Research Article

Removal of Chromium (VI) ions from Aqueous Solution using Tamarind Seeds as an Adsorbent

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ABSTRACT

Adsorption is one of the effective techniques for chromium (VI) removal from waste water. The present work deals with the determination of Cr(VI) removal capacity from aqueous solution using tamarind seeds which is a low cost adsorbent. The effect of various parameters such as pH, contact time, adsorbent amount, initial Cr(VI) concentration and temperature for the adsorption of Cr(VI) on tamarind seeds is investigated. The maximum adsorption of Cr(VI) on tamarind seeds is obtained at pH 2. The equilibrium time obtained is 240 min for Cr(VI) adsorption on tamarind seeds. The experimental equilibrium adsorption data are tested for the Langmuir, Freundlich and Temkin equations. The Langmuir isotherm model is found to be most suitable for the Cr(VI) adsorption using tamarind seeds and also confirms the monolayer adsorption of Cr(VI) onto the adsorbent surface. The maximum adsorption capacity obtained using the Langmuir isotherm model is 29.41 mg/g at pH 2. Results indicate the following order to fit the isotherms: Langmuir > Freundlich> Temkin. Adsorption kinetic data were modeled using the Pseudo first order, Pseudo second order and Elovich models. The results indicate that the second order model best describes adsorption kinetic data. Experimental results demonstrate that the adsorbent has a significant capacity for adsorption of Cr(VI) from wastewater streams.

Keywords: Adsorption, Chromium (VI), Tamarind seeds, Isotherms, Kinetics.

1. INTRODUCTION

Chromium exists usually in both trivalent and hexavalent forms in aqueous systems. Cr(III) is relatively insoluble and required by microorganisms in small quantities as an essential trace metal nutrient, while Cr(VI) is a great concern because of its toxicity. Cr(VI) has been reported to be a primary contaminant to animals, plants and microorganisms and it is known to be carcinogenic. Sources of chromium waste leading to water pollution includes electroplating, steel fabrication, paints and pigments, mining, leather tanning, textile dyeing, aluminum conversion coating operations, plants producing industrial inorganic chemicals and wood treatment units[1].

Due to environmental concern, discharge limits of both Cr(III) and Cr(VI) have been instituted by most industrial countries. Their concentration in industrial wastewaters ranges from 0.5 to 270 mg/L. The tolerance limit for Cr(VI) for discharge into inland surface waters is 0.1 mg/L and in potable water is 0.05 mg/L. The Ministry of Environment and Forest (MOEE); Government of India has set minimal national standards (MINAS) of 0.1 mg/L for safe discharge of effluent containing Cr(VI) in surface water[2].

In general Chromium (VI) is removed from waste water by various methods such as chemical precipitation, electrochemical reduction, sulfide precipitation, cementation, ion-exchange, reverse osmosis, electro dialysis, solvent extraction, and evaporation, etc. These methods have been found to be limited, since they often involve high capital and operational costs. Among these methods adsorption is the most efficient technique because of its convenience, easy operation and simplicity of design[9]. Adsorption which have proven to be effective for the removal of Cr(VI) from aqueous solutions and industrial effluents. Agricultural by-products have been widely studied for metal removal from waste water. These include peat, wood, pine bark, banana pith, soybean and cotton seed hulls, rice husk, saw dust, tamarind seeds, compost and leaves[8].

In the present study, activated tamarind seeds are used as a low-cost adsorbent for the removal of Cr(VI) from aqueous solutions. It is one of the biological waste products which is widely available in all over the world and can be easily cultivated in variety of soils. The influence of pH, contact time, metal ions and adsorbent concentrations were investigated and experimental data obtained were evaluated and fitted using adsorbent equilibrium isotherms, and kinetic models.

2. MATERIALS AND METHODS
2.1. Materials
Potassium dichromate (A.R) was used for the preparation of stock Cr (VI) solution in distilled water. For pH adjustments throughout the experiment hydrochloric acid and sodium hydroxide solutions were used. Concentrated sulfuric acid (98% w/w) was used for the treatment of Tamarind seeds.

