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PHYSICOCHEMICAL STUDY AND REMOVAL OF CHROMIUM FROM TANNERY EFFLUENT BY LOW COST BIOADSORBENT

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Keywords

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Abstract

In the present study, we used various plant parts (barks and leaves) to utilize the feasibility of removing chromium from tannery effluents. In batch experiments, some physical parameters (like adsorbent dose and contact time) that primarily influence adsorption were studied at a fixed pH of 3. The present evaluated results (i.e., equilibrium adsorption) are well supported by Freundlich plus Langmuir's model. Here, the highest monolayer adsorption capacities (qe) were found to be 19.23, 18.98, 16.78, 15.27, and 18.66 mg/g for Azadirachta indica leaf (AI), *Platanus orientalis* (PO), *Tamarind indica* bark (TI), *Mangifera indica* bark (MI), and *Eucalyptus obliqua* plant bark (Eu) respectively. However, at optimum conditions (12-hour contact period, 15g dose), the adsorption capacity was measured 14.48, 12.61, 14.6, 12.76, and 14.46 mg/g by AI, PO, TI, MI, and Eu, respectively. Under this operating condition, total dissolved solids (TDS) (81.28 to 94.40 %), conductivity (82.26 to 94.61 %), and turbidity (80.47 to 93.95 %) of effluent were also significantly reduced. All of this bio-adsorbent could be used to heal tannery sewages/effluents and other domestic wastewater.

1. Introduction

Pollution is a major concern today due to its hazardous health impacts. However, previous records indicate that industrial wastes act as a major source of pollutants, and to reduce recent pollution, on-site management is a prime need before discharging these into the sewage system Islam, M., Rahaman, A., Mahdi, M.M. & Hossain, M., (2022). The increasing industrialization and manufacturing process initiate and facilitate both direct plus indirect unfavorable results on the entire environment. Rampant pollution becomes a common feature with rising effluents, contaminants, and toxins produced from unbalanced industrialization. Currently, Bangladesh has approximately 30000 manufacturing companies. However, only 10% of these units emit treated effluents or waste. The remaining 90% release effluent and other contaminants into the river channel without any treatment or minimal treatment. The result is continuous contamination of adjacent channels, agricultural lands, irrigation channels, surface/groundwater, and surrounding aquatic environments. Later, easily initiated into the food cycle and contaminated the cycle at a magnifying rate Islam, M., Rahaman, A., Mahdi, M.M.& Hossain, M., (2022) Younas, F., Niazi, N.K., Bibi, I., Afzal, M., Hussain, K., Shahid, M., Aslam, Z., Bashir, S., Hussain, M.M. and Bundschuh, J., (2022). Tannery industries ranked

