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Successive Treatment of Chromium Contaminated Water by Indian Siris

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*Abstract***—** Chromium (Cr) shows hazardous effects and potential toxicity in water. A major significance lies in the exclusion of Cr (VI) from contaminated aqueous solution for a hygienic surroundings and wellbeing. Numerous methods for hazardous metal ions subtraction from an assortment of dissipated water sources embrace biosorption, chemical, membrane, electric, and photocatalytic-based treatments. This work observed the significance of utilizing an inexpensive bio-adsorbent *Albizia lebbeck* (Indian Siris) in Chromium contaminated waters. The batch experiment showed the impact of preliminary Cr (VI) concentration, adsorbent quantity, pH, temperature, contact time, and mixing speed on the biosorption of chromium (Cr) by the seed of Indian Siris. Langmuir's adsorption isotherm and Freundlich's isotherm model examined the effectiveness of organic and inorganic compounds on various adsorbents at equilibrium. Batch investigations were performed for material removal by adsorbents and Atomic Absorption Spectroscopy (AAS) for heavy metal analysis. The adsorbed amount showed the difference between the initial and final concentrations of the Chromium ion in an aqueous solution (mg/L). The biosorption capability was pH-dependent and increased with a boost in the initial concentration of adsorbate. During the early stages, the metal-binding rate with the biomass was more predominant, which gradually decreased and became steady after equilibrium. The high availability of Indian Siris (seed) behaves as a suitable adsorbent in removing the Cr metal under different conditions.

Keywords— Biosorption, adsorption isotherm, Albizia lebbeck, industrial waste, batch experiment, Atomic Absorption Spectroscopy

I. INTRODUCTION

Improvement in contemporary industrial custom is one of the most considerable grounds of environmental pollution. The quick expansion of civilization has upshot many objectionable pollutants. The chief problems of industrial wastes are the massive quantity of wastewater generated, and before it releases into the environment, elevated investment requires the handling of the effluents. In the last few decades, water pollution by dangerous pollutants has magnetized extreme consideration. The present industry is principally responsible for the contamination of the environment. Heavy metals cause toxicity to the living world, imitating the accomplishment of a crucial constituent in the food chain, hampering the metabolic route, and causing deterioration of the environment may accumulate in the food chain. Heavy metals' toxicity disrupts the central nervous processes, lowers energy levels, and harms the kidneys, liver, lungs, blood composition, and other essential organs. Chronic exposure may be upshot little by continuing physical, and neural disintegrative progressions that emulate Parkinson's disease, Alzheimer's disease, muscle dystrophy, epigastric pain, lung tumors, carcinogenicity, mutagenicity, teratogenicity, and multiple sclerosis [1]. The study

observed the significance of using an adsorbent *Albizia lebbeck* (Indian Siris) to remove Chromium metal from contaminated waters. The effect of initial Cr (VI) ion concentration, adsorbent dosage, pH, temperature, contact time, and amalgamation speed on the adsorption of chromium (Cr) by the seed of *Albizia lebbeck* was examined by the batch experiment. Langmuir adsorption and Freundlich's isotherms evaluated the biosorption of untreated water pollutants on an extensive range of adsorbents at equilibrium.

Scientific Classification:

Kingdom: Plantae Division: Tracheophyta Class: Magnoliopsida Order: Fabales Family: Fabaceae Genus: *Albizia* Species: *lebbeck*

Over the last few decades, the entrance of heavy metals via natural or anthropogenic sources shows absurd dejection in scientific society. Humans, aquatic fauna, and flora suffer from incurable diseases by readily adsorbing toxic elements on the surface of cell membranes.

Cr(VI) adsorption depends on numerous factors, including the surface modification, adsorbent's surface property, nature and type of adsorbent, substrate's pre-treatment, and investigational conditions (equilibration time, pH, initial Chromium ion concentration, ionic potency). Cr(VI) adsorption data was studied using stoichiometry of proton, adsorption kinetics, Langmuir, Freundlich's isotherms, and surface complexation model (SCM). Heavy metals become toxic or poisonous even at low concentrations (i.e., specific gravity >5). Heavy metal such as Chromium (Cr) is widespread and highly hazardous with specific gravity 7.19 [2].

