

# Biosorption Performance of *Wrightia Tinctoria* Leaf Powder for the Removal of Manganese

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**Abstract:** This study determines Kinetic studies on adsorption of Pb ions from an aqueous solution using an adsorbent from plant source, low-cost called as “Parthenium Stem powder” leaves, they have a common name as “Parthenium”. The adsorption studies were carried out in a batch process and at six different parameters such as agitation time and adsorbent dosage, initial concentration of Parthenium stem in the aqueous solution, pH of aqueous solution and the temperature effects were determined.

From the experimentation it is determined that 0.5g of “Parthenium” stem powder of 53 µm size was obtained to be enough to remove 60.08% of 100mg/L concentration of Pb from 50mL of aqueous solution in 50 min. Results showed that adsorption of Pb increased with increase in adsorbent dosage. A significant increase in percentage removal of Pb was obtained as pH value increases from 4 to 5 and the percentage removal is Maximum as pH value increases from 6 to 7. The percentage removal decreases as pH value is beyond 7.

The Freundlich and Langmuir models were applied to describe the equilibrium isotherms and isotherm constants were determined. The Kinetic studies show that the adsorption of Pb on the “Parthenium” stem powder follows pseudo-second-order kinetics. Various thermodynamic parameters such as enthalpy, Gibbs free energy and entropy of adsorption were also determined.

Hence, the results show that “Parthenium” stem powder is effective in Pb removal and can be appreciably considered as most versatile, economical and feasible adsorbent for reclamation of Pb from aqueous solutions.

**Keywords:** *Wrightia Tinctoria* Leaf Powder, “Parthenium” Stem powder.

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## 1. INTRODUCTION

Disposal of solid, liquid and gaseous wastes produce by the domestic households, industrial units and man have been polluting the environment. The natural resources land water and air being contaminated every day by this type of pollution. About 80% of earth’s Surface is covered by water, but only some part is available for drinking, agriculture, domestic and industrial purposes as the rest of the water is locked up in oceans as salt water, polar ice caps, glaciers and underground. There has been increasing concern in the recent years over the discharge of the industrial wastewaters containing heavy metals besides other trace chemicals, which interferes with the designated best use of water. It is well perceived that there is permissible limit of each metal, above which they are generally toxic and some forms are even fatal to human and animal beings.

The toxic metals occur in very small quantities in the earth’s crust (<100ppm) and hence called “ trace metals” which are further arbitrarily divided on the basis of their densities less than 5gm/cc called light metals and those above 5gm/cc are called heavy metals. Biosorption of heavy metals is one of the possible alternative technologies involved in the removal of toxic metals from industrial waste stems and natural waters using low cost adsorbents. It is a potential and interesting alternative to conventional processes for the removal of metals such as ion exchange processes. Biosorption of metal ions from aquatic system using microbial biomass, including algae, fungi and bacteria has gained importance in the recent

year. It is a rapid, reversible, economical and eco friendly technology in contrast to conventional chemical methods of removal metal ions from industrial effluent.

The effluents from different metallurgical industries containing various heavy metal ions are usually treated with lime to precipitate the metal ions so that the metal ion concentrations can be reduced to minimum for easy disposal of the effluent. Although the lime precipitation is universally accepted technique but it suffers from several drawbacks such as high consumption of reagents, decrease in efficiency of precipitation in the presence of complex ions.

The most important aspect is that the precipitated mass is very unstable which requires elaborate storage technique. To overcome these drawbacks, various alternative methods such as chemical precipitation, ion exchange, reverse osmosis, evaporation, electro dialysis, electrolysis, ultra filtration and adsorption have been developed for the treatment of industrial effluents. Most of the methods required higher expenditure. Out of all these techniques adsorption is the most important technique is highly effective, cheap and an easy method for treatment of metal contaminated water. The effluents released from the industries contained considerable amounts of heavy metal ions like Cu, Zn, Cr and Ni. Metal ions are most important pollutants both in surface water and in ground water. Since levels of metals in the environment have increased because of industrial pollution. The elimination of such ions from water is essential to protect public health. Heavy metal ions such as Cobalt, Zinc, Chromium, Mercury, Lead, Nickel, Cadmium etc are considered as a serious factor, since they are tolerated only micro levels. Of these metals, in particular, Cobalt represents a major toxic pollutant, which calls for special attention. Heavy metals are emitted into the environment by the disposal of domestic, municipal and industrial wastes.

## 2. EXPERIMENTAL PROCEDURE

The present experimentation is carried out batch wise, on biosorption of manganese metal from aqueous solution by a biosorbent – *wrightia tinctoria* leaf powder.

The experimental procedure consists of the following steps.

1. Preparation of the biosorbent,
2. Preparation of the 1000 mg/L of manganese stock solution,
3. Studies on equilibrium biosorption,
4. Studies on biosorption kinetics and
5. Studies on thermodynamics.

