

Comparative Study for Removal of Chromium and Copper by Using Red Sandal Wood Leaf Powder from Aqueous Solution

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Abstract:

Rapid Industrialization causing long lasting environmental problems. The pollutants released from them pollute the land, air, water. Pollution from the heavy metals is one of the major concerns. Heavy metals include Nickel, Lead, Chromium, Copper, Arsenic. In this paper, we studied the adsorption of chromium and copper on the Pterocarpus Santalinus. This adsorbent was developed from the leaves of the Red Sandal Wood tree. The adsorbent was a fine powder. Adsorption is one of the best techniques for the removal of heavy metals. All the experiments were carried out in batch mode at room temperature. The effect of various parameters and thermodynamic parameters were studied, Langmuir, Freundlich isotherms were studied and published in results. Kinetic data was analyzed and published.

Samples are analyzed using a Shimadzu UV – 1900 spectrophotometer at 540nm wavelength.

Keywords: Heavy Metals, Chromium, Copper, Pterocarpus Santalinus, Freundlich isotherm, Langmuir isotherm, Kinetic Studies, Thermodynamic Parameters.

1. Introduction

Small quantities of minerals and vitamins are required for the growth and various biochemical and physiological functions in living organisms. But when the concentration limits exceed, they became toxic[6]. The environmental quality is reducing day by day by reaching saturation points. Most of the industries discharging waste into the water bodies without proper treatment methods. The aqueous waste from the industries contains heavy metals. Heavy metals are the elements atomic weights ranging from 63.5 to 200.6. Heavy metals are major pollutants of the water. These contaminants found to be toxic and carcinogenic and effecting both the human and the aquatic life. The most common metals found in waste water are Nickel, Lead, Copper, Chromium, Arsenic and Cadmium[9]. Access to the clean water is the essential requirement for all the organism. Due to increase in pollutants the contaminants increasing day by day in the natural resources. Adsorption is one of the best techniques and can be effectively used to remove pollutants from waste water[2].

In this work we aim to develop adsorbent from the leaves of the Pterocarpus Santalinus tree to treat the Chromium and Copper³.

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Published: 28 January 2026

DOI: <https://doi.org/10.70558/IJST.2026.v3.i1.241172>

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1.1 Sources

Copper: Pesticides, Fungicides, Copper Jewelry, Copper Utensils

Chromium: Steel and Textile industry

1.2 Effects

Copper: Mental disorders, anemia, arthritis, hypertension, insomnia, autism, Inflammation and enlargement of liver, cystic fibrosis.

Chromium: Rashes, Respiratory problem, weak immune system, kidney and liver damage, lung cancer, pulmonary fibrosis.

1.3 Maximum concentrations limit standards

Copper – 0.25 mg/L

Chromium – 0.05 mg/L

1.4 Need for removal of heavy metals

Effluents from the industries discharge into the rivers and lakes and cause the deposition of the heavy metals. Heavy metals can't degrade biologically. Reports from all over the world shows the incidence of accumulation heavy metals in fishes and other components of aquatic systems, some heavy metals at low concentrations are toxic to aquatic organisms. These elements do not degrade and biomagnified in human through food chain. So, it is needed to remove the heavy metals.

1.5 Toxic effects of aquatic life

- Elevated levels cause sub-lethal effects
- Changes in biochemistry, enzyme activity, behavior, reproduction.
- Suppression in growth and development.

1.6 Methods of removal

- Chemical Precipitation
- Ion Exchange
- Electrodialysis
- Coagulation/Flocculation
- Ultrafiltration
- Membrane Filtration
- Reverse Osmosis
- Adsorption

1.7 Adsorbents used in adsorption

- Activated Carbon
- Silica Gel
- Activated Alumina
- Hydrogels

1.8 Low-cost adsorbent

Adsorbents are termed as low cost if they require a little processing and they availability

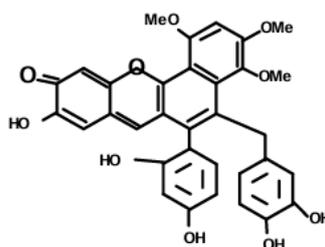
is abundant in nature, or they can get as the waste or by-product from the industries. Naturally from agriculture is one of the sources for low-cost adsorbents.

- Lignin
- Cast iron fillings and steel wool
- Zeolites
- Saw Dust
- Fly Ash
- Carbon Anode Dust
- Agriculture Waste, Tea Waste
- Rice Husk, Coconut Husk
- Neem Leaves, Eucalyptus tree parts [9].

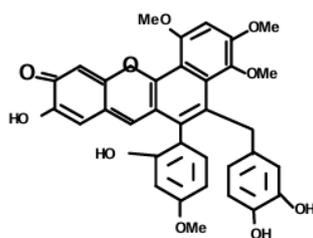
1.9 Red Sandal Wood

Pterocarpus Santalinus, commonly known as red sandalwood, is a deciduous tree natively belonging to the southern Eastern Ghats of India, especially in the state of Andhra Pradesh. The species is highly valued for its dense, dark red heartwood, which contains the pigment Santalin, and for its medicinal and cultural significance. Chemically, red sandalwood biomass is composed of cellulose, hemicellulose, lignin, and phenolic compounds, all of which provide active functional groups such as hydroxyl, carboxyl, and phenolic moieties. These groups are capable of binding with heavy metal ions, making the tree's powdered biomass as good source for adsorption studies.

Pterocarpus species contains santalin, isoflavonoids, terpenoids, phenolic compounds, beta-sitosterol, epicatechin, lupeol⁵.