A necessity is felt for inexpensive and efficient technology to treat metal-containing wastes for the elimination of the metal ion concentrations to environmentally tolerable levels to surmount some limitations of physiochemical treatments [3]. The most frequently employed conventional procedures; for removing toxic metal ions from streams include ion exchange, chemical precipitation, solvent extraction, reverse osmosis, ultrafiltration, adsorption, and membrane filtration [4].

Biosorption is efficient in biological, physical, and chemical systems. It is applicable in industrial appliances widespread, for instance, synthetic resins and activated charcoal for water purification. Adsorption shows benefits because of its trouble-free devise without sludge and trims expenses. The adsorbed substance is called adsorbate, and the adsorbing material has phrased the adsorbent [5]. Studies were conducted on removing toxic metal ions from synthetic wastewater using Groundnut shells and Rice husk [6], fly ash [7], coconut husk and palm Kernel fiber, peanut husk, charcoal, and natural zeolite [8], fava beans[9], rice straw [10] as effective adsorbents. Wastewater treatment using low-cost bio-adsorbents concluded that *Alisma Plantago Aquatica and* mango peel were highly likely to be employed for toxic ion subtraction [11]. A low-cost adsorbent such as Pinata leaves powder in removing Lead (II) ion from aqueous solution was performed with five parameters [12].

Various former constraints such as BOD, COD, Cl-, SO42- , and Fe were also diminished noticeably in the adsorption process. Several inexpensive bio-adsorbents and their operational methods for a range of agricultural and horticultural wastes were examined, such as bagasse, orange peels, almond shell, sugarcane rice husk, sawdust, sago waste, soybean hulls, cottonseed hulls, rice bran, coconut tree sawdust, palm pressed fiber, coconut shell, mango seed, clay, waste tea leaves, wood barks, and some bio adsorbents [13]. These adsorbents are readily available in India to eliminate heavy metals from wastewater.

Owing to the elevated cost associated with handling waste matters, including toxic metals in the environment, the incessant unprocessed release of chromium-restraining fabric industrial water has numerous harmful consequences on plants, ecological systems, and humans [14]. Hence, this research focused on using a low-cost, bioadsorbent *Albizia lebbeck* seed for chromium (VI) biosorption from contaminated aqueous effluent.

II. RELATED WORK

A study indicated that the acid-activated carbon preparation from the bark of *Albizia lebbeck* for the ferrous ion adsorption [15]. The adsorption phenomenon for that ferrous ion onto carbon was examined via XRD, IR spectra, and SEM images. By physical activation, the activated carbon characterization prepared from *Albizia lebbeck* was observed in a study [16]. The outcome signified the surface phenomenon and functional groups of the *Albizia lebbeck, such as c*arbon, as recognized by FTIR. A non-edible plant material, *Albizia lebbeck* (seed), was used for the removal and analysis by Atomic Absorption Spectroscopy of Pb metal ion from water medium to detect the impact of adequate parameters, for instance (pH, initial heavy metal ion concentrations, adsorbents dose, contact time, rotation per minute) to examine the Langmuir and Freundlich adsorption isotherm models [17].

III. METHODOLOGY

This experimental study was conducted on *Albizia lebbeck* as a bio-adsorbent of chromium (VI) from water medium on a laboratory scale in the Department of Environmental Sciences, Sambalpur University, Jyoti Vihar, Burla, Sambalpur, Odisha, during 2018-2020 by analyzing the bio-adsorbent's physicochemical characteristics and Chromium metal ion and working conditions (i.e., temperature, adsorbent quantity, pH, adsorption time, and initial Chromium concentration [18].

I. *Adsorbate preparation (Cr (VI)):*

All analytical-grade reagents were utilized as provided. Reagents and solutions were prepared using standard techniques. A standard stock solution of Chromium metal ion (Cr (VI)) as preparation of adsorbate was carried out by adding 2.82g of Potassium dichromate $[K_2Cr_2O_7]$ (Merck, Germany) in 100 ml double-distilled water (DDW) in the volumetric flask and then was diluted to 1ltr. A standard solution of $1.000 \mu\text{g/cm}^3$ (100 ppm)of Cr(VI) was prepared in vitro from the 1000 ppm stock solution by pipetting 10.0 $cm³$ in a volumetric flask up to 100.0 $cm³$ marked with DDW. To acquire the whole number values of pH between 3 and 8, the dilute solutions of 0.1 M HCl and 0.1 M NaOH were used. Analytical grade chemicals and doubledistilled water were utilized wherever necessary for experimental work.