1st position for emitting the highest pollutants during the different manufacturing processes. There are approximately 200 tanning industries in Bangladesh, most of which are located in the Hazaribagh area of Dhaka. Around 90% of these industries are connected to different chrome tanning processes where they use toxic chromium sulfate, acids, chemicals, and salts Islam, M., Rahaman, A., Mahdi, M.M. and Hossain, M., (2022). The tannery generates different types of toxins, and these are well recognized for producing excess hazardous chemicals, negative impact on salinity, organic load (biological and chemical oxygen demand or load), inorganic matter (heavy metals), dissolved, suspended solids, etc. Finally, enhance severe environmental and health threat by destroying the food chain Younas, F., Niazi, N.K., Bibi, I., Afzal, M., Hussain, K., Shahid, M., Aslam, Z., Bashir, S., Hussain, M.M. and Bundschuh, J., (2022). Maritim, G.K., Kipkemboi, P. and Lusweti, K., (2022). In every step of tanning huge amount of water is used; among the used water, \sim 90- 92% is released as effluent Maritim, G.K., Kipkemboi, P. and Lusweti, K., (2022). Silambarasan, T.S., Balakumaran, M.D., Suresh, S., Balasubramanian, V., Sanjivkumar, M., Sendilkumar, B. and Dhandapani, R., (2022). The permitted limit of hexavalent and trivalent chromium (Cr) in wastewater is respectively 0.5 mg/l and five (5) mg/l. In the case of edible water tolerable limit of chromium is 0.1 mg/l US Department of Health and Human Services, (1991). Chromium in drinking-water. Background document for development of WHO Chromium used in the tanning industry because of chrometanned weather is characterized by high hydrothermal stability and excellent physical properties. In the tanning process, up to 60-70% of chromium is bonded with rawhide, and skin and wastewater contain a large amount of unused chromium. Chrome tanning wastewater generally contains 1500- 3000 mg/l of trivalent chromium Islam, M., Rahaman, A., Mahdi, M.M. & Hossain, M., (2022), Dave, H., Vajpayee, M. and Ledwani, L., 2022. Trivalent chromium acts as a major catalyst in glucose, lipid, and amino acid metabolism, but prolonged exposure to chromium causes serious health problems Islam, M., Rahaman, A., Mahdi, M.M. and Hossain, M., (2022). Longtime chromium exposure causes respiratory tract problems, lung cancer, skin disorders, acute tubular and kidney damage, gastric damage, and finally, death in severe cases. Islam, M.S. and M. Hoque, (2014). There are several methodologies have been used in removing chromium from different effluents, like precipitation, electroplating, ion exchange, bioaccumulation, Nano filtration, chemical reduction, etc. Jain, M., Garg, V.K. and Kadirvelu, K., (2009). These processes suffer from various disadvantages like incomplete removal, production of secondary sludge, costly, high reagent cost, and energy requirements. Researchers around the world try to create an easy, efficient, simple, low-cost methodology to release various trace elements from industrial wastewater. In this study, we try to develop bio-adsorbents derived from Lingo cellulose materials such as plant leaves and bark. Many bio-adsorbents have been used in the past, like maple sawdust. Kobya, M., (2004). neem sawdust, coconut fiber. Lata, *et al* (2007). eucalyptus bark Freundlich, H., (1906). The bark of several pine species Malik, *et al*, (2007). soya coke Malkoc et al (2005). green algae Malkoc, E. & Y. Nuhoglu, The removal of chromium (VI)tea waste Maritim, *et al.,* (2022, plum tree bark McKay, G., (1998). and Chitosan. This work aims to evaluate some low-cost adsorbents from agricultural biomaterials such as plant leaf and bark to remove chromium from real tannery wastewater and also measure some physical parameters that are reduced by the adsorption with these bio-adsorbents.

2.Experimental methodology

2.1 Preparation of bio-adsorbent

Locally available plant bark and leaf samples were collected separately and then thoroughly rinsed with de-ionized water until the removal of different unwanted particles (i.e., sand, dirt, mud). After proper cleaning, the samples were dried at 60° C \pm 5 in an oven for 24 hours. Then these were grounded properly and strained to 120 mesh size. After sieving, obtained products were stored separately in airtight plastic containers. This powdered bioadsorbent was used for an adsorption study without any pretreatment.

Name	Representation			
Eucalyptus plant bark	Eu			
Tamarind indicia bark	ТI			
Mangifera indica bark	МĪ			
Azadirachta indica leaf	AI			
Platanus orientalis leaf	PO.			

Table 1: Description of the studied plants collected near the Hazaribagh industrial area.

2.2 Collection of Tannery effluent

Chrome tanning effluents were collected in plastic bottles from the Samina, Choudhury, and Ruma

tannery industries from Hazaribagh. Before sampling, the plastic bottles were treated with 0.5M HNO³ acid and stored at 4º C till analysis.

Table 2: Initial metal concentration, TDS, Turbidity and conductivity of the studied tannery effluents.

Sample ID	Cr concentration		TDS(gm/l)		Turbidity
	(ppm)			(mS/cm)	(NTU)
Chowdhury	1524.3		593	117.8	359
Samina	1291		57.2	114.8	297
Ruma	1100.3		53.2	105.8	281