Santalin A



Santalin B

2. Materials and Methods

2.1 Adsorbent preparation

Red Sandal Wood leaf powder is used as the adsorbent in this work. Red Sandal Wood leaves collected from the Chemical Engineering Department, Sri Venkateswara University College of Engineering, Tirupati.

Leaves are repeatedly washed for several times with water to remove the dirt. Then the leaves are sun dried; dried leaves were grounded to fine powders using domestic mixers. The adsorbent was washed and dried before the start of the experiment to further remove the impurities and color present in it. All the experiments were conducted in batch mode and at the room temperature. Samples were analyzed by using Shimadzu UV-1900 spectrophotometer at the wave length of 540nm. pH of the samples were measured by using Hanna pH meter. Experiments were conducted for various parameters, by varying initial concentrations(50-250 mg/L), varying contact time (10-120 min), varying pH (2-10) and dosage of adsorbent (0.1 – 1g), varying temperature (30 to 60°C)

2.2 Adsorbent capacity

It is defined as the quantity of adsorbate that can take up by adsorbent per unit mass of adsorbent and can be calculated by using the formula as follows[6].

$$q_e = (C_0 - C_e)V/m$$

where q_e is the adsorption capacity

C_0 is the initial metal ion concentration (mg/L)

C_e is the equilibrium metal ion concentration (mg/L)

V is the volume of sample taken (L)

m is the mass of the adsorbent (g)

The percentage removal of the metal from the solution is given by

$$\% \text{ Removal} = ((C_0 - C_e)/C_0) * 100$$

2.3 Stock solution preparation

Copper aqueous solution

2.68 g of Copper Chloride was taken in a 500ml volumetric flask and then diluted with distilled water up to the mark. The resulting concentration of the copper solution is 1000 mg/L. From this copper stock solution (1000 mg/L), different initial concentrations of 50, 100, 200, 300, 400 and 500 mg/L were prepared by diluting with required distilled water[4].

Chromium aqueous solution

2.828 g of $K_2Cr_2O_7$ (analytical grade) is dissolved in one liter of double distilled water to prepare 1000 mg/L of Chromium Stock Solution. 1000 mg/L Chromium aqueous solution is prepared by diluting 50 ml of 1000 mg/L chromium stock solution with distilled water in 500

ml volumetric flask. From this 100 mg/L chromium stock solution, different initial concentrations of 50, 100, 200, 300, 400, 500 mg/L were prepared¹. 0.1N H₂SO₄, 0.1N NaOH are used to maintain desired pH of solution[6].

3. Experimental Results

Experiments conducted by varying time, concentration, adsorbent dosage, p^H. 50 ml solution of desired pH, concentration and adsorbent dosage was taken into a 250 ml conical flask and placed in the rotary shaker of fixed rpm and rotated for specific time. The samples were filter and analyzed[6].

3.1 Effect of Contact Time

The experiment was conducted for 120min, the obtained results were shown. At regular interval of 10 min of time the sample was taken out from the rotary shaker and analyzed. The % removal of Chromium is 80.67 % at 10 min and it increased to 90.52% at 90 min and then equilibrium was obtained. The % removal of Copper is 68.2% at 10 min and it increased to 87.8 at 90 min and after that attained equilibrium.

Maximum adsorption has been achieved during the first 10 min because of the availability of the surface area. After that the surface was occupied and the further there is no availability of surface for metal ion to penetrate and so it reaches the Equilibrium.

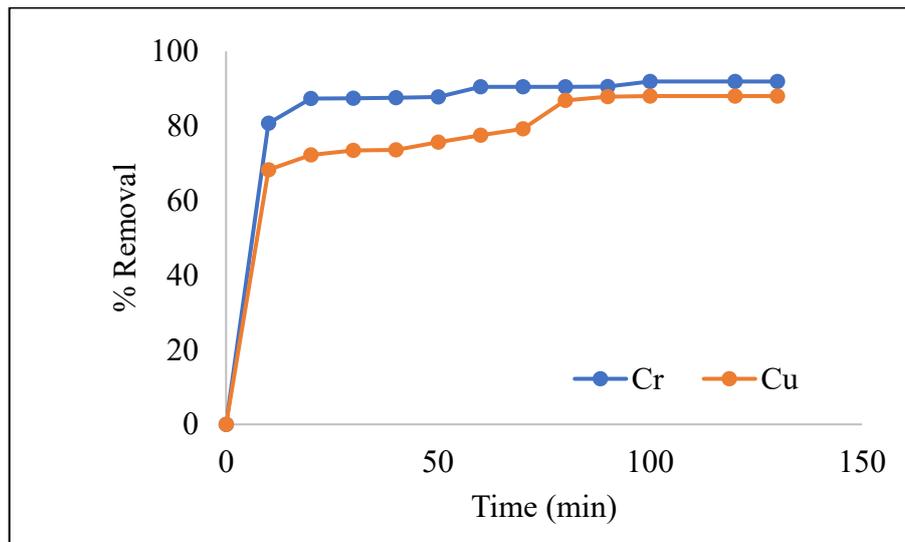


Figure 1: Effect of Contact Time, at 50 mg/L of initial Concentration

3.2 Effect of initial concentration

The Removal of Metals using Red Sandal wood leaf powder at various initial concentration from 50 to 250 mg/L at a dosage of 1 g/L and at room temperature.

The % removal of Chromium decreases from 91.91% to 61.45% and for the Copper it decreased from 93% to 70%.

The adsorbent capacity for Chromium increased from 4.59 to 15.36, and for the Copper it shows that the adsorbent capacity increased from 4.65 to 17.5.

The percentage of the metal removed with increase in initial concentration is decreased due availability of more active sites for adsorption. Conversely the adsorption factor increases due to enhancement in the driving force.