2.2. Adsorbent preparation
Tamarind seeds are collected from local market and washed with distilled water and sun dried for three days. The dried seeds are crushed in small particles by using Roll crusher. Then it is treated with concentrated sulphuric acid (98% W/W) in 1:1 weight ratio and is kept in oven at 105^0 C for 24 hours. Treated seeds are washed with distilled water to remove free acid and again dried at 110^0 C for 4 hours. Dried material was ground and sieved to obtain particles of size 0.3 mm.

2.3. Batch experiments
All experiments were carried out at room temperature, i.e., 30±2 ^{0}C. Using 100 ml of test solution batch adsorption studies were carried out at the desired pH value, contact time and adsorbent dosage level. Different initial concentration of Cr(VI) solutions was prepared by proper dilution from stock 1000 ppm Cr(VI) standard. pH of the solution was monitored by adding 1M HCl and 1M NaOH solution as required. The solution was stirred by using mechanical stirrer. The time required for reaching the equilibrium condition estimated by drawing samples at regular intervals of time till equilibrium was reached. The contents of the flask were filtered through filter paper and the filtrate was analyzed for remaining Cr(VI) concentration in the sample using UV visible spectrophotometer with 1,5-diphenylcarbazide in acid medium by following APHA, AWWA standard methods for examination of water and wastewater[7]. The absorbance of purple-violet colored solution is measured at a wavelength of 540 nm. The amount of Cr(VI) adsorbed per unit mass of the adsorbent was evaluated by using the following mass balance equation,

$q = [(C_i - C_e) \times V]/W$

where V is the volume of the solution (L) and W, the amount of adsorbent (g); C_i and C_e (mg/L), the initial and equilibrium metal concentrations.

The percent removal of Cr(VI) was calculated as follows:

$\%\text{Removal of Cr(VI)} = [(C_{initial} - C_{final}) / C_{initial}] \times 100$

Adsorption experiments for the effect of pH were conducted by using a solution having 200 mg/L of Cr(VI) concentration with an adsorbent dosage of 10 g/L and stirring the same for a contact time of 3h. The effect of adsorbent dosage level on percent removal of chromium was studied using Cr(VI) concentration of 200 mg/L, pH 2 and adsorbent concentration ranging from 2 g/L to 10 g/L. Effect on variation of initial concentration was studied using Cr(VI) solution of concentration 200 mg/L to 700 mg/L at pH 2, contact time of 4 h and adsorbent dosage level of 10 g/L.

Adsorption isotherm studies were carried out with six different initial concentrations of Cr(VI) from 200 mg/L to 700 mg/L at pH 2, contact time 4 h and adsorbent dosage level of 10g/L. The kinetic parameters for the adsorption process were studied on the batch adsorption of 200 mg/L of Cr(VI) at pH 2. The contact time was varied from 30 min to 240 min and the percent removal of Cr(VI) was monitored during the study.

3. RESULTS AND DISCUSSION
3.1. Effect of pH
The pH of the solution has a significant impact on the uptake of heavy metals, since it determines the surface charge of the adsorbent, the degree of ionization and speciation of the adsorbate. In order to establish the effect of pH on the bio-sorption of chromium (VI) ions, the batch equilibrium studies at different pH values were carried out in the range of 2-10. Fig. 1 shows that the maximum percent removal of chromium (VI) ions on the adsorbent was observed at pH 2 and significantly decreased at higher pH values.

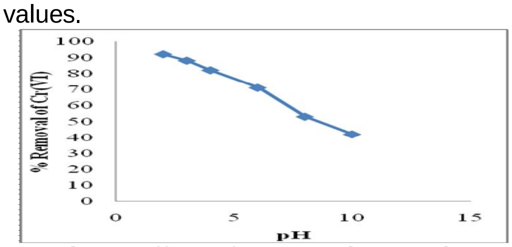

Fig. 1: Effect of contact time on the adsorption of Cr(VI) by tamarind seeds. Initial concentration 200 mg/L, adsorbent dosage 10 g/L, contact time 3h

