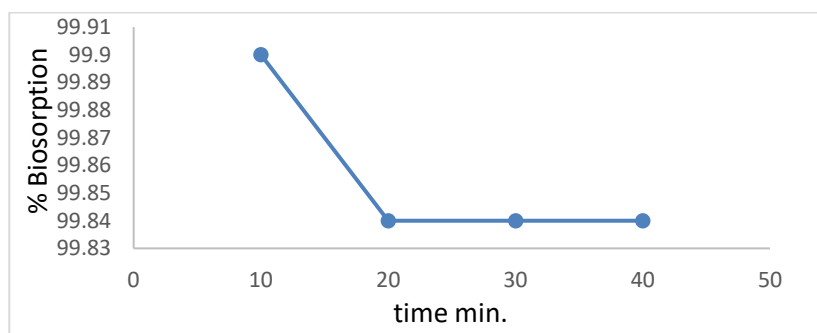
### 3.6. Effect of Agitation Time on % Biosorption of Zinc

Effect of agitation time was conducted to determine the % biosorption, it was observed that a rapid absorption occurred at 10 min, there after constant value was recorded as presented in table 3 and figure 3. The changes in the rate of Pb removal might be due to the fact that initially all adsorbent sites were vacant and the solute concentration gradient was high, hence the rate of adsorption was also high. As these sites were exhausted, the uptake rate was controlled by the rate at which the adsorbate is transported from the exterior to the interior sites of the adsorbent particles (Dorris et al., 2000). The Zinc

uptake rate by adsorbent was observed to maintained value after first absorption, due to the decrease in number of adsorption sites as well as Zinc concentration.

**Table 6.** Effect of agitation time on % biosorption of Zinc

Time t	Final conc. $C_e$ mg/l	% biosorption $q_e$	Metal uptake $q_t$ mg
10	6.2480	99.90	326.5876
20	10.247	99.84	326.3877
30	10.464	99.84	326.3768
40	10.772	99.84	326.3614



**Figure 6.** Effect of agitation time

### 3.7. Kinetic Study of Biosorption of Lead and Zinc

#### 3.7.1. Lagergren Pseudo First-order

This model proposed by Lagergren was used to describe the kinetics. The linearized form is shown in equation below

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

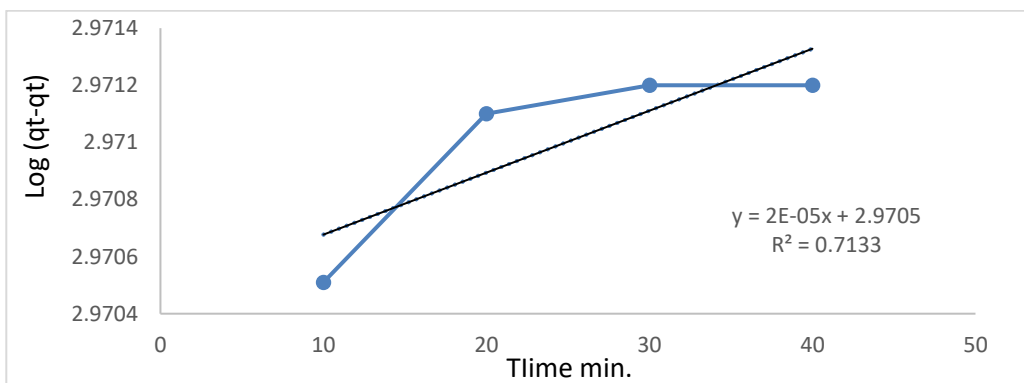
Where  $q_e$  (mg/g),  $q_t$  (mg/g) are adsorption capacity at equilibrium and at time  $t$ , respectively.  $K_1$  is the rate constant of pseudo first-order adsorption (L/min). The value of the constants  $q_e$ ,  $K_1$  and  $R^2$  obtained from the linear plot of  $\log(q_e - q_t)$  vs  $t$  (fig 4.1.4 and 4.1.9) of Lead and Zinc are shown in tables below. The model provided a better fit both Lead and Zinc kinetic studies with regression  $R^2 < 0.9$ . For zinc, a regression  $R^2 < 0.9$ , but lead is more fit to the model when compared between the two elements.

**Table 7.** First order kinetic of biosorption of Lead

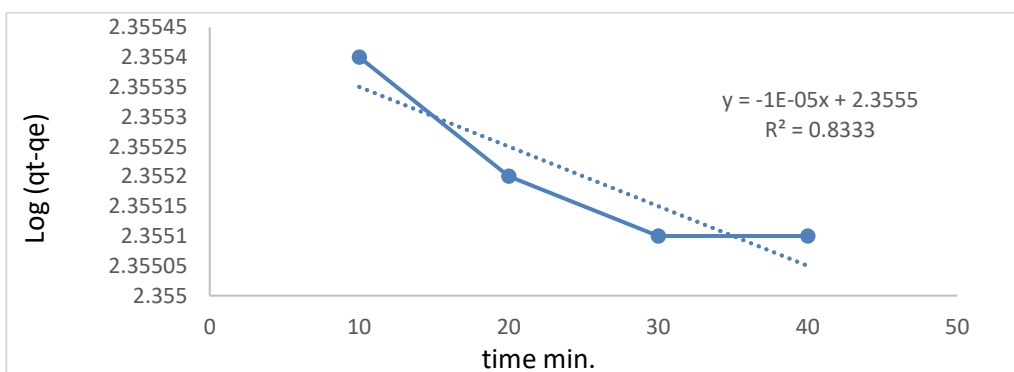
Time t (min)	Final Conc. (mg/l)	% Biosorption (mg)	qe (mg/g)	Metal uptake qt (mg/g)	Log (qt-qe)
10	35.347	99.83		1634.2326	2.9705
20	10.637	99.95		1035.4683	2.9711
30	3.012	99.98		1035.8494	2.9712
40	2.738	99.99		1035.8631	2.9712