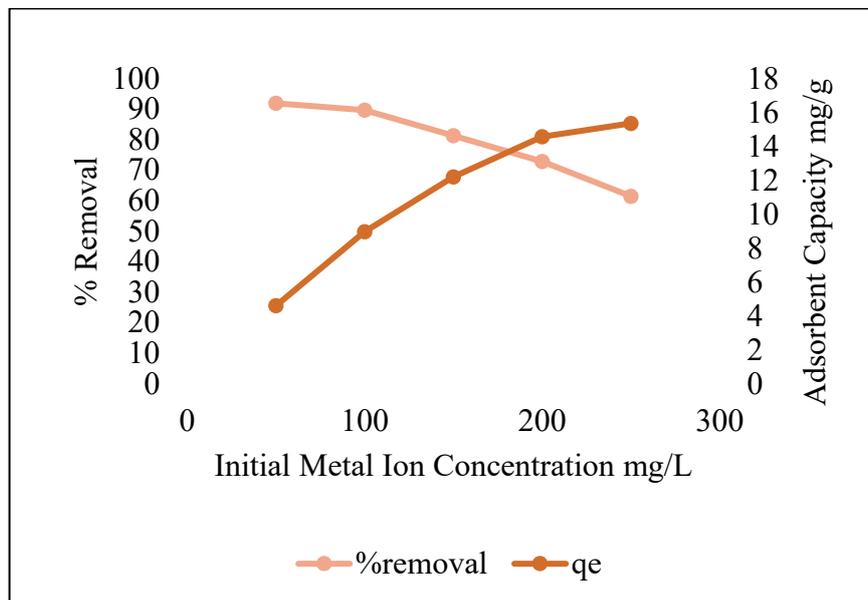


Figure 2: Effect of initial metal ion concentration of Chromium on % Removal and adsorption capacity at adsorbent dose of 1g, volume of 0.1L, and at room temperature

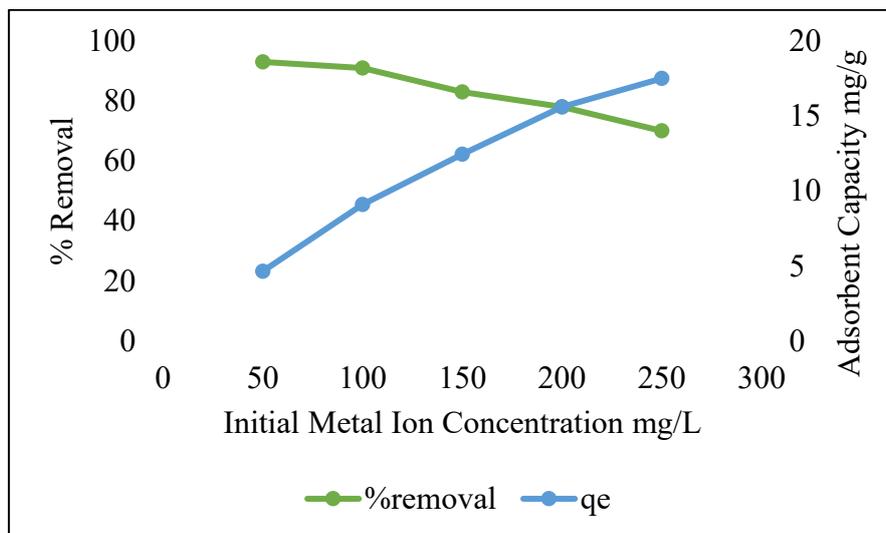


Figure 3: Effect of initial metal ion concentration of Copper on % Removal and adsorption capacity at adsorbent dose of 1g, volume of 0.1L, and at room temperature

3.3 Effect of pH

Effect of the Solution p^H on the percentage removal of metal ions was shown. p^H has important role in the removal of metal ions in the adsorption. It has effect on the both site dissociation and the solution chemistry. The p^H is varied from 2 to 10 and the % removal of Chromium was decreased from 90.44 % to 56.89% and for the Copper the % removal was increased from 82.3% at 2 p^H to 92.4% at p^H of 6 and then reduced to 79%.

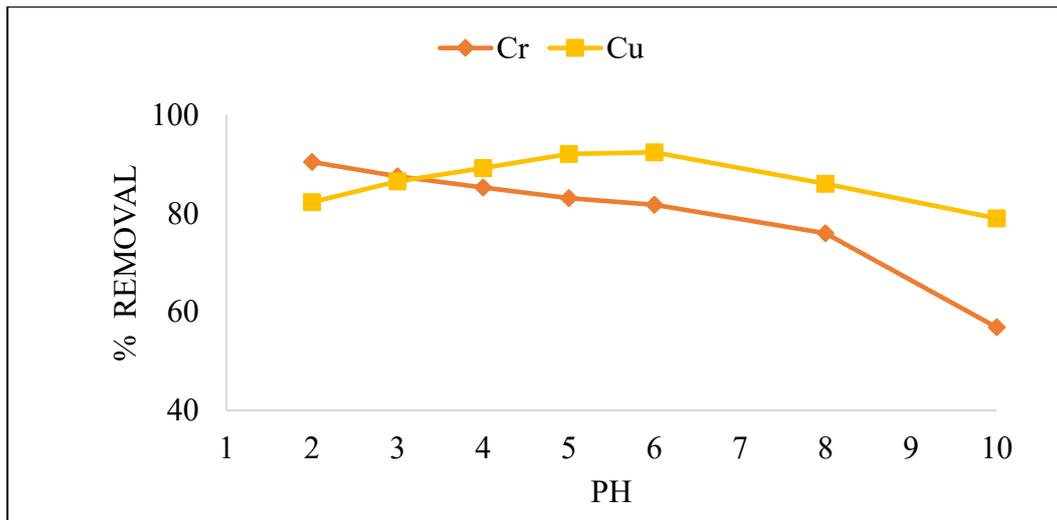


Figure 4: Effect of pH at the initial concentration of 50mg/L, adsorbent dosage of 2g and at room temperature.