### 2.1 Preparation of the biosorbent:

*Wrightia tinctoria* leaf is collected near TLN Sabha Hall in Andhra University, Visakhapatnam and is washed with water and then with distilled water to remove dust and soluble impurities. They are dried at room temperature. The crisp leaves are obtained by sun-drying. The dried leaves are finely powdered and sized by passing the powder through a set of sieves ranging from 53, 75, 103, 125 and 150  $\mu$ m mesh sizes. The powder is collected separately and preserved in glass bottles for use as biosorbent.

### 2.2 Preparation of manganese stock solution:

MnSO<sub>4</sub>.H<sub>2</sub>O is used as the source of manganese stock solution. All the required solutions are prepared with analytical reagents and double-distilled water. 3.11 g of 99% MnSO<sub>4</sub>.H<sub>2</sub>O is dissolved in distilled water of 1.0 L volumetric flask up to the mark to obtain 1000 ppm (mg/L) of manganese stock solution. Synthetic samples of different concentrations of manganese are prepared from this stock solution by appropriate dilutions. 100 mg/L manganese stock solution is prepared by diluting 100 mL of 1000 ppm manganese stock solution with distilled water in 1000 mL volumetric flask up to the mark. Similarly solutions with different metal concentrations such as (20, 40, 80, 120 and 160 mg/L) are prepared. The pH of aqueous solution is adjusted to the desired value by addition of 0.1 N H<sub>2</sub>SO<sub>4</sub> or 0.1N NaOH solution.

$$\text{Manganese equivalent to 1 g is given by} = \frac{(\text{Molecular weight of MnSO}_4 \cdot \text{H}_2\text{O}) * 100}{(\text{Atomic weight of manganese}) * (\text{Purity})}$$

### 2.3 Studies on equilibrium biosorption:

The initial concentration of manganese in the aqueous solution is analyzed in an Atomic Absorption Spectrophotometer (Perkin Elmer A Analyst 200 model). Wave length is 279.5 nm and sensitivity check is 2.5 mg/L. The procedure adopted for the biosorption of manganese is as follows.

### 2.4 Effect of agitation time and dosage:

50 mL of aqueous solution is taken in a 250 mL conical flask and 10 g/L of biosorbent having a size of 53  $\mu\text{m}$  is added. This sample is shaken on an orbital shaker at 180 rpm at room temperature (30  $^{\circ}\text{C}$ ) for 1 min. Similarly 14 more samples are prepared in conical flasks adding 10 g/L of biosorbent and exposed to varying agitation times (3, 5, 10, 15, 20, 25, 30, 40, 50, 60, 90, 120, 150 and 180 min). These samples are filtered separately with whatman filter paper and the filtrates are analyzed in AAS to obtain final concentrations of manganese. Similarly the experiments are carried out for five more dosages (10, 15, 20, 25, 30, 40, 50 and 60 g/L) with different agitation times. The percentage biosorption of manganese is calculated as  $(C_0 - C_e) \times 100 / C_0$ . Graphs are plotted between the agitation time and % biosorption of manganese to identify the equilibrium agitation time and optimum dosage.

### 2.5 Effect of initial concentration of the manganese in aqueous solution:

50 mL of aqueous solution containing 20 mg/L manganese is taken in a 250 mL conical flask and 10 g/L of 53  $\mu\text{m}$  size biosorbent is added. The sample is kept in continuous contact for equilibrium agitation time by shaking on an orbital shaker at room temperature. The sample is filtered. The final manganese concentration of the filtrate is determined in an AAS. The same procedure is repeated for other initial concentrations of manganese in aqueous solution (20, 40, 80, 120 and 160 mg/L).

### 2.6 Effect of pH of the solution:

To study the influence of pH on manganese biosorption, 50 mL of aqueous solution is taken in each of seven conical flasks. The pH values of the solutions are adjusted to 2, 3, 4, 5, 6, 7 and 8 in separate flasks by adding required amounts of 0.1 N  $\text{H}_2\text{SO}_4$  or 0.1N NaOH. 10 g/L of 53  $\mu\text{m}$  size biosorbent is added separately to these flasks. The samples are shaken on an orbital shaker at room temperature for equilibrium agitation time. Then samples are filtered. The manganese concentrations of the filtrates are determined by using AAS.

### 2.7 Studies on biosorption kinetics:

In order to examine the biosorption and to determine the order of the rate of biosorption, 50 mL of aqueous solution containing 20 mg/L of manganese is taken in each of fifteen conical flasks. 10 g/L of biosorbent having 53  $\mu\text{m}$  size is added in each of flasks. The conical flasks are shaken on an orbital shaker for different time intervals at room temperature. They are filtered and the filtrates are analyzed to find the concentrations of manganese in aqueous solutions.

### 2.8 Studies on thermodynamics:

The following procedure is followed to determine the effect of temperature on the rate of biosorption and to evaluate the enthalpy ( $\Delta H$ ), entropy ( $\Delta S$ ) and Gibbs free energy ( $\Delta G$ ). 50 mL of aqueous solution containing 20 mg/L of manganese is taken in each of five conical flasks. 30 g/L of 53  $\mu\text{m}$  size biosorbent is added in each of these flasks. The above five conical flasks are shaken in an orbital shaker for optimum agitation time for five different temperatures 283, 293, 303, 313 and 323 K. These samples are filtered. The manganese concentrations of the filtrates are determined by using AAS. The values of parameters investigated are compiled in table – 4.1.