II. *Sample collection and adsorbent preparation:*

The plant species used in this study as adsorbent, *Albizia lebbeck,* is often called "Indian Siris." Seeds of *Albizia lebbeck* collection were performed in local areas of the Bargarh district of Odisha. For three days, the seeds were then soaked in 1 M HCl to eliminate the original concentration of the Chromium ion. Then the Albizia seeds were taken for washing in distilled water, drying in the air,

and grinding with the use of a mortar and pestle. Ethanol/acetone mixture was used for mixing, and it was then centrifuged after shaking many times in anticipation that zero fat was remaining in the biomass. The wet seed powder of *Albizia lebbeck* was then dried after centrifugation in a hot oven at a temperature of 30^0C for 24 hours duration. Again the above was crushed and sieved through a sieve of 0.5 mm. Atomic absorption spectrophotometer (AAS) provided the initial chromium quantity and its leftover parts in the solution.

III. *Wastewater sample collection for Cr(VI) analysis:*

Samples of wastewater were assembled in acid-rinsed polythene bottles and were filtered through a 0.45 μm of Whatman filter paper and stored at 4°C until analysis. The prepared solution was taken within a pH 8–8.5 and standard buffer medium by obeying USEPA Method 218.6.

IV. *Material Characterization:*

Under adequate laboratory conditions, the **Batch experiment** was performed in the well-settled laboratory of the Department of Environmental Sciences, Sambalpur University. In REMI Bench top orbital Shaker Incubator (CIS- 24 B.L., RIS-24 BL), the solution was continuously shaken by taking 100ml of solution in 250 ml conical flasks. Then for further analysis in Atomic Absorption Spectroscopy (AAS), the water samples were taken out after a specific time interval and were sent to CPCB Kolkata.

V. *Adsorption experiment:*

Atomic Absorption Spectrometer (AAS) measured the Chromium metal ion concentration in the solution. The impact of pH, adsorbent amount, initial Cr concentration, contact time, and mixing speed was scrutinized on chromium ions' effectiveness and adsorption power by the adsorbents. For data analysis, Microsoft Excel and SPSS software were used. The featured conditions of this experiment were proved as follows. Each adsorption experiment was carried out more than twice for effective results.

The heavy metal chromium's adsorption equilibrium percentage and capacity were calculated using Equations (1) and (2).

$$
A\% = \frac{Co - Cf}{Co} \times 100
$$
 (1)

$$
Qe = \frac{(Co - Ce)}{M}V
$$
 (2)

In the above formula, "A" symbolizes the adsorption percentage; "qe" represents the quantity of adsorption capability; "Co" symbolizes the initial metal ion concentration in ppm (before mixing adsorbent) in solution in mg/l, "Cf " represents the equilibrium concentration in ppm of metal ion in solution (after mixing adsorbent); "Qe" is the quantity of metal ion adsorbed at equilibrium (mg/g), "Ce" indicates the equilibrium concentration of Cr (VI) in solution in mg/L and "m" means adsorbent mass(g), and "V" represents the volume of solution(L).

VI. *Langmuir adsorption Isotherm:*

Adsorption of molecules at a fixed number of definite active locations and its homogeneous distribution over the adsorbent surface were based on Langmuir's adsorption theory. These active sites showed a similar affinity for monomolecular layer adsorption, and no interaction was noticed between the adsorbed molecules [19].

The Langmuir equation [20] is articulated by Equation (3):
\n
$$
q_e = \frac{qm \, KL \, ce}{1 + KL \, ce}
$$
\n(3)

Its linear form representation is given below.

$$
\frac{\text{Ce}}{\text{qe}} = \frac{1}{\text{qm KL}} + \frac{\text{Ce}}{\text{qm}}
$$
(4)

In the above Equation, "Ce" (mg/L) represents the Chromium metal's equilibrium concentration in solution, "qe" signifies the adsorption equilibrium value (mg/g) , "qm" represents the maximum adsorbed quantity of Chromium metal ion per unit mass of sorbent, K.L. (L/mg) is the free energy of adsorption constant (mg/L).