The dominant form of Cr(VI) at pH 2 is $HCrO_4^-$ which arises from the hydrolysis reaction of the dichromate ion ($Cr_2O_7^{2-}$) Increasing the pH will shift the concentration of $HCrO^{4-}$ to $Cr_2O_7^{2-}$ and other forms as CrO^{4-}. Maximum adsorption at pH 2 and 3 indicates that it was the $HCrO^{4-}$ form of Cr (VI), which was the predominant species at this pH range and adsorbed preferentially on the adsorbents. Better adsorption capacity observed at low pH values with optimum range of pH between 2 and 3 may be attributed to the large number of H^+ ions present at these pH values, which in turn neutralize the negatively charged hydroxyl group (OH^-) on adsorbed surface thereby reducing the hindrance to the diffusion of dichromate ions. At higher pH values, the reduction in adsorption may be possible due to abundance of OH^- ions causing increased hindrance to diffusion of dichromate ions[1].

3.2. Effect of contact time

The experimental runs measuring the effect of contact time on the batch adsorption of Cr(VI) at initial pH value 2 and initial Cr(VI) concentration of 200 mg/L is shown in Fig. 2. The removal of chromium ions increases with time and attains saturation in about 240 min, basically the removal of sorbate is rapid, but it gradually decreases with time until it reaches equilibrium. The rate of percent metal removal is higher in the beginning due to a larger surface area of the adsorbent being available for the adsorption of metal. The two stage sorption mechanism with the first rapid and quantitatively predominant and the slower and the quantitatively insignificant[21].

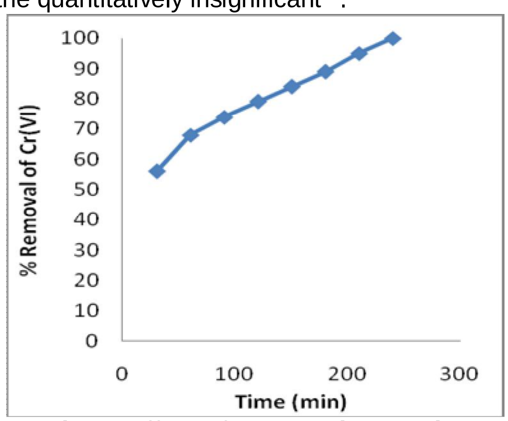

Fig. 2: Effect of contact time on the adsorption of Cr(VI) by tamarind seeds. pH 2, initial concentration 200 mg/L, adsorbent dosage 10 g/L.

3.3. Effect of initial metal ion concentration

The efficiency of Cr(VI) removal was affected by the initial metal ion concentration, with decreasing removal percentages as concentration increases from 200 mg/L to 700 mg/L at constant pH 2, adsorbent dosage level 10 g/L and contact time of 4 h duration as shown in Fig. 3. At low concentrations, metals are adsorbed by specific sites, while with increasing metal concentrations the specific sites are saturated and the exchange sites are filled[21].

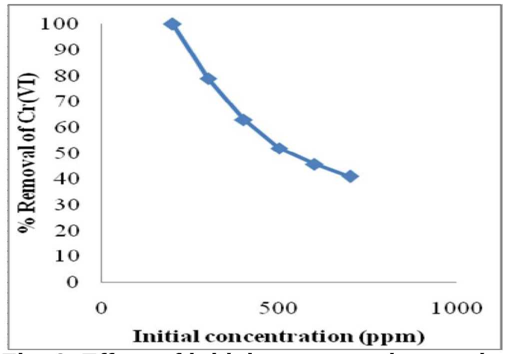

Fig. 3. Effect of initial concentration on the adsorption of Cr(VI) by tamarind seeds. pH 2, contact time 4h, adsorbent dosage 10 g/L.

3.4. Effect of adsorbent concentration

One of the parameters that strongly affect the sorption capacity is the concentration of the adsorbent. With the fixed metal concentration it can easily be inferred that the percentage removal of metal ions increases with increasing weight of adsorbent as shown in Fig. 4. After certain adsorbent dosage the removal efficiency is not increased so significantly. It is evident that maximum removal efficiency was achieved at an adsorbent dosage level of 10 g/L. Therefore, the following experiments were carried out at adsorbent concentration of 10 g/L.