3.4 Effect of adsorbent dosage

The % Removal of metal ion increases with increasing in the amount of the adsorbent, this happens due to the increase in the surface area and more availability of the adsorption sites for the adsorption. Conversely the adsorption capacity decreases.

The % removal of Chromium is 68% at 0.1g adsorbent dosage and it increased to 91.91% at adsorbent dosage of 1g. The adsorbent capacity decreases from 34mg/g to 4.595mg/g at adsorbent dosage of 0.1g to 1g.

The % removal of Copper increases from 64% to 93% as the adsorbent dosage varies from 0.1g to 1g. The adsorbent capacity of Copper decreases as 32mg/g at 0.1g of adsorbent dosage and 4.65mg/g at 1g of adsorbent dosage.

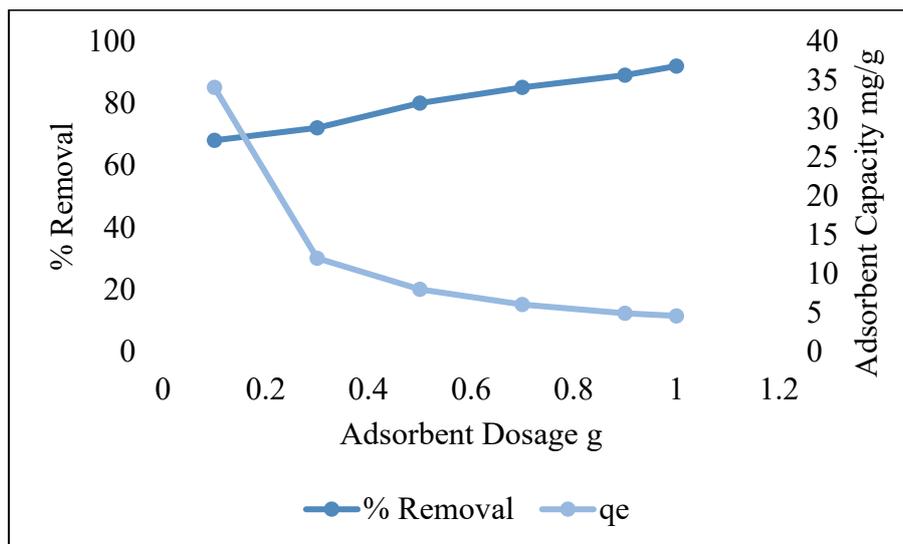


Figure 5: Effect of adsorbent dosage on % removal and adsorption capacity of Cr

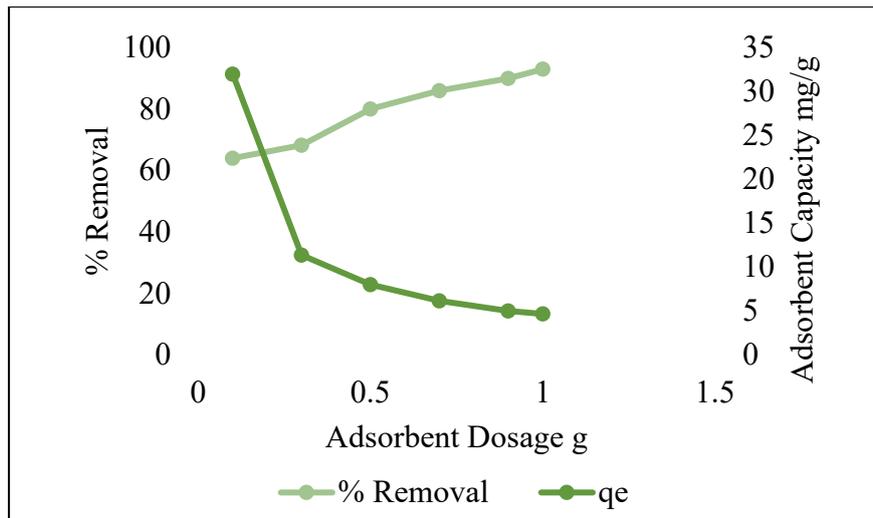


Figure 6: Effect of adsorbent dosage on % removal and adsorption capacity (mg/g) of Cu

3.5 Effect of temperature

In adsorption process temperature plays an important role. The % removal of Chromium and Copper onto the red sandal wood leaf powder at various temperature. The utmost % removal of Chromium and Copper was found to be 82% and 84% at the temperature of 50°C. The % removal at first was increased as the temperature increases and reaches a maximum and after that as the temperature increases the % removal decreases. The decrease in the adsorption as the temperature is due to the increase in the kinetic energy of the adsorbate molecules.

