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Biosorption of Hexavalent Chromium (Cr(VI)) from contaminated water using charred tea waste

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Abstract

The goal of this study is to develop charred tea waste (CTW) via chemical process for the removal of Cr(VI) from contaminated water. Batch adsorption experiments were conducted as a function of pH, initial concentration, contact time and adsorbent dosage. Characterization of the adsorbent was analyzed by FT-IR and XRD. Maximum adsorption capacity (q_m) of the CTW was found to be 85.32 mg/g at optimum pH 2 in 120 minutes. The adsorption on CTW was well fitted to the Langmuir isotherm and the kinetic data is consistent with the pseudo-second order kinetic model. The findings suggest that CTW could be an efficient and promising adsorbent for the removing Cr(VI) from aqueous solution.

Keywords
Hexavalent Chromium, Charred tea waste, Langmuir isotherm, Pseudo-second order.

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1 Introduction

Access to safe drinking water is internationally recognized human right. Presence of organic pollutants, inorganic pollutants and microbes are causing water pollution. Presence of metals or metal ions such as cadmium, mercury, lead, arsenic, chromium, nickel, etc. in environment is causing heavy metal ions pollution which brings about an adverse effect on human health as well as aquatic flora and fauna [1]. Discharge of heavy metal ions such as chromium rich effluent from tanning, leather, metallurgy and electroplating industries into water body is being a serious issue these days. Its prevalence is increasing due to industrial production of dyes, leather, textiles, electroplating materials, and chemical manufacturing and some similar channels [2]. Different oxidation states of chromium are present, and they all affect people differently. Cr(VI) is toxic and highly carcinogenic, despite the fact that trace amounts of chromium (III) have been reported to be necessary for proper lipid, insulin, and glucose metabolism [3]. World health organization (WHO) has assigned its upper permissible level in drinking water as 50 ppb [4]. Chromium gets exposed to the environment via natural as well as anthropogenic routes [5]. Many chromium-based materials are based on hexavalent chromium which exhibits its strong oxidizing power. Hexavalent chromium either directly enters into the workers through dermal contact or enter into the human via water resource. Industrial wastewater and human activities are polluting the environment more than a bearable level. Studies have been undertaken using various types of adsorbents for its efficient and selective removal due to its extreme toxicity and pollution concerns. Over use of heavy metals pollutes the water. The use of heavy metals has health, environmental and aquatic effects [6].

In connection to the increasing amount of Cr(VI), various absorbents have been synthesized for the effective and selective removal of such heavy metal ions. Among the different types of adsorbents, the biomass-based adsorbents are getting interest these days due to their non-toxic nature towards environment, low-cost and tunable functionality. In order to remove chromium from industrial effluents, varieties of methods and materials are used for the effective and efficient removal of heavy metal ions from the sources. Some of the commonly used approaches are chemical precipitation, ion-exchange methods, electrochemical precipitation, chemical reduction, solvent extraction, membrane filtration or separation and so on [7]. However, many of these methods are less sensitive for adsorptive removal of heavy metal ions below the tolerance level due to lack of sensitivity or selectivity.

Removal of heavy metal ions by adsorption methods is the most promising for overall treatment. Though different types of nanoparticles, polymers/conductive polymers, especially heteroatom containing conductive polymers [8,9], layered compounds [10] are being used for the removal of heavy metal ions, biomass-based low-cost materials are getting interest for the effective removal of toxic metal ions form water. These adsorbents are easily available, eco-friendly, and cost effective with higher capacity of adsorption. In addition, such types of biomass-based materials can be chemically processed to increase its effectiveness. Commonly, activated carbons are being widely used as adsorptive materials for waste water treatment [11,12].

Commonly used waste materials used to prepare biomass-based adsorbent are banana peels, plum kernels, oil palm shells, tea leaves, saw dust, wheat bran, coconut husks, coffee grounds, rice husks, jackfruit peels and many others.