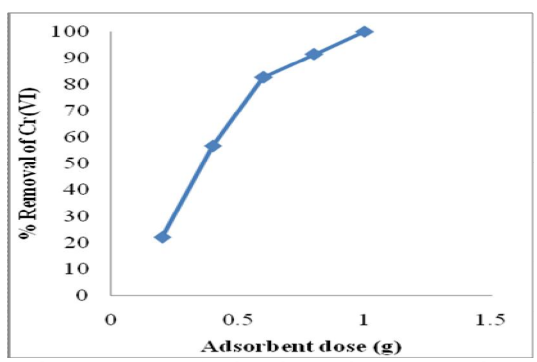

Fig. 3: Effect of adsorbent dosage on the adsorption of Cr(VI) by tamarind seeds. pH 2, contact time 4h, initial concentration 200 mg/L.

3.5. Effect of temperature
Temperature dependence of the adsorption process is associated with several thermodynamic parameters. Fig. 5 shows an increasing trend of Cr(VI) removal with the rise in temperature from 30^0C to 50^0C. The reason may be that, at high temperature some polymers might have released from the sorbent which assist in adsorption, because the tamarind seed contains some free sugars. The increase in Cr(VI) uptake may also be due to creation of some new sorption sites on the sorbent surface or the increased rate of intra-particle diffusion of sorbate ions into the pores of adsorbent at higher temperature, as diffusion is an endothermic process. Up to certain extent, enhancement of adsorption capacity of tamarind seed at higher temperatures may be attributed to enlargement of pore size and/ or activation of the adsorbent surface.

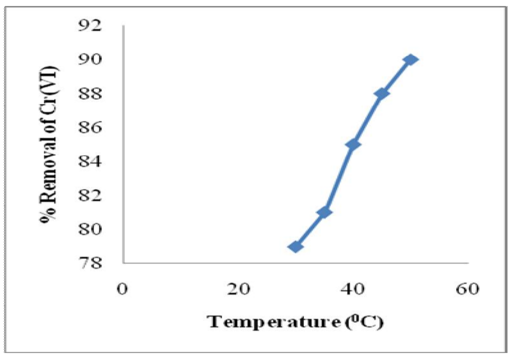

Fig. 5:. Effect of temperature on the adsorption of Cr(VI) by tamarind seeds. pH 2, contact time 4h, initial concentration 200 mg/L, adsorbent dosage 10 g/L.

3.6. Adsorption isotherms
The adsorption isotherms for the Cr(VI) removal were studied using initial concentration of Cr(VI) from 200 mg/L to 700 mg/L at an adsorbent dosage level of 10 g/L. The adsorption equilibrium data are conveniently represented by adsorption isotherms, which correspond to the relationship between the mass of the solute adsorbed per unit mass of adsorbent q_e and the solute concentration for the solution at equilibrium C_e.
In order to successively represent the equilibrium adsorptive behavior, it is important to have a satisfactory description of the equation state between the two phases composing the adsorption system. Three kinds of several isotherms equations were tested to fit the experimental data[10].
Langmuir equation: $C_e/q_e = (C_e/q_{max}) + [1/ (q_{max} b)]$

Freundlich equation: $\log q_e = \log K + (1/n) \log C_e$
Temkin equation: $q_e = a + b \ln C_e$
Where q_e is the amount adsorbed at equilibrium (mg/g) and C_e is the equilibrium concentration of metal ions in solution (mg/L).
The other parameters are different isotherm constants, which can be determined by regression of the experimental data. In the Langmuir equation, q_{max} (mg/g) is the measure of adsorption capacity under the experimental conditions and b is a constant related to the energy of adsorption. Freundlich treatment gives the parameters, n, indicative of bond energies between metal ion and the adsorbent and K, related to bond strength.

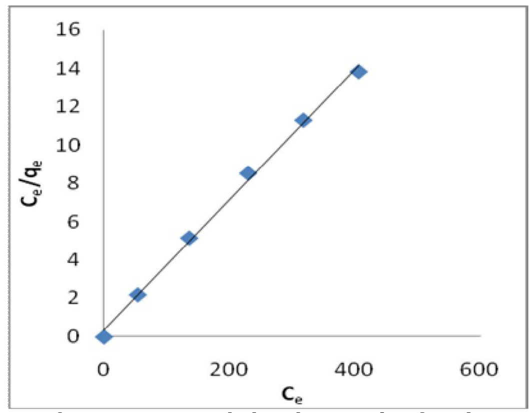

Fig . 6: Langmuir isotherm plot for the adsorption of Cr(VI) by tamarind seeds. pH 2, adsorbent dosage 10 g/L.