Table Experimental conditions investigated

S.No.	Parameter	Values Investigated
1	Agitation time, t, min	1, 3, 5, 10, 15, 20, 25, 30, 40, 50, 60, 90, 120, 150 & 180
2	Biosorbent size, $\mu\text{m}$	53, 75, 103, 125 and 150
3	Biosorbent dosage, w, g/L	10, 15, 20, 25, 30, 40, 50 and 60
4	Initial manganese concentration, $C_0$ , mg/L	20, 40, 80, 120 and 160
5	pH of aqueous solution	2, 3, 4, 5, 6, 7 and 8
6	Temperature, K	283, 293, 303, 313 & 323

### 3. RESULTS AND DISCUSSION

In the present investigation, the potential of dry *wrightia tinctoria* leaf powder as biosorbent for biosorption of manganese metal present in an aqueous solution is investigated. The effects of various parameters on biosorption of manganese are studied. The measured data consist of initial and final concentration of manganese in the aqueous solution, agitation time, biosorbent size, biosorbent dosage, pH of the aqueous solution and temperature of the aqueous solution. The experimental data are obtained by conducting batch experiments.

#### 3.1 Equilibrium studies on biosorption:

From the experimentations on biosorption of manganese, the percentage biosorption of manganese is found from the relation  $= \frac{C_0 - C_e}{C_0} \times 100$

The amount of manganese biosorbed per unit mass of the biosorbent,  $q_t$  in mg/g is computed by using the expression:

$$q_t = \frac{C_0 - C_e}{w}$$

The effects of various parameters on biosorption of manganese are discussed below.

#### 3.2 Effect of agitation time:

Duration of equilibrium biosorption is defined as the time required for heavy metal concentration to reach a constant value during biosorption. The equilibrium agitation time is determined by plotting the % biosorption of manganese against agitation time as shown fig. 5.1 for the interaction time intervals between 1 to 180 min. For 53  $\mu$ m size of 10 g/L biosorbent dosage, 62.26 % (1.2452 mg/g) of manganese is biosorbed in the first 5 min. The % biosorption is increased briskly up to 50 min reaching 77.475 % (1.5495 mg/g). Beyond 50 min, the % biosorption is constant indicating the attainment of equilibrium conditions [1, 2].

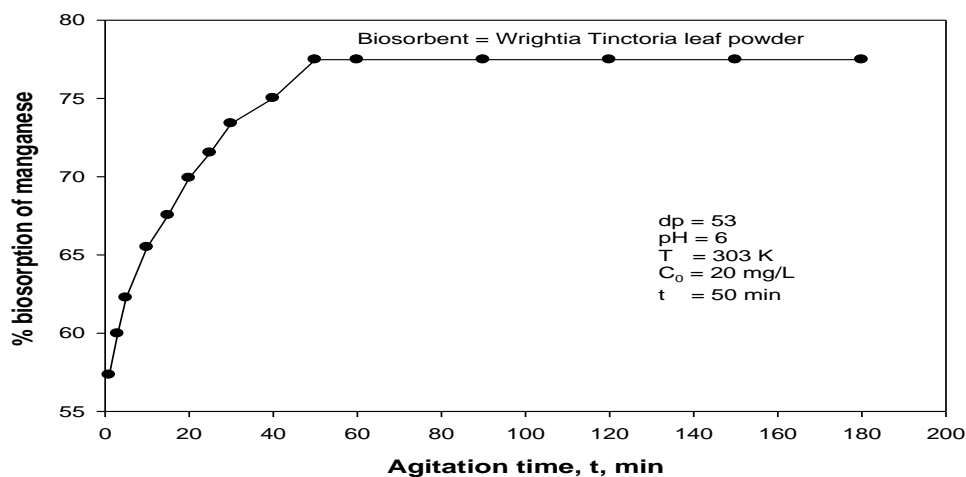


Fig. 5.1 Effect of agitation time on % biosorption of manganese

The maximum biosorption of 77.475 % (1.5495 mg/g) is attained for 50 min of agitation time with 10 g/L of 53  $\mu$ m size biosorbent mixed in 50 mL of aqueous solution ( $C_0 = 20$  mg/L). The rate of biosorption is fast in the initial stages because adequate surface area of the biosorbent is available for the biosorption of manganese. As time increases, more amount of manganese gets biosorbed onto the surface of the biosorbent due to Vanderwaal's forces of attraction and resulted in decrease of available surface area. The biosorbate, normally, forms a thin one molecule thick layer over the surface. When this monomolecular layer covers the surface, the biosorbent capacity is exhausted. The maximum percentage of biosorption is attained at 50 minutes. The percentage biosorption of manganese becomes constant after 50 min. Therefore, all other experiments are conducted at this agitation time.