In this context, most commonly used bio-adsorbents are fruit gum, wheat bran, cork powder, sugar beet, rice bran, rice husk, nut shells, tea leaves and so on. Each of the mentioned adsorbent capacitates to adsorb chromium form aqueous solution [5]. Activated carbon is considered as a potent adsorbent with high surface area, microporosity and surface chemistry [13]. One of the biomass-based precursors for the preparation of activated carbon could be Tea waste which is a common material left after the preparation of tea. Nepal is rich in tea production. With the processing and refining into tea powder, tea leaves remain as residue. Such waste materials can be employed for the preparation of activated carbon and further chemical treatment. Furthermore, the composition of tea leaves may be different as a function of geography and climate.

Tea waste contains cellulose, hemicellulose, lignin and a lot of pectin in the cell walls. Pectin is polysaccharides in which pectic acid and acidic polysaccharides, mainly composed of D-galacturonic acid and methyl ester, are present. Tea leaves contain various polyphenols including epigallocatechin gallate (usually known as EGGCG), flavonoids and other catechins [14]. Large-scale hot water extraction of tea leaves is employed for the instant production of canned or bottled teas and producers have to struggle to dispose the discarded tea-leaves wastes. Therefore, the use of such waste materials is highly preferable for the synthesis of activated carbon [15]. In this context, tea waste can be regarded as a potential precursor which can be used for the removals of Cr(VI) ions from aqueous media. This method is benefitted by the facile mode of processing, easily available, cost-effective and environmentally friendly features [16].

The synthesis of charred carbon materials from tea waste of Nepal origin has not been reported for the adsorptive removal of chromium (VI) listed in the comparison table 1. In this work, activated carbon has been prepared from the tea waste followed by charring with concentrated sulphuric acid. The objective of the present study is to find out the maximum uptake capacity of prepared charred tea waste for the removal of Cr(VI) from contaminated water. Batch adsorption experiments would be carried out changing the pH of solution, adsorbent dosage and contact time. The biosorbent CTW is ecofriendly and it can be incinerated with zero emission due to its natural constituents. On the other hand, the adsorbent is cost-effective since it is prepared from agricultural byproduct of tea. It could be an efficient and promising biosorbent for the treatment of Cr(VI) from contaminated water.

2 Materials and Methods

2.1 Preparation of Chromium (VI) solution

Stock solution of Cr(VI) of 1000 ppm concentration was prepared by dissolving 1.414 g of potassium dichromate in water followed by the addition of 100 mL solution of 2 N HNO_3. Then the volume of the solution was made exactly 500 mL by adding distilled water. All the chemicals used in this study were of analytical grades.

2.2 Preparation of Charred Tea Waste

Waste tea leaves were collected from the tea shop, Lazimpat, Kathmandu. The tea waste was washed with distilled water several times, dried and ground. For charring process, 250 grams of ground tea waste were placed in a beaker of 1 liter capacity followed by the addition of concentrated sulphuric acid at an installment with constant stirring until the content turns to the black. In this step, ring opening of cellulose unit occurs. The reaction was left for 24 h for completion. The carbon was washed with deionized water several times to bring up to neutralization. Then the sample was dried in vacuum at 50 °C for 9 h. The dried charcoal was ground in an agate mortar using pestle and sieved through 200 microns. Finally, a sample was completely dried keeping in desiccator which is known as Charred Tea Waste (CTW) and stored in an airtight bottle.

2.3 Instrument used

UV-Visible double beam spectrophotometer (Labtronics, LT-2802, India), FTIR-spectrometer (Perkin Elmer 10.6.2, USA), Digital pH meter (HANA, HI2002-01 edge), four digit weighing machine (Phoenix, PH2204C), Hot air oven (ELITE Oven), Magnetic stirrer (LABINCO L34) and Sieve (New Dehli-110055).