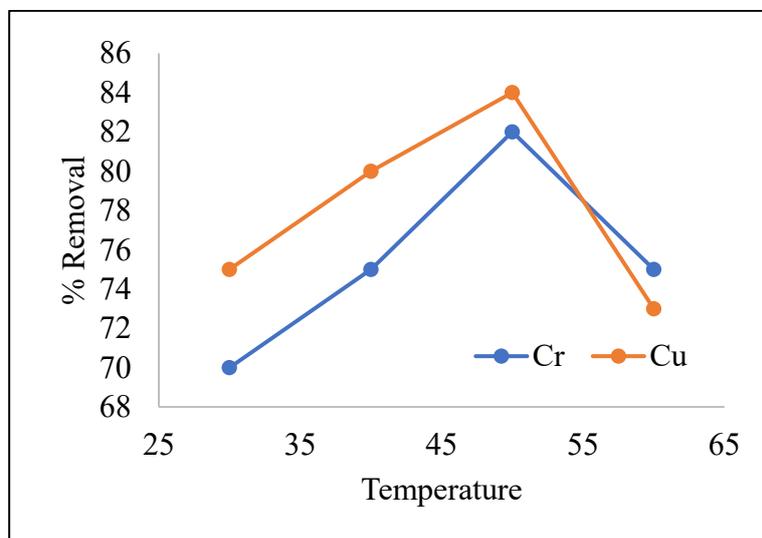


Figure 7: Effect of Temperature on adsorption when the initial concentration of 50mg/L and adsorbent dosage of 1mg.

3.6 Langmuir Isotherm

Langmuir adsorption isotherm is based on monolayer adsorption of metal on the surface of the red sandal wood leaf powder[2]. This model assumes the finite number of active sites evenly distributed across the surface and there is no interaction between the molecules adsorbed because active sites have same affinity for adsorption of monomolecular layer[10].

$$\frac{C_e}{q_e} = \frac{1}{bq_m} + \frac{C_e}{q_m}$$

Where C_e = equilibrium concentration in mg/L

q_e = adsorption capacity in mg/g

q_m = maximum monolayer adsorption capacity in mg/g

b = Langmuir constant in L/mol

$$R_L = 1/(1+b \cdot C_0)$$

The value of the R_L tells us whether the adsorption is favorable or unfavorable. If R_L lies between 0 and 1 is favorable for adsorption. If R_L is > 1 unfavorable for adsorption. $R_L = 1$ then adsorption is linear. $R_L = 0$ irreversible[10].

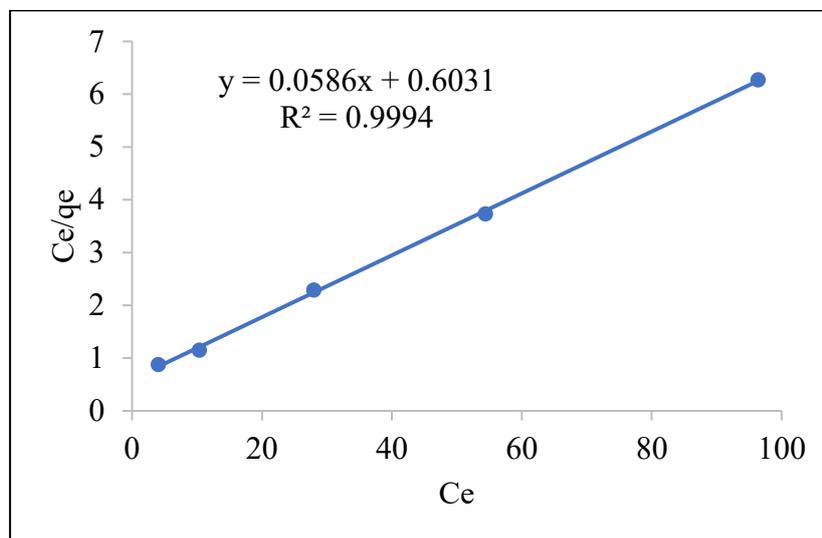


Figure 8: Langmuir isotherm for Chromium

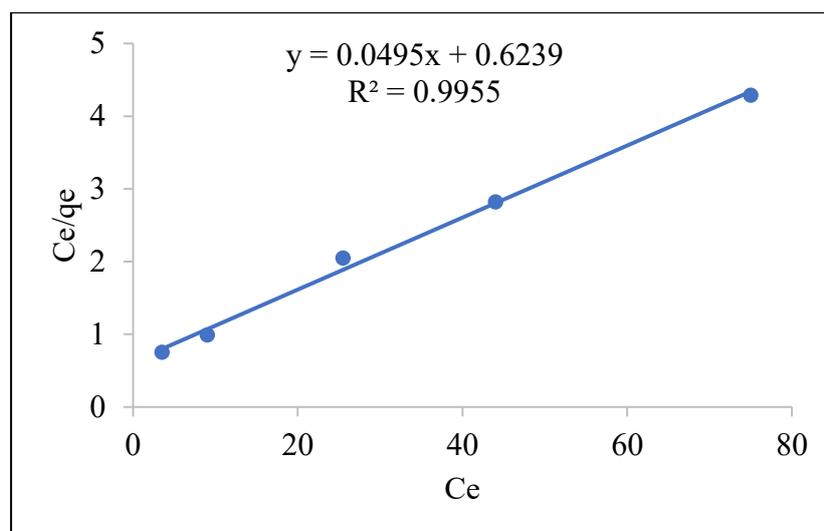


Figure 9: Langmuir isotherm for Copper

3.7 Freundlich Isotherm

Freundlich isotherm is not only for the monolayer adsorption it can also be used for adsorption on heterogenous surface with interaction between the adsorb molecules. This model assumes the adsorbate concentration increase the concentration of adsorbate on adsorbent surface also increases[10].

$$\ln(q_e) = \ln(K_F) + \frac{1}{n} \ln(C_e)$$

C_e = equilibrium concentration in mg/L

q_e = adsorption capacity in mg/g

K_F = Freundlich Constant

$1/n$ = sorption intensity, indicates capacity of adsorbent to the adsorbate.