#### 3.3 Effect of biosorbent size:

The variations in % biosorption of manganese from the aqueous solution with biosorbent size are obtained. The results are drawn in fig.5.2 with percentage biosorption of manganese as a function of biosorbent size. The percentage

biosorption is increased from 60.83 (1.2166 mg/g) to 79.39 % (1.5878 mg/g) as the biosorbent size decreases from 150 to 53  $\mu\text{m}$ . This phenomenon is expected, as the size of the particle decreases, surface area of the biosorbent increases; thereby the numbers of active sites on the biosorbent are better exposed to the biosorbate.

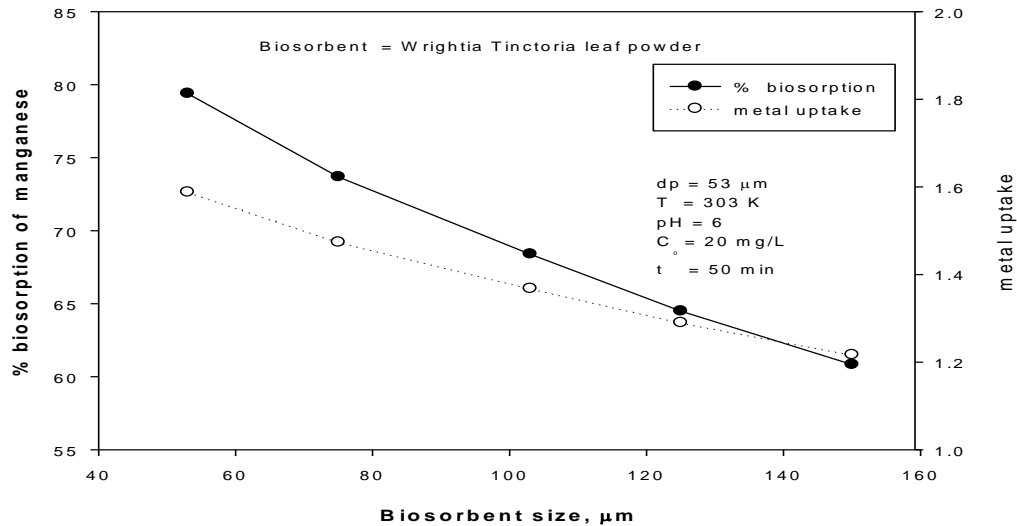


Fig 5.2 Effect of biosorbent size on % biosorption of manganese

### 3.4 Effect of pH:

pH controls biosorption by influencing the surface change of the biosorbent, the degree of ionization and the species of biosorbate. In the present investigation, manganese biosorption data are obtained in the pH range of 2 to 8 of the aqueous solution ( $C_0 = 20 \text{ mg/L}$ ) using 10 g/L of 53  $\mu\text{m}$  size biosorbent. The effect of pH of aqueous solution on % biosorption of manganese is shown in fig.5.3. The % biosorption of manganese is increased from 58.77 % (1.1754 mg/g) to 80.17 % (1.6034 mg/g) as pH is increased from 2 to 6 and decreased beyond the pH value of 6 [3]. % biosorption is decreased from pH 7 to 8 reaching 66.67 % (1.3334 mg/g) from 73.83 % (1.4766 mg/g). Low pH depresses biosorption due to competition with  $\text{H}^+$  ions for appropriate sites on the biosorbent surface. However, with increasing pH, this competition weakens and Manganese ions replace  $\text{H}^+$  ions bound to the biosorbent [4, 5].

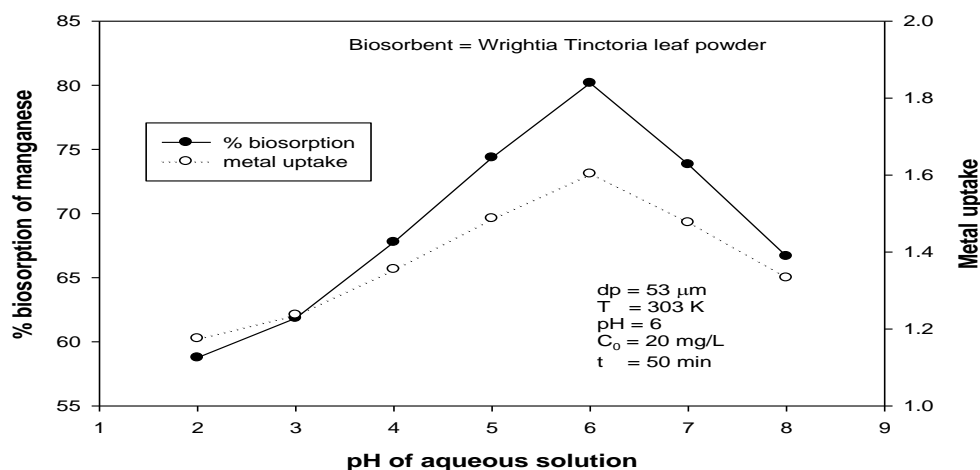


Fig. 5.3 Effect of pH on % biosorption of manganese

### 3.5 Effect of initial concentration of manganese:

The effect of initial concentration of manganese in the aqueous solution on the percentage biosorption of manganese is shown in fig.5.4. The percentage biosorption of manganese is decreased from 80.88 % (1.6176 mg/g) to 62.358 % (9.9774 mg/g) with an increase [6] in  $C_0$  from 20 mg/L to 160 mg/L. Such behavior can be attributed to the increase in the amount of biosorbate to the unchanging number of available active sites on the biosorbent.