2.3 Batch Adsorption Experiment

We conducted several batch experiments to assess the influence of adsorbent dose and contact time on chromium (Cr) release from industrial waste. All the experiments were conducted by adding 5, 10, 15, and 20 gm of adsorbents in 250 ml chromium solution (tannery effluent) and then keeping the solution inside a rotary shaker at $30\pm1\degree C$ temperature. The solutions were extracted from the rotary shaker at programmed periods (i.e., 3, 6, 9, and 12-hour intervals). Later the solution was kept at a centrifuge at 5000 rpm for 10-12 minutes to separate the adsorbent. Various tanneries effluents were used to

determine the effects of opening Cr ion concentration on the elimination of Cr from the sewages and the influence of physical parameters on the release of Cr from sewage. The concentration of chromium varies from tannery to tannery due to the unique tanning process. However, in the tanning process, pH is maintained between 2.5 to 3.0 for the proper penetration of chromium in the rawhide and skin during chrome tanning operation Mella, B., A.C. Glanert, and M. Gutterres, (2015). Therefore, all of the experiments were conducted at that pH range by adding 1M HCl or 1M NaOH. Chromium concentration was then analyzed by Flame Atomic

Adsorption Spectrophotometer, followed by wet digestion. Isotherm experiments were carried out by shaking various amounts of adsorbent (i.e., 5 - 30 grams) in 250 ml at varying concentrations of tannery waste of Cr solution for a constant time interval (12 hours) for each bio-adsorbent. There were three replications for each experiment, and the data analysis was carried out based on mean results. The total Cr adsorbed by used adsorbent crushed powder was determined as follows:

%Removal =
$$
\frac{C_{o} - C_{c}}{C_{o}} \times 100
$$

$$
Q_{\text{max}} = \frac{C_{o} - C_{e}}{M} \times V
$$

M

Where, Q_{max} = Adsorption capacity (Cr ion absorbed/ g of adsorbent), C_0 = initial concentration, C_e = final concentration, M = mass of the adsorbent (gm) and $V =$ volume of Cr solution (litter) taken for the adsorption study.

2.4 Digestion of Water Sample

Digestion was conducted in a microwave digester (CEM, MARS Xpress, USA; Model-907511) based on manufacturer guidelines. Firstly, samples were properly cleaned and chopped into minute sizes. Then 5ml sample was poured into a clean Teflon vessel. Pre-digestion was carried out by standing open the Teflon vessel for 15-20 minutes before sealing. Later digestion was carried out with the addition of dilute $HNO₃$ (12 ml $HNO₃$, 69%, Merck, Germany) plus H_2O_2 (4 ml) and kept at 200°C for 15 minutes with a constant power supply (1600W) in a CEM microwave. Then de-ionized water was added until it reached a constant volume (50 ml) of the entire solution. By using a 0.45 µm syringe, filter digested samples were filtered and then, until analysis, stored at 4° C in polypropylene tubes (50 ml) [21-22].

2.5 Analysis of the sample solution

The concentration of the chromium was measured by a flame atomic absorption spectrometer (Perkin-Elmer, Model- 800, USA) operated with a Zeeman background corrector. An atomizer with an acetylene/air burner was utilized for analyzing the elements. A hollow Cathode Lamp with 357.9 nm wavelengths was used for Cr determination [22]. Results were summarized and represented in mg/l unit. For the analysis of solutions extracted, an aliquot of 100 µl of the sample was kept manually to the sample injector of a spectrometer connected with a nebulizer. The lower detection limit (LoD) for Cr metal was 0.08µg/L in FAAS [23].

2.6 Adsorption isotherm studies

Equilibrium isotherms have been carried out to get an enhanced insight into different adsorption systems. Among many other theories regarding adsorption equilibrium, "three isotherm equations," specifically Freundlich and Langmuir, have been trialed in the recent study due to their high acceptability. The Langmuir adsorption model explains a homogeneous surface and assumes that every adsorption sites possess the same activity. Thus, the adsorption site has no effect on adsorption from any specific angle/adjacent site. An additional hypothesis is that only a monolayer is formed during maximum adsorption, and the adsorption takes place through a similar mechanism [24]. This model may be represented as follows:

$$
\frac{1}{q_e} = \frac{1}{K_L q_o} \cdot \frac{1}{C_e} + \frac{1}{q_o}
$$
 (3)

Here, $Ce = equilibrium concentration of Cr$ ion (mg/L) ; qe = quantity of Cr adsorbed during equilibrium (mg/g) ; qo = highest adsorption capability/capacity (mg/g), and $KL = constant$ (L/mg) relay on adsorption's energy. From the slope and intercept of the plot 1/qe against 1/Ce, the constant (KL) and the highest adsorption capability (qo) is obtained. The Freundlich model describes the equilibrium based on heterogeneous surfaces without assuming monolayer capacity [25, 26]. The presentation of the Freundlich model for adsorption is as follows:

$$
\log q_e = \log K_F + \frac{1}{n} \log C_e \tag{4}
$$

Where, C_e = Chromium ion in equilibrium (mg/l); q_e $=$ total Cr adsorbed during equilibrium (mg/g); K_F & $n =$ Freundlich constants (K_F is the indicator of the adsorption capability/capacity and n is the indicator of the adsorption intensity; which were obtained

from the slope and intercept of the logarithmic plot of q_e vs C_e).