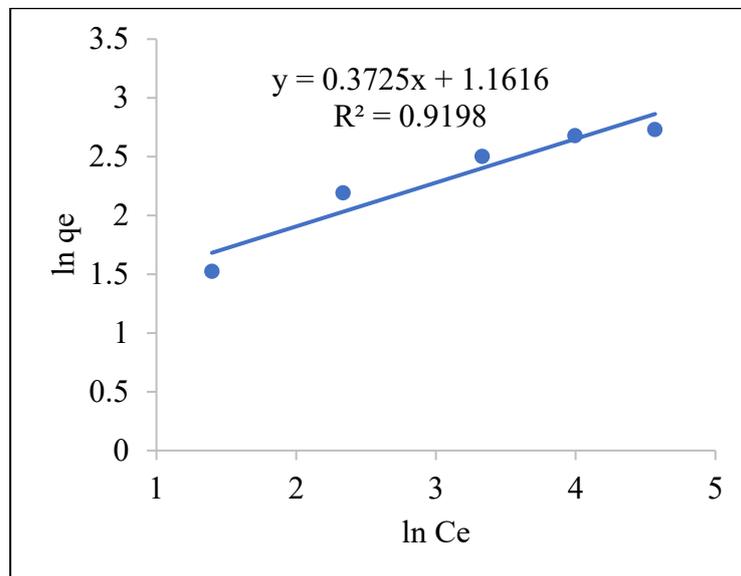


Figure 10: Freundlich isotherm for Chromium

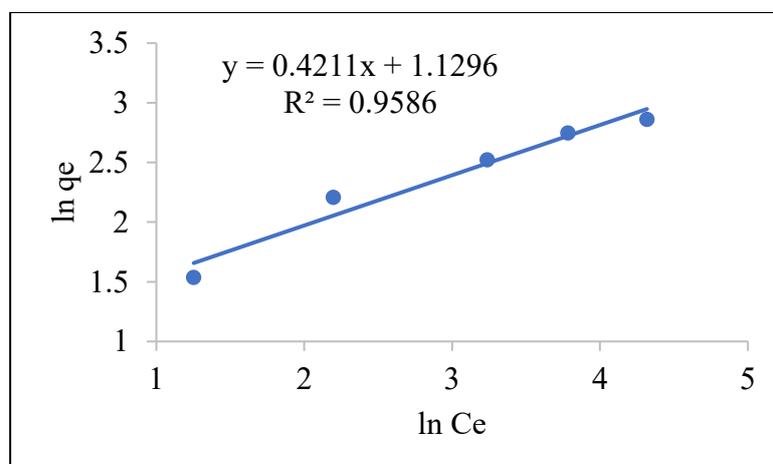


Figure 11: Freundlich isotherm for Copper

Table 1: constants for the adsorption isotherms

		Chromium	Copper
Langmuir	q _m	17.0648	20.202
	b	0.0972	0.0793
	K _L	0.17	0.20
	R ²	0.9994	0.9955
Freundlich	1/n	0.3725	0.4211
	K _F	3.195	3.0944
	R ²	0.9198	0.9586

3.8 Thermodynamic Parameters

Temperature effects the adsorption process. So, this adsorption studies were studied at different temperature ranges. The thermodynamics analysis for the adsorption was performed by using the equation

$$\log\left(\frac{q_e}{C_e}\right) = \frac{\Delta S^0}{2.303 \cdot R} + \frac{\Delta H^0}{2.303 \cdot R \cdot T}$$

$$\Delta G^0 = \Delta H^0 - T \Delta S^0$$

C_e = equilibrium concentration in mg/L

q_e = adsorption capacity in mg/g

R = universal gas constant 8.314 J / mol K

T = Temp in Kelvin

The plot of log(q_e/C_e) vs 1/T will give a straight line the slope will give the enthalpy and the intercept will give the entropy; by using them we can get the Gibbs free energy at various temperatures[2].

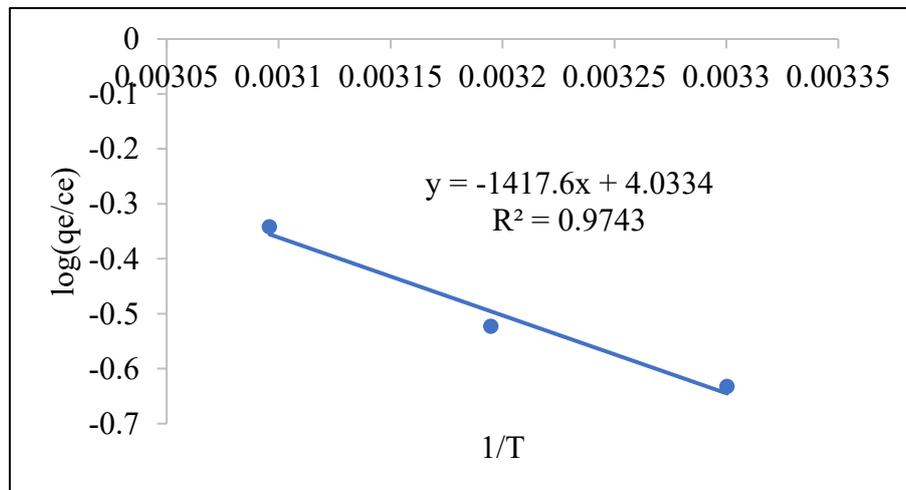


Figure 12: Thermodynamic parameters estimation for Chromium, initial concentration 50mg/L and adsorbent dosage 1mg