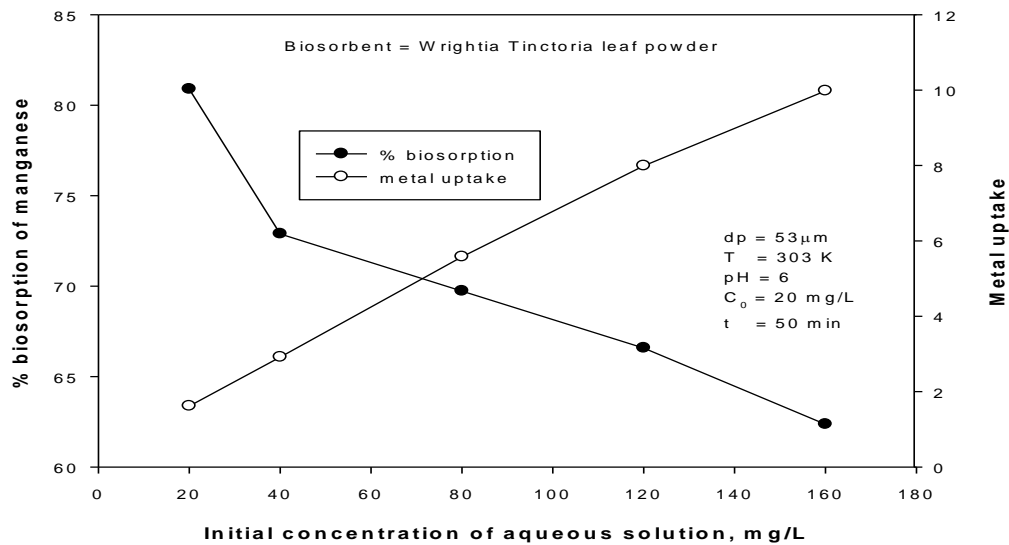


Fig: 5.4 Effect of initial concentration for the biosorption of manganese

### 3.6 Effect of biosorbent dosage:

The percentage biosorption of manganese is drawn against biosorbent dosage for 53  $\mu\text{m}$  size biosorbent in fig.5.5. The biosorption of manganese increased from 79.91 % (1.5982 mg/g) to 87.76 % (0.58506 mg/g) with an increase in biosorbent dosage from 10 to 30 g/L. Such behavior is obvious because with an increase in biosorbent dosage, the number of active sites available for manganese biosorption would be more. The change in percentage biosorption of manganese is marginal from 87.76 % (0.5850 mg/g) to 88.075 % (0.2935 mg/g) when 'w' is increased from 30 to 60 g/L. Hence all other experiments are conducted at 30 g/L dosage.

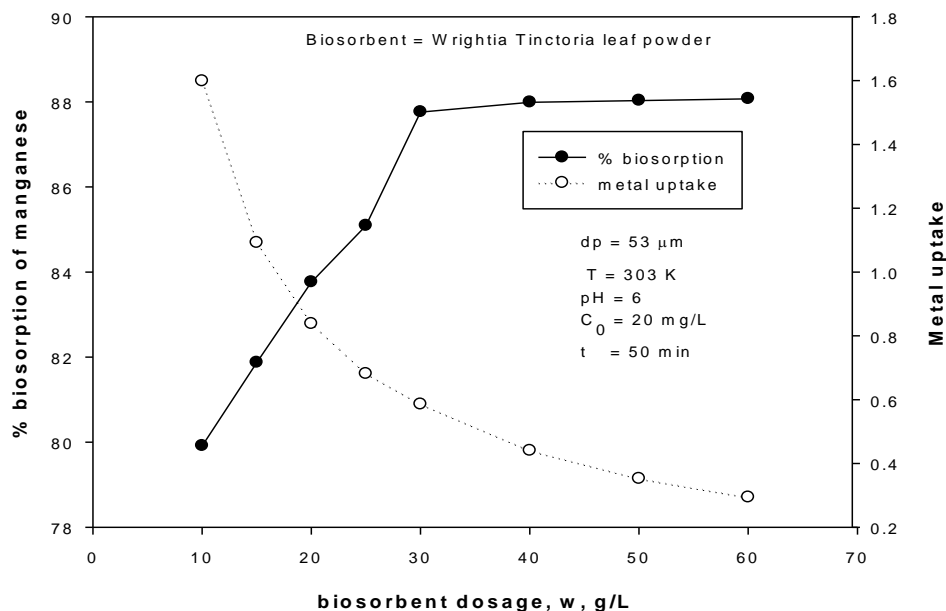


Fig. 5.5 Effect of biosorbent dosage on % biosorption of manganese

### 3.7 Effect of Temperature:

The effect of temperature on the equilibrium metal uptake was significant. The effect of changes in the temperature on the manganese uptake is shown in Fig.5.6. When temperature was lower than 303 K, Manganese uptake increased with increasing temperature, but when temperature was over 303 K, the results were on the contrary. This response suggested a different interaction between the ligands on the cell wall and the metal. Below 303 K, chemical biosorption mechanisms

played a dominant role in the whole biosorption process, biosorption was expected to increase by increase in the temperature [7] while at higher temperature, the plant powder were in a nonliving state, and physical biosorption became the main process. Physical biosorption reactions were normally exothermic, thus the extent of biosorption generally is constant with further increasing temperature.