VII. *Freundlich adsorption Isotherm:*

Freundlich's adsorption isotherm model interprets the biosorption phenomenon on various surface interactions between the adsorbed molecules. It was not restricted to the formation of a monolayer. This isotherm was commonly applied to the organic and inorganic compound adsorption [19].

Freundlich isotherm [21] is expressed by Equation (5):

$$
qe= Kf(Ce)^{1/n}
$$
 (5)

"qe" represents the absorbed quantity in mg/g, "Ce" signifies the adsorbed ions' equilibrium concentration in mg/L, "Kf" signifies the Freundlich isotherm constant, and "n" symbolizes the constant integrating all parameters affecting the adsorption process.

The linear representation of Freundlich adsorption isotherm (6) was employed for better evaluation of the sorption data and is represented in Equation (6):

$$
Log qe = log Kf + 1/n log Ce \tag{6}
$$

Scheming of log qe against log Ce represents a slope with a $1/n$ value and an intercept scale of log Kf.

IV. RESULTS AND DISCUSSION

This study took a powerful adsorbent Indian Siris (seed) to remove selected heavy metals like chromium. The adsorption of chromium in an aqueous solution on *Albizia lebbeck* seed was monitored by optimizing a range of physicochemical parameters (Table-1). Indian Siris (seed) was utilized as an adsorbent for the elimination of Cr (VI) from the aqueous sample in this study.

Effects of pH on biosorption

PH is a crucial factor in chromium adsorption from the aqueous solution and influences the adsorbent's surface charge and adsorbate's degree of ionization as described by Korwiak et al., 2011**.** The impact of several pH ranges between 3 to 7 was examined upon the chromium metal ion uptake, initial concentration of 20 mg/L chromium (VI), adsorbent dose of 10 g/L, contact time of 30 minutes, and mixing speed of 200 rpm. In this study (Table-1), removal percentages of chromium (VI) at pH 3, pH 4, pH 5, pH 6, pH 7, was 73.95, 74.65, 75.4, 73.45, and 70.45 respectively. The exclusion of chromium (VI) metal was maximum in the pH of 5(Figure -1, Table - 2); hence the optimum pH chosen was 5.0.

Effects of initial concentration on biosorption

A vital driving force is an initial concentration that affects the adsorption process. The impact of initial Cr(VI) concentration was observed within 10–50 mg /L, adsorbent dose 10 g/L, pH 5, and 30-minute contact time. The mixing speed was constant and equal to 200 rpm for all treatments. In all the optimization cases, chromium (VI) concentration of 20 mg/L., other concentrations were studied well to learn the possibility of chromium (VI) removal in other concentrations with the identical optimization situation. The result showed that at 10 mg/L, the adsorption was very low; it was 19 %. In 30 mg/L, 40 mg/L, and 50 mg/L, the chromium (VI) adsorption was found to be 65.9%, 59.42%, and 57.72% respectively. In 20 mg/L the study showed highest adsorption value of 75.95 % (Figure- 2, Table - 2).

Effects of adsorbent dose on biosorption

The metal ion adsorption is enhanced with an increase in adsorbent dose. The effect of a diverse quantity of adsorbent was within 10-50 g/L at an initial concentration of 20 mg/L Cr (VI), pH 5, 30 minute contact time, and 200 rpm. Increased adsorbent dosage implied a greater surface area and more binding sites present for the constant amount of chromium (VI). An optimum value of 40 g/L of the adsorbent dose was chosen (Figure - 3, Table - 2). An adsorbent dose ranging from 10-50 g/L was taken in the present study. At adsorbent dose 10 g/L, 20 g/L, 30 g/L and 50 g/L, the study showed 75.15 %, 74.05%, 76.35% and 79.55% chromium (VI) adsorption respectively. At 40 g/L study showed 81.45% adsorption, which was optimum.