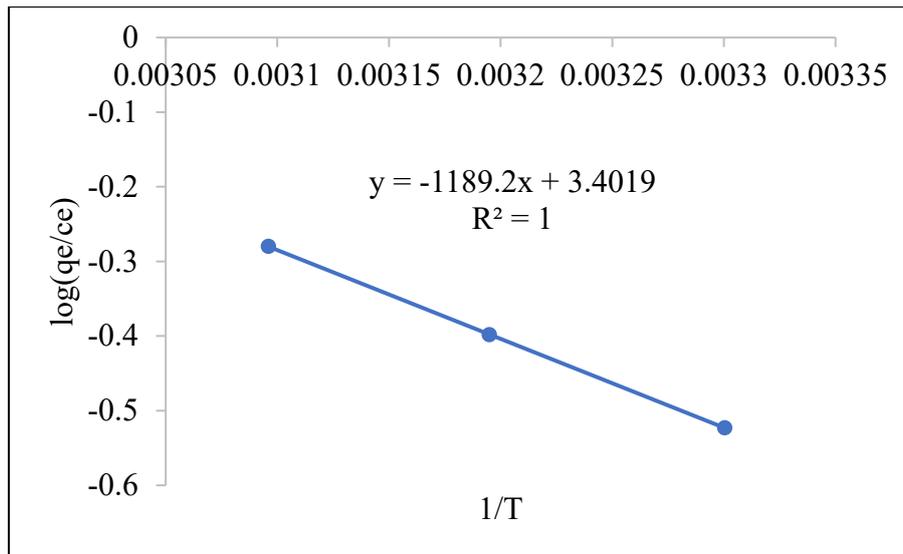


Figure 13: Thermodynamic parameters estimation for Copper, initial concentration 50mg/L and adsorbent dosage 1mg

Table 2: Thermodynamic Properties

Chromium	ΔH KJ/Kmol	-27.143		
	ΔS KJ/Kmol.K	0.0772		
	ΔG KJ/Kmol	303K	313K	323K
		-50.535	-51.307	-52.079
Copper	ΔH KJ/Kmol	-22.77		
	ΔS KJ/Kmol.K	0.0651		
	ΔG KJ/Kmol	303K	313K	323K
		-19.48	-19.43	-19.38

3.9 Kinetic Studies

The process of removal of metals from aqueous phase by using adsorbent can be described by using the kinetic models. Kinetics of removal of metals are explained by using the pseudo first order, pseudo second order models.

3.9.1 Pseudo first order kinetics

$$\frac{dq_t}{dt} = k_{ad}(q_e - q_t)$$

Integrating the equation by using the initial conditions $q_t=0$ at $t=0$

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

Plot of $\log(q_e - q_t)$ vs t will give a straight line for the pseudo first order adsorption kinetics

The parameter $\log q_e$ is found not equal to the intercept of the plot, for the actual first order kinetic model the value of $\log q_e$ must equal to the intercept. So, from this model we can only estimate the k_{ad} [11].

3.9.2 Pseudo second order kinetics

In this model we can estimate both q_e and k_2 , the rate equation is given by

$$\frac{dq_t}{dt} = k_2(q_e - q_t)^2$$

Where k_2 = second order rate constant (g/mg.min)

q_t = amount absorbed at any time t

q_e = amount absorbed at equilibrium time t

on integrating equation by using the boundary conditions $q_t = 0$ to q_t and $t = 0$ to t , the equation becomes

$$\frac{1}{q_e - q_t} = \frac{1}{q_e} + k_2 t$$

This can be written in linear form as

$$\frac{t}{q_t} = \frac{1}{h} + \frac{1}{q_e} t$$

Where $h = k_2 q_e^2$

The plot of t/q_t vs t should give a linear relationship, from that can estimate q_e and k_2 [11].

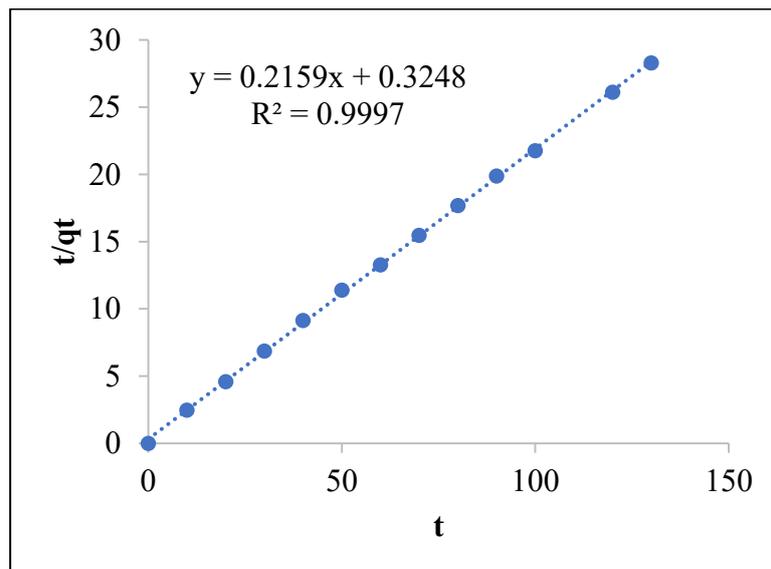


Figure 14: Adsorption kinetics for Chromium initial concentration 50mg/L and adsorbent dosage of 1mg.