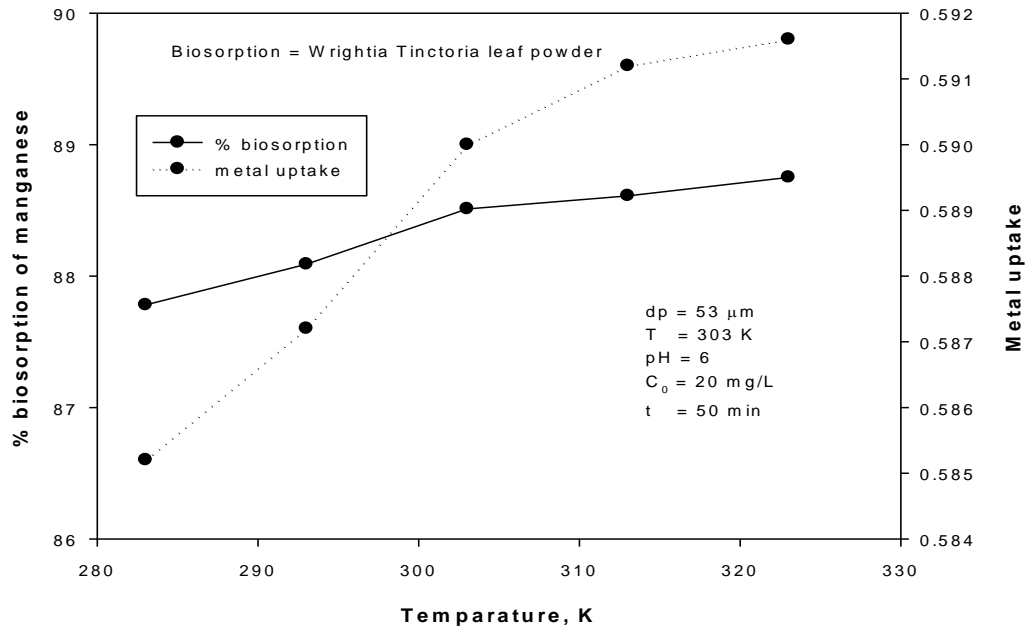


Fig.5.6 Effect of temperature for the biosorption of manganese

### 3.8 Langmuir isotherm:

Irving Langmuir [8] developed an isotherm named Langmuir isotherm. It is the most widely used simple two-parameter equation. This simple isotherm is based on following assumptions:

Biosorbates are chemically biosorbed at a fixed number of well-defined sites

Each site can hold only one biosorbate specie

All sites are energetically equivalent

There are no interactions between the biosorbate specie

The Langmuir relationship is hyperbolic and the equation is:

$$q_e/q_m = bC_e / (1+bC_e) \quad \dots\dots\dots (5.1)$$

Equation (5.1) can be rearranged as

$$(C_e/q_e) = 1/(bq_m) + C_e/q_m$$

From the plots between  $(C_e/q_e)$  and  $C_e$ , the slope  $\{1/(bq_m)\}$  and the intercept  $(1/b)$  are calculated. Further analysis of Langmuir equation is made on the basis of separation factor,  $(R_L)$  defined as  $R_L = 1/(1+bC_e)$

$0 < R_L < 1$	indicates	favorable adsorption
$R_L > 1$	indicates	unfavorable adsorption
$R_L = 1$	indicates	linear adsorption
$R_L = 0$	indicates	irrepressible adsorption

Langmuir isotherm is drawn for the present data and shown in Fig.5.7. The equation obtained ' $n$ '  $C_e/q_e = 0.0583 C_e + 2.678$  with a good linearity (correlation coefficient,  $R^2 \sim 0.9286$ ) indicating strong binding of manganese ions to the surface of *Wrightia Tinctoria* leaf powder.