Tuble 0. Effect of separation factor on hotherin shape					
R^2 Value	Type of isotherm				
$R^2 > 1$	Unfavorable				
$R^2 = 1$	Linear				
$0 < R^2 < 1$	Favorable				
$R^2 = 0$	Irreversible				

Table 3: Effect of separation factor on isotherm shape

3. Results and Discussion

3.1 Batch studies

3.1.1 Consequence of adsorbent dose on chromium removal

The adsorption of chromium against the selected absorbent (i.e., AI, PO, TI, MI, and Eu) was studied at various adsorbent quantities (5, 10, 15, 20, 25, and 30 gm/250 ml solution). In the batch experiment, chromium concentration was maintained at 1100.3 ppm (wastewater from the Ruma tannery industry), temperature 30°C, and pH 2.5-3 to maintain natural effluent pH from the tannery industry for optimum contact time (12 hours). After 12 hours' percentage of chromium adsorbed was calculated. It was found that with the rising in adsorbent dose, the percentage of chromium removal from the solution also rises (for each case). It is because surface area increases which increase the adsorption sites absorb more Cr. Thus, automatically with increasing adsorbent dose, the removal of Cr increases (Figure 1). However, the removal of Cr was significantly increased up to the addition of 20 gm absorbent, but after a 20 gm dose, chromium removal from the solution was insignificant. 4.11, 7.84, 4.05, 8.31, and 10.15% chromium was reduced by AI, PO, TI, MI, and Eu for extra 10 gm adsorbent addition. Maximum chromium adsorption was assessed by TI (97.96%), and the minimum was found for PO (88.58%). However, adsorption capability is reduced with increasing adsorbent dose after a certain adsorbent dose at a constant concentration (1100.3 ppm for Ruma tannery effluent), as shown in Figure 1.

Maximum exposed adsorption sites enhance this type of higher adsorption capacity, and quick saturation on the surface occurs at a low dose. But with the increasing dose, scarcity of higher adsorption sites with a larger fraction of lower energy sites obtained in the solution. Adsorption surface area may overlap or be aggregated at the higher dose and lower active site available to chromium [27], producing lower adsorption capacity Younas et al (2022).Tokalioğlu, et al, (2000). Determination of heavy metals and their speciation in lake sediments by flame atomic absorption spectrometry after a four-stage sequential extraction procedure. Anal.Chim. Acta., 413(1-2); 33-40. From figure 1, it was found that the optimum dose for all bio adsorbents was 15 g; at that point, chromium removal percentage and capacity were in optimum condition.

Table 4: Constants for Freundlich and Langmuir adsorption isotherms for bio-adsorbents for chromium removal

Figure 1: Adsorption capacity/capability and percentile removal of chromium by different bio-adsorbent at different doses: a) AI, b) PO, c) TI, d) MI, and e) Eu.

Figure 2: Adsorption capacity/capability of chromium by adsorbents at different time periods.

3.2 Effect of time at chromium removal

Figure 2 shows the relation between contact time (3 to 18 hours) vs. adsorption capacity (mg/g) of chromium on the different bio-adsorbent from 1100.3 ppm solution (Ruma tannery effluents) at pH of 3, and the temperature was maintained at 30°C. From figure 2, it can be said that the Cr adsorption rate is high at the beginning due to the initial higher presence of chromium in the solution that was in close proximity with the adsorbent as well as higher

active sites were also present and later reduced gradually when equilibrium points were entirely achieved a saturation point (i.e., 12 hours for each of the bio-adsorbent). Adsorption capacity after 12 hours was 12.41, 11.84, 13.47, 11.37, and 11.82 mg/g for AI, PO, TI, MI, and Eu, respectively. After 12 hours, though, adsorption capacity, as well as chromium removal, occurred, but the rate was quite insignificant.