Effects of Contact Time on biosorption

The prime motive of the experimentation is to measure the contact time needed to attain the equilibrium between the solid stage (biomass) and liquid phase (effluent). The contact time effect was examined within 30–120 minutes, pH 5, 20 mg/L chromium as initial concentration, 10 g/L adsorbent dose, and mixing speed of 200 rpm. In the present study, at 30 min, 90 min, and 120 min of contact time, the result illustrated 75.35 %, 85.20 %, and 85.55 % of chromium (VI) adsorption. The study revealed optimum chromium (VI) adsorption at 60 min. contact time with chromium (VI) removal of 85.45 %(Figure -4, Table- 2).

Effects of mixing speed on biosorption

Rotation per minute (rpm) performs an imperative function in the adsorption of toxic heavy metals. Toxic metal adsorption amplifies with the rise in mixing speed. The study was conducted at pH 5, 10 g/L of adsorbent dosage, initial Cr (VI) ion concentration of 20 mg/L, 30 min of contact time, and mixing speed varied from 50-250 rpm. At 50 rpm, 100 rpm, 150 rpm, and 250 rpm, the result showed 36.5 %, 43.85 %, 45.3 %, and 75.2 % of biosorption of chromium (VI) respectively. At 200 rpm, the result showed optimum adsorption of chromium (VI) of 75.2 % (Figure -5, Table- 2). Therefore an optimum value of mixing speed was noted as 200 rpm.

Adsorption Isotherms

Parameters of Langmuir isotherm "qm" and "b" were determined from the Ce/qe against the Ce plot. Langmuir adsorption isotherm showed qm value 85.45 and b value - 0.47 for chromium (VI). In Langmuir's isotherm model, R^2 was approved with the investigational data (Table-3). The investigational outcomes divulged that *Albizia lebbeck* could be utilized as a competent adsorbent to confiscate chromium (VI).

The linear representation of the Langmuir isotherm model for change in pH showed that the model fits well to an R^2 value of 0.99, as shown in figure - 6(a) and Table-3. Similarly, results can be observed in the alteration in contact time and mixing speed of Chromium (VI) by *Albizia lebbeck* with R^2 values 1 and 0.97, as shown in figure - 6(d) and figure - 6(e), respectively. Langmuir adsorption isotherm model was not fit well in case of change in initial Chromium metal concentration and change in adsorbent dose, as shown in figures - 6(b) and figure $-6(c)$.

The Langmuir and Freundlich adsorption isotherms were employed in the adsorption process, and their constants were assessed.

Discussion

The present experimental work was performed to assess the effects of the operational parameters on Chromium adsorption by Indian Siris (seed). The operating parameters comprised the adsorbent dose, the pH of the solution, the time required for equilibrium, the initial concentration of metal, and the mixing speed.

The highest Cr (VI) metal ion adsorption was found at 20 mg/L. Results signified that higher adsorption took place at a lower concentration. It was caused by the ions' interaction in the aqueous solution with the binding sites. Further, Chromium ions remained unabsorbed at higher metal concentrations in an aqueous solution because of the adsorption sites' degree of saturation [22]. The number of Chromium ions adsorbed from higher solution concentrations was more significant than that eliminated from less concentrated solutions. The percentage uptake was higher at a lower metal ion concentration attributable to the adsorbent's more large surface area accessible for adsorption. While Chromium ion concentration increased,

the removal percentage diminished as the existing adsorption sites befell fewer. At a higher Chromium ion concentration, the initial Chromium ion amounts to the adsorption sites were greater, ensuring a lesser adsorption percentage [23].

The experimental result showed that the utmost percentage of Chromium ion adsorption occurred in chromium (VI) - 40 gm/L. The removal percentage of Chromium ion augmented with the growing quantity of adsorbent dose [22],[24]. Equilibrium is achieved between the adsorbentbound Chromium ions and the leftover unabsorbed in the aqueous solution owing to almost all ions' binding to the adsorbent.

The experiment's primary purpose signified the examination of the contact time to attain the equilibrium stuck between the adsorbent and the synthetic water. The investigational result showed that the optimum contact time chosen for Cr (VI) was 60 min. From the result, we can remark that the uptake percentage amplifies with time, and it achieves a constant value after some time where no further Chromium ions can be eliminated from the solution [22]. The quantity of Chromium ions adsorbed represented the dynamic equilibrium state with the number of Chromium ions desorbed from the adsorbent. Equilibrium time symbolized the time needed to attain the equilibrium state with the utmost adsorption capability of the adsorbent under specific conditions [9], [23]. The outcome illustrated that Chromium ion adsorption enhanced with time up to 60 min. It became nearly constant at the end of the experiment. It can be inferred that the metal-binding rate with the biomass was more predominant during the initial stages that progressively decreased and remained nearly constant after equilibrium time. The active adsorption sites of the adsorbent were entailed in Chromium metal complexation while the adsorbent was introduced into the system.