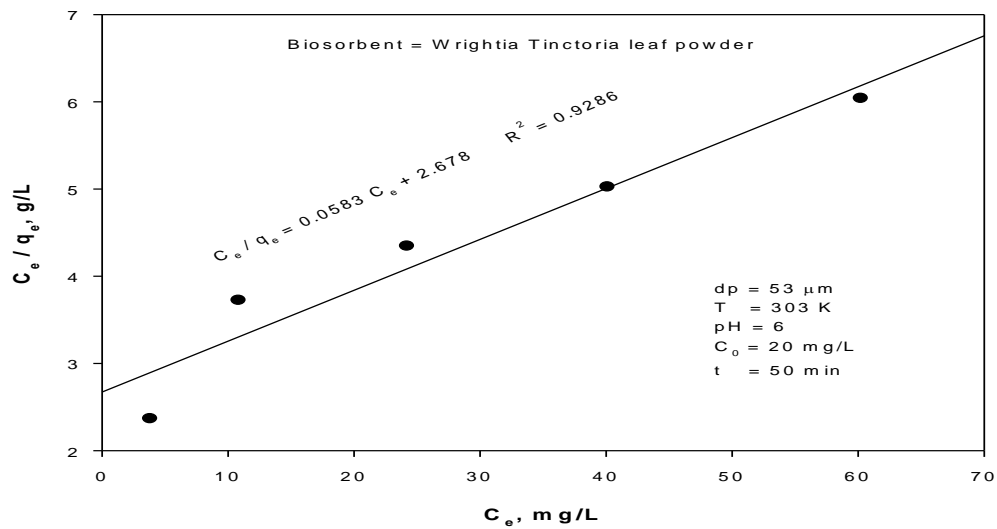


Fig: 5.7 Langmuir isotherm for biosorption of manganese

### 3.9 Freundlich isotherm:

Freundlich [9] presented an empirical biosorption isotherm equation, that can be applied in case of low and intermediate concentration ranges. It is easier to handle mathematically in more complex calculations.

The Freundlich isotherm is given by

$$q_e = K_f C_e^n$$

Where  $K_f$  (mg) represents the biosorption capacity when metal equilibrium concentration and  $n$  represents the degree of dependence of biosorption with equilibrium concentration Taking logarithms on both sides, we get

$$\log q_e = \log K_f + n \log C_e$$

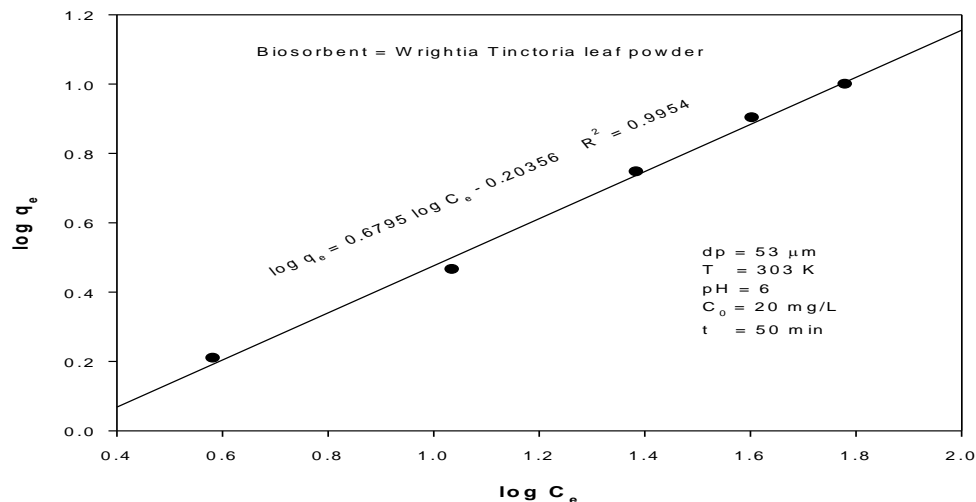


Fig.5.8 Freundlich isotherm for biosorption of manganese

Freundlich isotherm is drawn between  $\log q_e = 0.6795 \log C_e - 0.20356$ ;  $\log C_e$  and  $\log q_e$  in Fig.5.8 for the present data. The resulting equation has a correlation coefficient of 0.9954.

The 'n' value in the above equations satisfies the condition of  $0 < n < 1$  indicating favorable biosorption.

### 3.10 Kinetics of biosorption:

The order of biosorbate – biosorbent interactions have been described using kinetic model. Traditionally, the first order model of Lagergren [11] finds wide application. In the case of biosorption preceded by diffusion through a boundary,



the kinetics in most cases follows the first order rate equation of Lagrangen:

$$(dq_t/dt) = K_{ad} (q_e - q_t)$$

where  $q_e$  and  $q_t$  are the amounts adsorbed at  $t$ , min and equilibrium time and  $K_{ad}$  is the rate constant of the pseudo first order biosorption [12].

The above equation can be presented as

$$\int (dq_t / (q_e - q_t)) = \int K_{ad} dt$$

Applying the initial condition  $q_t = 0$  at  $t = 0$ , we get

$$\log (q_e - q_t) = \log q_e - (K_{ad}/2.303) t$$

$$\log (q_e - q_t) = 0.2023t + 0.6497$$

Plot of  $\log (q_e - q_t)$  versus 't' gives a straight line for first order kinetics, facilitating the computation of adsorption rate constant ( $K_{ad}$ ).