Figure 3: Adsorption isotherms of chromium by different bio-adsorbent: a) AI (Langmuir), b) AI (Freundlich), c) PO (Langmuir), d) PO (Freundlich), e) MI (Langmuir), f) MI (Freundlich), g) TI (Langmuir), h) TI (Freundlich), i) Eu (Langmuir), b) Eu (Freundlich).

3.3 Adsorption isotherm modeling

Two adsorption isotherm models (viz. Langmuir and Freundlich) were assessed to know their actual applicability. The value of all adsorption constants and regression coefficient for each of these isotherms are summarized in table 5. These were then computed against the best-fit lines. According to Mckay *et al*., for the best fit of Freundlich, the isotherm n value should be 1 to 10 US Department of Health and Human Services, (1991). This model fits the studied bio-adsorption process since the n values for each bio-adsorbent ranged from 2.9 to 5.7, and the regression coefficient of 0.9001 to 0.9640 indicates that this model fits the studied bio-adsorption process. On the other side, the obtained data of Langmuir constants point toward positive situations for adsorption. Again, higher correlation coefficients demonstrate that the Langmuir model (Figure 3) is comparatively better than Freundlich Model for the selected bio-absorbent. However, these findings are also well supported by some previous literature studies Silambarasan, T.S., Balakumaran, M.D., Suresh, S., Balasubramanian, V., Sanjivkumar, M., Sendilkumar, B. and Dhandapani, R., (2022).This observation clarifies that under the applied (applied during the recent research) experimental conditions, both heterogeneous surface and monolayer adsorption conditions may co-exist. From the Langmuir adsorption constant (qe), the maximum adsorption capacity on adsorbents was obtained from 15.24 mg/g and 19.23 mg/g for MO and AI, correspondingly. Differences in metal adsorption capacity have resulted from different structural properties, surface area, functional groups, etc., of various bio-adsorbent.

3.4 Reduction of conductivity, turbidity, and TDS value of Tannery effluents

Analysis of different Physicochemical characteristics of the collected effluents from the tannery wastewater is represented in Table 2. It was apparent that these effluents were polluted with various inorganic and organic contagions. Thus, these effluents show higher TDS, conductivity, and turbidity values**.**

The present study was undertaken only to reduce all those parameters using discarded plant parts so that these plants can be used as effective adsorbents in the future. The Cr concentrations, TDS, conductivity, and turbidity collected from three different tannery industries were noted to be higher than their allowable limits to release for horticultural uses like irrigation, as recommended by WHO. Under the batch experiment, 250ml of wastewater was treated on a rotary shaker at 30°C and 150 rpm for 12 hours with 20gm of each bio-adsorbent. All parameters were measured carefully from the initial to the finishing stage of each trial with an adsorbent. The presence of the mineral ion in wastewater is confirmed by its conductivity. But which ions might be present is not confirmed by conductivity. Higher conductivity indicates the presence of various types of mineral ions in the effluents; thus, removal of these ions may be difficult during treatment. Table 2 shows how effectively bio-adsorbent can be applied for the reduction of conductivity. It was found that Tamarind indicia bark can reduce the maximum amount $(87.72 \text{ to } 94.61 \text{ %})$ of conductivity which means it has a maximum site to adsorb minerals.

Name of	Chowdhury			Samina			Ruma		
Bio-	Initial		Final Reduced (%)	Initial	Final	Reduced $(\%)$	Initial		Final Reduced (%)
absorbent									
AI		20.90	82.26		15.00	86.93		8.30	92.16
PO ₁	117.80	18.10	84.63	114.80	16.70	85.45	105.80	9.90	90.64
TI		10.10	91.43		14.10	87.72		5.70	94.61
Eu		13.00	88.96		13.10	88.59		9.80	90.74
MI		16.50	85.99		16.50	85.63		10.60	89.98

Table 4: Removal of conductivity (mS/cm) from tannery effluents using bio-adsorbents