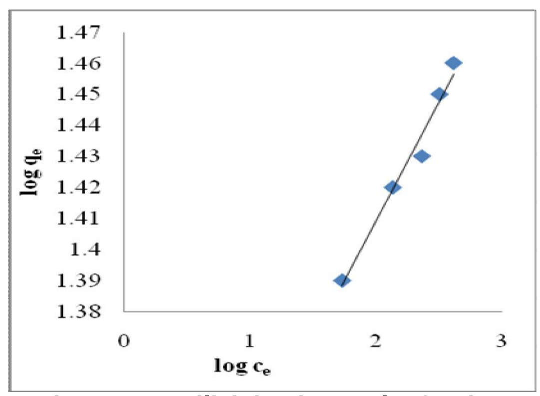

Fig. 7: Freundlich isotherm plot for the adsorption of Cr(VI) by tamarind seeds. pH 2, adsorbent dosage 10 g/L.

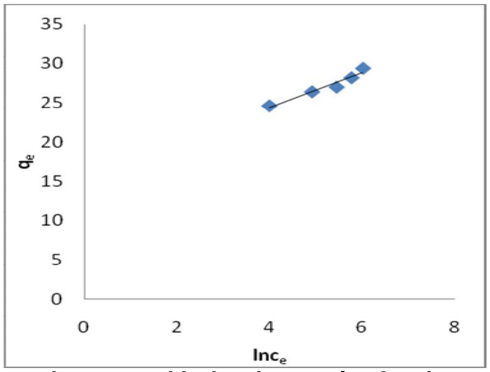

Fig. 8: Temkin isotherm plot for the adsorption of Cr(VI) by tamarind seeds. pH 2, adsorbent dosage 10 g/L.

The linearized Langmuir, Freundlich and Temkin isotherms of chromium (VI) is shown in Fig. 6-9. The estimated model parameters with correlation coefficient (R^2) for different models are shown in Table. 1. The values of R^2 are regarded as a measure of the goodness of fit of experimental data on the isotherm models. Applicability of the three isotherm models for the present data approximately follows the order: Langmuir> Temkin> Freundlich.

Table 1: Langmuir, Freundlich and Temkin adsorption isotherm constants for adsorption of Cr(VI) onto Tamarind seeds

Adsorbent	Langmuir Isotherm			Freundlich Isotherm			Temkin Isotherm		
	q_{max} (mg/g)	b (L/mg)	R^2	K_F	n	R^2	a	b	R^2
Tamarind seeds	29.41	0.104	0.997	17.95	12.99	0.976	15.59	2.207	0.946

3.7. Adsorption kinetics

In order to analyze the adsorption kinetics of heavy metal ions, the pseudo-first and pseudo second order and simple Elovich kinetic models were applied to data. The first order rate equation of Lagergren is most widely used for the sorption of a solute from liquid solution [15] and is represented as:

$\ln(q_e - q_t) = \ln q_e – K_1 t$

where q_e is the mass of metal adsorbed at equilibrium (mg/g), q_t is the mass of metal adsorbed at time t (mg/g), K_1 is the first order reaction rate constant (min^{-1}). The pseudo first order considers the rate of occupation of adsorption sites to be proportional to the number of unoccupied sites. Pseudo second order equation based on adsorption equilibrium capacity may be expressed in the form:

$t/q_t = [1/(K_2 q_e^2)] + (t/q_e)$

where K_2 is the second order reaction rate equilibrium constant (g/mg. min). A plot of t/q_t against t should give a linear relationship for the applicability of the second order kinetic. Finally, the simple Elovich may be expressed in form:

$q_t = \alpha + \beta \ln t$

A plot of q_t versus ln t should give a linear relationship for the applicability of the simple Elovich kinetic.