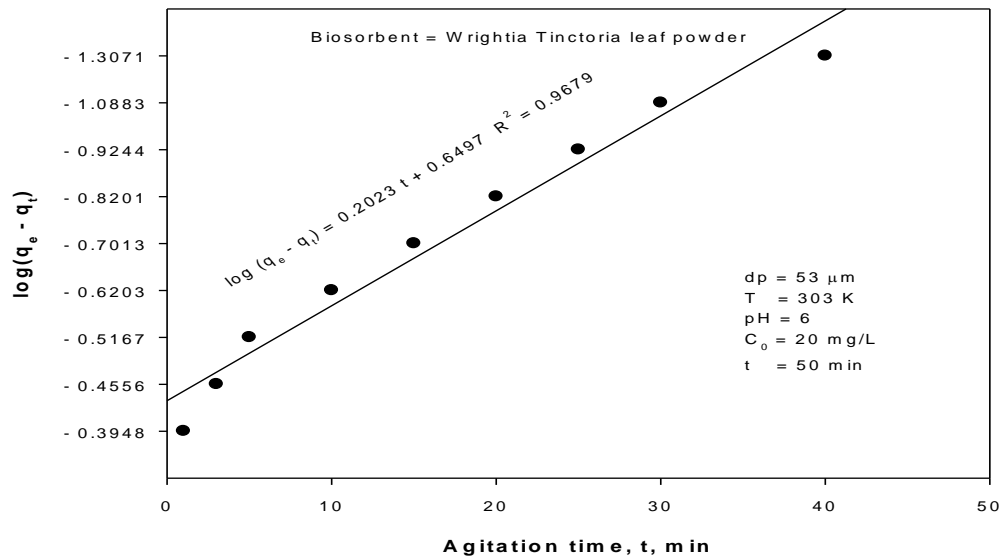


Fig.5.10 first order kinetics for biosorption of manganese

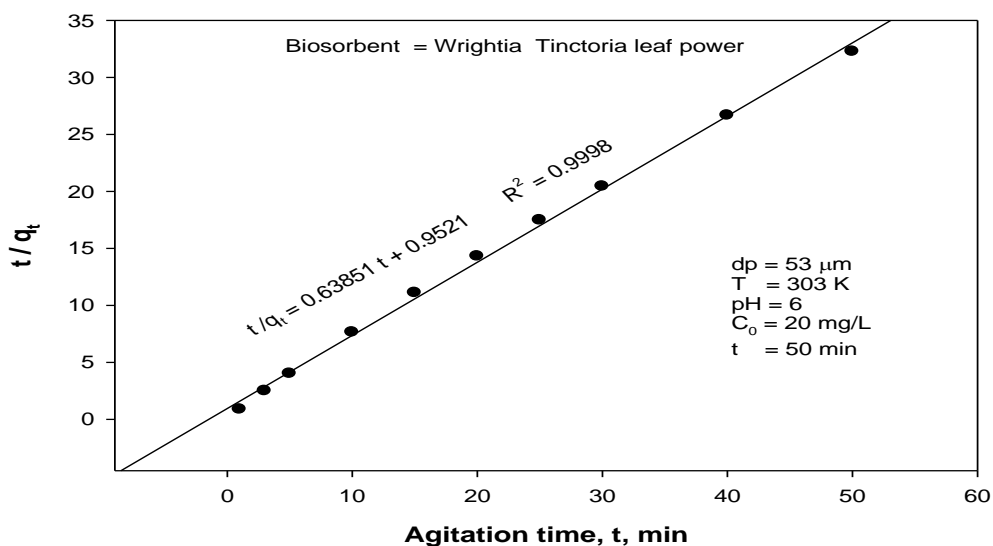


Fig: 5.11 Second order Kinetics for biosorption of manganese

#### 4. CONCLUSIONS

Investigations are carried out to find out the equilibrium, kinetics and Thermodynamic parameters for biosorption of manganese from an aqueous solution using *Wrightia Tinctoria* leaf powder. The analysis of the experimental data result in the following conclusions

1. The equilibrium agitation time for manganese biosorption is 50 min.
2. The percentage biosorption of manganese is increased from 60.83 (1.2166 mg/g) to 79.39 % (1.5878 mg/g) as the biosorbent size decreases from 150 to 53  $\mu\text{m}$ .
3. The % biosorption of manganese is increased from 58.77 % (1.1754 mg/g) to 80.17 % (1.6034 mg/g) as pH is increased from 2 to 6 and decreased beyond the pH value of 6. % biosorption is decreased from pH 7 to 8 reaching 66.67 % (1.3334 mg/g) from 73.83 % (1.4766 mg/g)
4. With an increase in the initial concentration of manganese in the aqueous solution, the percentage biosorption of manganese from the aqueous solution is decreased.
5. The percentage biosorption of manganese is increased significantly with increase in biosorbent dosage up to 30 g/L and thereby remained constant. The biosorption of manganese increased from 79.91 % (1.5982 mg/g) to 87.76 % (0.58506 mg/g) with an increase in biosorbent dosage from 10 to 30 g/L
6. The kinetic studies show that the biosorption of manganese is better described by pseudo second order kinetics. ( $K_2 = 0.4282$ ),  $R^2 = 0.9998$
7. The thermodynamic data show that percentage biosorption increases with marginal increase in temperature.

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