2.4 Sorption study

Stock solution of Cr(VI) was used for the study of Cr(VI) adsorption by charred tea waste. For this purpose, the stock solution of Cr(VI) was diluted to prepare a series of solution form 10-300 ppm. The effect of dosage, effect of initial concentration, pH of the solution and contact time of the adsorption process was investigated. The pH of solution was maintained by using 0.5 N NaOH and 0.5 N HCl solution where necessary. 20 mg of prepared adsorbents was taken in a stopper bottle and 25mL of chromium solution were agitated in a shaker at room temperature for 5-6 hours. Then, the solution was filtered and the filtrate solution was taken. The absorbance of metal ion solution before and after adsorption was determined by using double beam UV-Visible spectrophotometer. The amount of metal ion adsorbed onto adsorbent is calculated using the following relation given in equation (1).

$$q = \left(\frac{C_i - C_e}{W}\right) \times V \tag{1}$$

Here, q (mg/g) is the quantity of adsorbate adsorbed per unit mass of adsorbent. In the equation, V is the volume of solution in liter. W(g) is the mass of adsorbent, C_i and C_e are concentration of Cr(VI) solution, respectively. The percentage adsorption capacity (%A) of the metal ion can be used by the relation of equation (2):

$$Percentage\ adsorption(\%A) = \frac{C_i - C_e}{C_i} \times 100 \tag{2}$$

3 Results and Discussion

3.1 X-Ray Diffraction (XRD) Analysis of Adsorbents

X-ray diffraction (XRD) study was carried out to study the crystalline/amorphous nature of the adsorbent. The XRD pattern of the raw tea waste (RTW) and charred tea waste (CTW) exhibit a broad peak in the 2θ range of 14-23° indicating a predominantly an amorphous nature. Before and after modification, the XRD patterns of RTW and CTW are not so significantly different shown in figure 1. The amorphous biosorbent allows the Cr(VI) ions adsorption by penetrating the surface rapidly, hence increase in adsorption capacity [17].

3.2 Fourier Transform Infra-Red Spectra (FTIR) Analysis

The functional group study for the RTW and CTW materials were carried out in terms of FTIR analysis in the Department of Chemistry, Amrit Campus. The FTIR spectra of RTW and CTW are shown in figure 2. The broad absorption at 3200-3500 cm^{-1} corresponds to the –OH group of the adsorbent and small absorption band at 2917 cm^{-1} corresponds to the CH stretching in methylene group. The absorption band around 1704 cm^{-1} and1610 cm^{-1} correspond to the C=O and C=C group, respectively [18]. The C-OH stretching band at 1032 cm^{-1} of RTW got shifted and slightly deformed in CTW which could be associated with the charring process of the material [19]. The FTIR spectra reveal the intensity and frequency change in the transformation of RTW to CTW indicating the involvement of chemical reaction. This happens due to breakdown of chemical bonds during the process of charring.

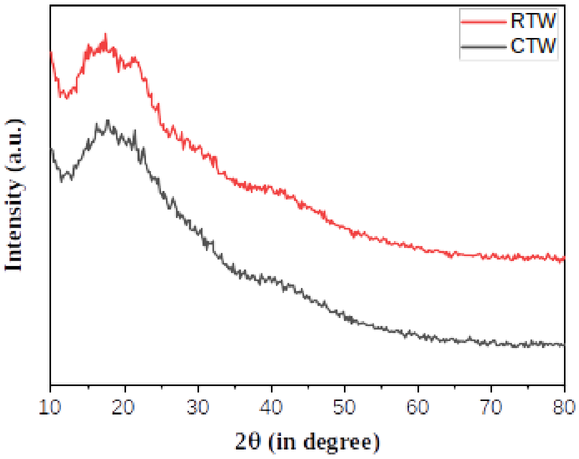

Figure 1: X- ray diffraction of RTW and CTW.

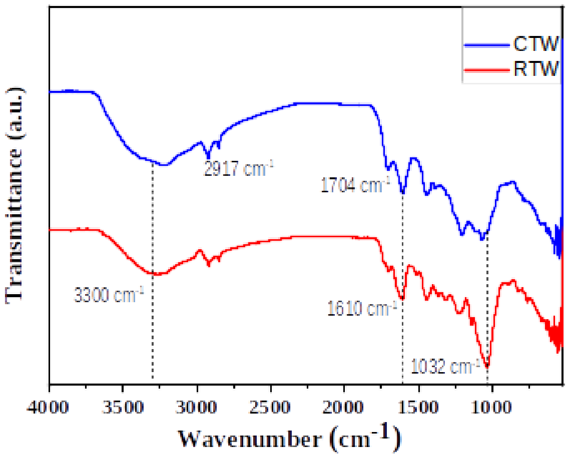

Figure 2: FTIR Spectra of RTW and CTW.