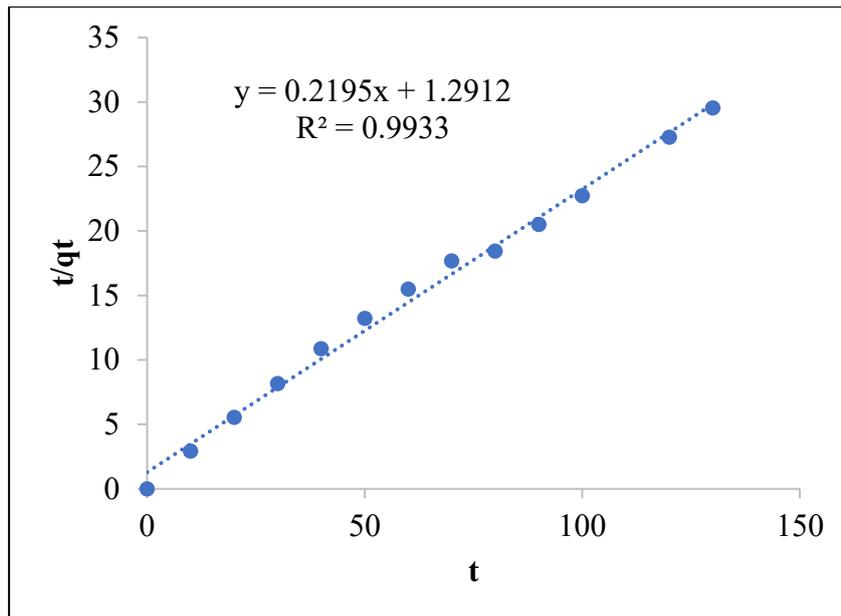


Figure 15: Adsorption kinetics for Copper initial concentration 50mg/L and adsorbent dosage of 1mg.

The pseudo second order kinetics best fit with R^2 values of 0.997, 0.9933 for Chromium and Copper. From the plots the value of q_e and k for chromium and copper are as follows 4.6318, 4.55 and 0.1435 g/mg.min, 0.0374 g/mg.min.

3.9.3 Intraparticle diffusion

For further understanding of the adsorption process of Chromium and copper on the red sandal wood leaf powder, we used the Weber-Morris equation $q_t = kt^{0.5}$. The plot shows that the intraparticle was also present in the adsorption process of Chromium and Copper on to the red sandal wood leaf powder. The linear lines in the plot are not passing through the origin which indicates that the pore diffusion alone is not the rate controlling. In this plot we can see the existing of two regions External mass transfer and the intraparticle diffusion[12].

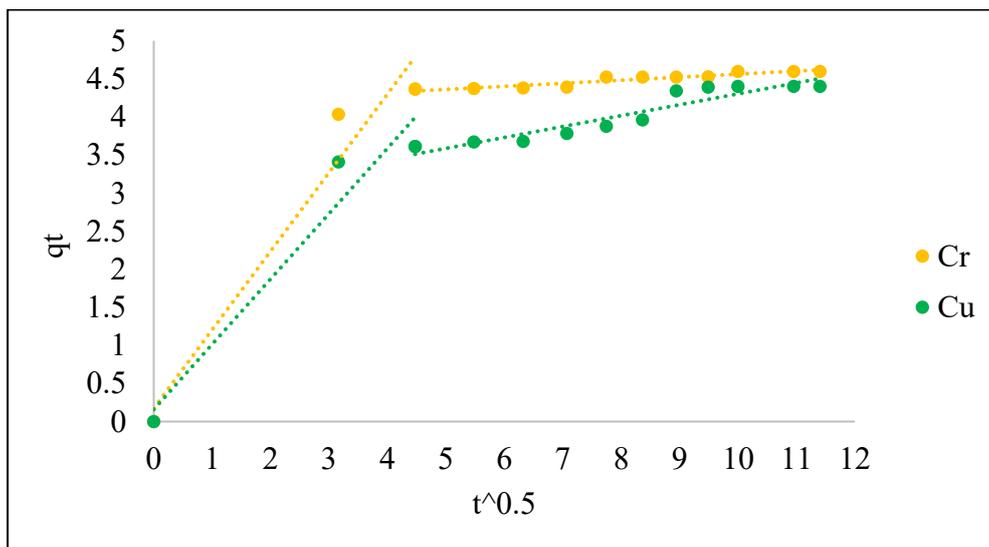


Figure 16: Weber-Morris plot for Chromium and Copper

4. Conclusion

The following conclusions can be drawn from the obtained results:

- Red Sandal wood leaf powder can be used as adsorbent for removal of Chromium and Copper in aqueous solutions.
- Experiments shows that the equilibrium is reached after 100 mins for both the Copper and Chromium.
- Maximum percentage removal of Chromium is 91.91 at 50mg/L initial metal ion concentration.
- Maximum adsorbent capacity for Chromium is 15.3625 at 250mg/L initial metal ion concentration.
- Maximum percentage removal of Copper is 93 at 50mg/L initial metal ion concentration.
- Maximum adsorbent capacity for Copper is 17.5 at 250mg/L initial metal ion concentration.
- Maximum percentage removal of Chromium is 90.44 at pH 2.
- Maximum percentage removal of Copper is 92.4 at pH 6.
- Maximum adsorbent capacity of Chromium is 34 at adsorbent dosage 0.1g.
- Maximum adsorbent capacity of Copper is 32 at adsorbent dosage 0.1g.
- Percentage removal of both Copper and Chromium decreasing with increasing initial metal ion concentration, whereas the adsorption capacity is increasing.
- As the pH increasing the percentage removal of chromium is decreasing, whereas for the copper as the pH increases, percent removal increases up to 6 and then decreases.
- If the adsorbent dosage is increasing then the percent removal is increasing and the adsorbent capacity is decreasing for both Chromium and Copper.
- The Langmuir adsorption isotherm is best set for the adsorption of Chromium and Copper on red sandal wood leaf powder.
- The optimal removal percentage of Chromium and Copper was found to be 82% and 84% at the temperature of 50⁰C.
- The adsorption of Chromium and Copper on red sandal wood leaf powder follows the pseudo second order kinetics and also studied the intraparticle diffusion.

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