Adsorption of heavy metal rises along with mixing speed and becomes nearly constant towards the end [10],[24]. In the present study, Chromium metal adsorption at 200 rpm mixing speed showed an optimal range of adsorption at 150 rpm mixing speed. The experimental result revealed a steady increase in the Chromium metal adsorption primarily, and then it became nearly constant towards the end. Consequently, Chromium metal adsorption was extremely slow at 50 rpm and 100 rpm. The adsorbent surface cannot be entirely rendered to the heavy metal solution.

Langmuir's adsorption model was most appropriate for explaining the adsorption process. The limitations of this study could be overcome by implementing Kinetic studies for the pseudo-first and second-order kinetic reactions. Also, thermodynamic parameters such as Gibbs free energy, entropy, and enthalpy could be used to make it thermodynamically favourable.

V. CONCLUSION AND FUTURE SCOPE

The experimental observations through the batch system reveal that biosorption occurs at several physiochemical parameters, and maximum adsorption occurs at pH 5, adsorbent dose 10 g/L, initial metal ion concentration 20 mg/L, showing the maximum temperature at 37 °C, at maximum contact time 120 min, and 200 rpm mixing speed offered the maximum adsorption percentage (85.55%) in this research.

PH range was maintained in-between 3 to 7 as adsorption could not be carried out beyond it. The maximum chromium (VI) removal percentages were 75.4% at pH 5. A steady boost in the amount of adsorption with time experimented. Adsorption increases from 19-57.72% in the time interval of 30 to 300 min. The adsorption was very low (19 %.) at an initial concentration 10 mg/L and optimum (75.95 %) at 20 mg/L. Among the adsorbent dose ranging from 10-50 g/L, a maximum of 81.45% adsorption was found at 40 g/L due to increased surface area and adsorption sites. Its corresponding increase with a decrease was observed. Optimum chromium (VI) adsorption was illustrated (85.45 %.) at 60 min. contact time. Cr concentration varied with an increase in agitation time. Among 50-120 rpm, the optimum adsorption of chromium (VI) was 75.2 % at 200 rpm.

The adsorption records best suited the Langmuir isotherm model with the R2 value of 0.99. The utmost adsorption capacity indicated the adsorbent's elevated efficiency and cost-effectiveness. *Albizia lebbeck* can be employed as a proficient bio adsorbent to remove chromium (VI).

This work will be beneficial in applying diverse approaches to boost the competence of adsorbents and physicochemical stipulations to remove heavy metals and dyes from contaminated streams in the future. The utilization of nano-adsorbents in industries will be implemented to reduce the harmful effect of solid and liquid waste. The most recyclable, low-cost, discriminating, and efficient treatment technique for eliminating toxic heavy metals and other organic pollutants from wastewater will be performed for future prospective.

Tables and Graphs

Figure 1 - Effects of pH on Chromium (VI) biosorption(%)

Figure 2- Effects of Initial Chromium (VI) concentration biosorption (%)

Figure 3 - Effects of adsorbent dose on Chromium (VI) biosorption (%)

(VI)biosorption(%)

Figure 6(a) - Variation in pH of Chromium (VI) by Langmuir adsorption isotherm

Figure 6(b) – Variation in initial Chromium (VI) concentration by Langmuir adsorption isotherm

Figure 6(c) – Variation in adsorbent dose of Chromium (VI) by Langmuir adsorption isotherm

Figure 6(d) -Variation in contact time of Chromium (VI) by Langmuir adsorption isotherm

Figure 6(e) - Variation in mixing speed of Chromium (VI) by Langmuir adsorption isotherm

Table 1. Physicochemical parameters setting for synthetic water

Table 2. Adsorption percentage of *Albizia lebbeck* (seed) with various Physico-chemical parameters for Chromium (VI)

Table 3. Parameters of Langmuir isotherm and correlation coefficient

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