Total dissolved solids (TDS) include dissolved inorganic salts (mainly Ca2+, Mg2+, K+, Na+, CO32-, Cl-, and SO42-) and little organic matter in the water. Generally, the TDS is the summation of the anions (negatively charged) and cations (positively charged) in the water. Thus, the TDS analysis presents only a qualitative measurement of the dissolved ions except for the relationships or nature of ions. Therefore, TDS is applicable as a marker/indicator to decide the common feature of the effluents/water. During analysis, it was found that effluents collected from three industries were highly polluted and contained higher TDS value; this is due to leather processing, and each operational

section uses different types of organic and inorganic

chemicals. Each of these chemicals is added to the effluent water and pollutes the environment. After treatment with bio-adsorbent, it is found that almost 90 % TDS value is reduced. It is due to the use of a higher dose of adsorbent was used during treatment containing amino and acidic groups (activation site) so that these adsorbents adsorb most of the anions and cations. From Table 4, it was found that *Tamarind indicia bark* has a maximum capacity to adsorb ions, and so the TDS value also reduced to a maximum of 90.39, 87.41, and 94.40% for Chowdhury, Samina, and Ruma tanneries' correspondingly (table 5).

Name of		Chowdhury		Samina			Ruma		
Bio-absorbent Initial		Final	Reduced	Initial	Final	Reduced	Initial	Final	Reduced
			$(\%)$			$(\%)$			$(\%)$
AI		11.10	81.28		7.50	86.89		4.10	92.29
P _O		9.80	83.47		8.30	85.49		4.90	90.79
TI	59.30	5.70	90.39	57.20	7.20	87.41	53.20	2.98	94.40
Eu		8.10	86.34		8.10	85.84		5.30	90.04
MI		5.10	91.40		6.90	87.94		4.90	90.79

Table 5: Removal of TDS (g/l) from Tannery effluent using different Bio-adsorbents.

Relative clarity or clearness of liquid can be easily measured by its turbidity. Generally, the turbidity of water increases with the increasing scattered intensity of light. Substance like finely divided organic and inorganic matter, clay, silt, soluble colored organic compounds, different plankton, algae, and some microorganism causes water to make turbid. Particulates having high concentrations affect light penetration in the aquatic environment; thus, productivity, habitat quality, and recreational values are ultimately reduced, sometimes causing streams to fill in quicker. In rivers, sometimes siltation and sedimentation increase, which destroys the normal habitation of fish and other aquatic life. Again, particles act as a connector and easily attach many pollutants, especially bacteria, viruses, trace metals, etc. That's why the turbidity tests also act as an indicator against the potential pollution of any water body. Many chemicals (acid, base, salt, fat,

359.00

dye, and oil) are used during tanning operations. These have the main responsibility for the higher turbidity value of tannery effluents. After treatment with adsorbent, it was found that a significant amount of turbidity was reduced. Table 4 shows that Tamarind indicia's barks have a maximum capacity to adsorb ions, and turbidity value also reduced maximum (90.81, 90.91, and 93.95% for Chowdhury, Samina, and Ruma tannery, respectively). So this bio-adsorbent can be used as a primary treatment (initial stage without using any chemicals) of wastewater. After treatment with the bio-adsorption process, most of the organic, inorganic dye molecules were reduced largely; additionally, no chemicals were needed further to treat these wastewater, so the ETP cost was largely reduced.

281.00

297.00

PO 359.00 57.00 84.12 297.00 58.00 80.47 281.00 35.00 87.54

Table 6: Removal of turbidity (NTU) from tannery effluents using bio-adsorbents

86.12

4. Conclusion

The present investigation concluded that all bioadsorbent can successfully remove Cr from tannery effluent without any treatment for their significant adsorption capacity. Bio adsorbents were selected in this study due to their easy availability, low cost, and biodegradability. In this study, results indicate that adsorbent dose and contact time affect bioadsorbents' chromium uptake capability. Freundlich and Langmuir's adsorption isotherms are used to evaluate the equilibrium. Adsorption isotherm shows that both models are suited for these selected

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adsorbents which means monolayer, multilayer, homogenous and heterogeneous adsorption for chromium are possible. To determine the maximum adsorption capabilities of bio-adsorbents for chromium release from effluents computed isotherm constants were utilized. This study obtained a significant reduction in conductivity, turbidity, and total dissolved solids (TDS). All of the investigated parameters signify that chosen bio-adsorbent could be a superior alternative to expensive chemicals/methods; hence, the waste-water treatment process may become very economical.

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