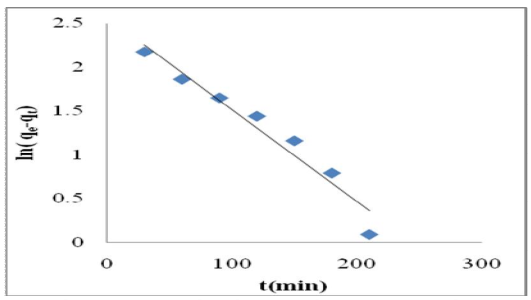

Fig. 9: Pseudo first order plot for the adsorption of Cr(VI) by tamarind seeds. pH 2, initial concentration 200 mg/L, adsorbent dose 10 g/L.

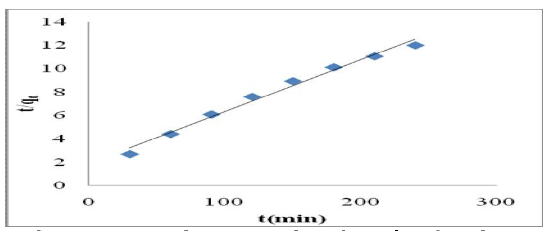

Fig. 10: Pseudo second order plot for the adsorption of Cr(VI) by tamarind seeds. pH 2, initial concentration 200 mg/L, adsorbent dose 10 g/L.

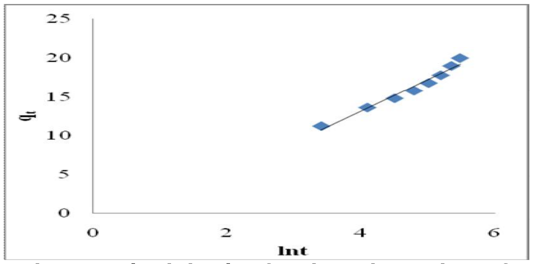

Fig. 11: Elovich plot for the adsorption of (VI) by tamarind seeds. pH 2, initial concentration 200 mg/L, adsorbent dose 10 g/L.

During the present study, the three different kinetic models were applied and showed in Fig. 9-11. The estimated model and the related statistic parameters are reported in Table. 1. The values of correlation coefficients indicated a better fit of pseudo- 2nd-order model with the experimental data.

Table 2: Calculated kinetic parameters for pseudo first-order, second order and Elovich kinetic models for the adsorption of Cr(VI) using tamarind seeds as an adsorbent

Pseudo first order			Pseudo second order			Elovich model		
K_1 (min^{-1})	q_e (mg/g)	R^2	K_2 ×10^{-3} (g mg^{-1} min^{-1})	q_e (mg/g)	R^2	α	β	R^2
0.010	13.13	0.950	1.039	22.73	0.986	3.069	4.061	0.97

4. CONCLUSIONS

Batch adsorption studies for the removal of Cr(VI) from aqueous solution have been carried out using activated tamarind seeds. The obtained results may be summarized as follows:

- The pH variation studies showed that the adsorption process is highly pH dependent .The optimum pH range for the removal was found to be 2.
- The effect of various process parameters showed that percentage of adsorption decreased with increase in initial metal ion concentration, whereas with increase in contact time, adsorbent dose and temperature, there was increase in percentage adsorption or removal of metal ions. Maximum uptake of Cr(VI) was obtained at adsorbent dosage of 10 g/L. Increase in adsorbent dosage leads to increase in Cr(VI) adsorption due to increased number of adsorption sites.
- The equilibrium data are best fitted with Langmuir Isotherm model which confirms the monolayer adsorption of Cr(VI) onto tamarind seeds. The maximum adsorption capacity is obtained with the application of Langmuir Isotherm model as 29.41 mg/g, which is comparatively good adsorption capacity. Results indicate the following order to fit the isotherms: Langmuir > Freundlich> Temkin.
- The kinetics for adsorption of Cr(VI) by tamarind seeds was best described by Pseudo second order model.
- The use of tamarind seed as an adsorbent seems to be an economical and worthwhile alternative over conventional methods.
- Finally, this works shows that activated tamarind seeds can be used as efficient adsorbent for Cr(VI) removal from waste water. However, further research should attempt to improve the adsorption capacity of tamarind seeds and apply this method to the removal of metals in large scale.

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