**Table 8.** First order kinetic of biosorption of Zinc

Biosorbent dorsage mg	Final Conc. Ce (mg/l)	% biosorption mg	qe	Metal uptake qt	Log (qt-qe)
10	6.2480	99.90		326.5876	2.3554
20	10.247	99.84		326.3877	2.3552
30	10.464	99.84		326.3768	2.3551
40	10.772	99.84		326.3614	2.3551



**Figure 7.** First order kinetic biosorption of lead



**Figure 8.** First order kinetic biosorption of Zinc

### 3.7.2. Pseudo Second-order Model

This model is based on the assumption that chemisorption is the rate-determining step (Das and mondal, 2011) expressed in equation below.

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e}$$

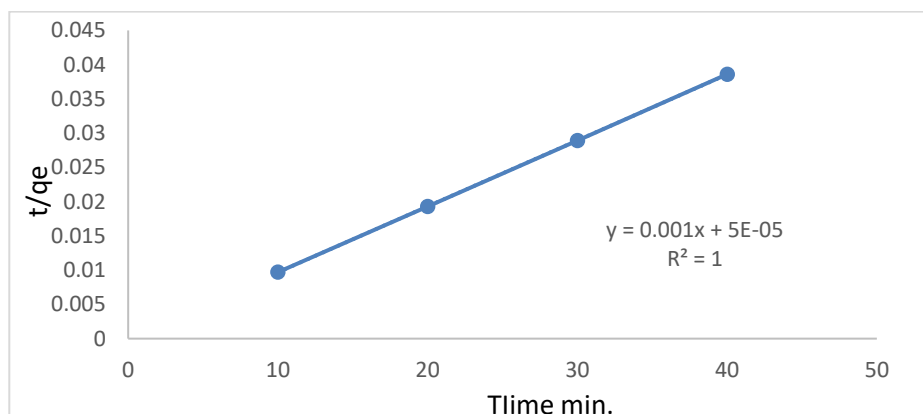
where  $K_2$  is the rate constant of pseudo second- order adsorption rate constant (g/mg/min). The values of  $k^2$ ,  $R^2$  and  $q_e$  were calculated from the plots of  $t/q_t$  versus  $t$  of lead and zinc (4.2.2.1 and 4.2.2.2) as shown in tables below. The regression  $R^2 > 0.9$ , for both lead and Zinc meaning that this model provided the best fit for the adsorption data than other kinetic models previously discussed.

**Table 9.** Second order kinetic of biosorption of lead

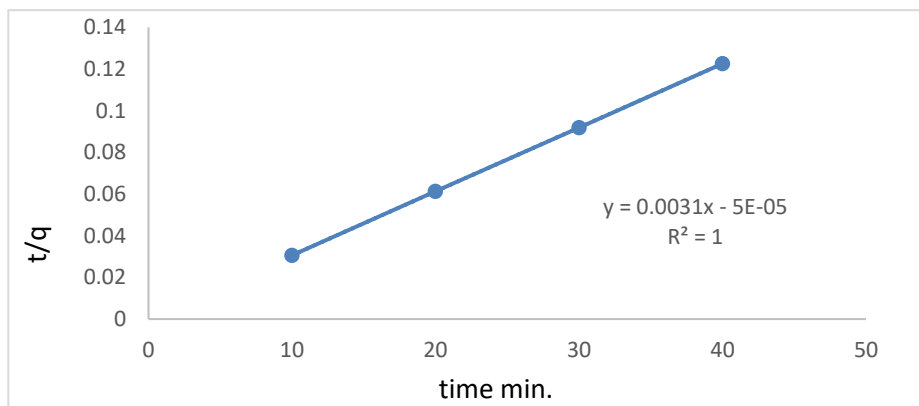
Time t (min)	Final Conc. $C_e$ (mg/l)	% Biosorption $q_e$	Metal uptake $q_t$ mg	$t/q_e$
10	35.347	99.83	1634.2326	0.0097
20	10.637	99.95	1035.4683	0.0193
30	3.012	99.98	1035.8494	0.0289
40	2.738	99.99	1035.8631	0.0386

**Table 10.** Second order kinetic of biosorption of Zinc

Biosorbent dorsage mg	Final Conc. $C_e$ (mg/l)	% Biosorption $q_e$	Metal uptake $q_t$ (mg)	$t/q_t$
10	6.2480	99.90	326.5876	0.0306
20	10.247	99.84	326.3877	0.0613
30	10.464	99.84	326.3768	0.0919
40	10.772	99.84	326.3614	0.1226



**Figure 9.** Second order kinetic Biosorption of Lead



**Figure 10.** Second order kinetic zinc

## 4. Conclusions

This present study confirms the industrial and general use of tea fibre (*camellia sinensis*) as good biosorbent for the treatment of industrial waste water containing heavy metals like lead (Pb) and Zinc (Zn). The obtained data could be used for further studies like the use of treated tea fibre to improve the efficiency of biosorption, processing direction of modeling, of regeneration and immobilization of biosorbents.

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