3.3 Effect of pH on Hexavalent Chromium Adsorption

The pH of the adsorbate solution has been reported to be one of the variable for Cr(VI) adsorption. For this study, 50 ppm of chromium(VI) solution was taken and its pH was maintained using pH meter in different reagent bottles from pH 1 to pH 5 keeping 0.02g of CTW in each bottle. From pH studies it was found that the % removal of chromium by CTW increased from 48.77% to 81.42% then again decreased up to 40.91%. The maximum percent Cr(VI) removal was found to be 81.42% by which we can conclude that the optimum pH found to be 2 as shown in figure 3. This finding is in quite agreement with the reported literature [6]. Below the pH 2, some of the chromium (VI) exists as H_2CrO_4 which is hardly adsorb onto the adsorbent. From pH 0 to 6.5, Chromium(VI) exists as $HCrO_4^-$ (hydrogen chromate) while above this pH range chromium(VI) exists as chromate ions (CrO_4^{2-}) predominantly. At the optimum pH value, chromium exists predominantly as $HCrO_4^-$ anion in aqueous media. With increasing the pH value the complexation process of Cr(VI) goes on decreasing resulting into the low uptake of Cr(VI).

3.4 Effect of dose

Adsorption experiment was carried out at lab temperature (25°C) taking different weight of adsorbent to find the optimize amount of adsorbent dosage for the removal of Cr(VI) from its 50 ppm aqueous solution. The upper tolerance level of chromium in drinking water is also 50 ppb. Adsorption capacity of adsorbents increases upto a certain weight then it remains constant keeping in Cr(VI) solution. With the increase of adsorbent dosages from 5 mg to 100 mg under the experimental conditions like pH, temperature and adsorbate concentration constant, the

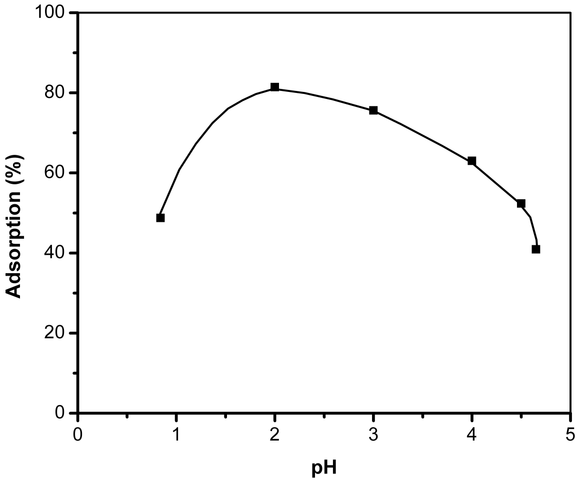

Figure 3: Effect of pH for adsorption of Cr(VI) onto CTW.

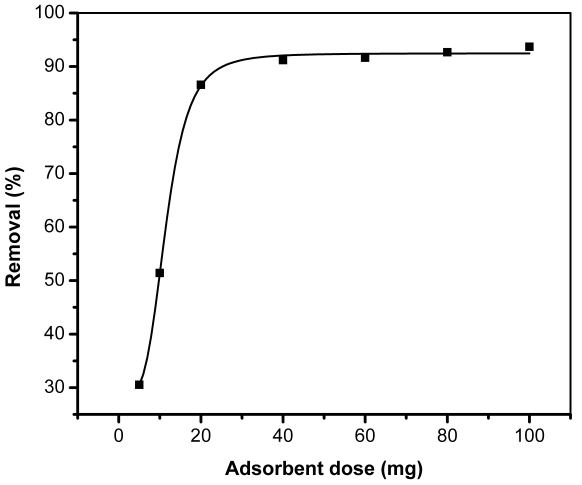

Figure 4: Effect of CTW dose on Cr(VI) adsorption.

adsorption study was carried out. Low mass of adsorbent corresponds to the low active sites and high mass of adsorbent corresponds to the high active sites. With the increase of active sites, the percentage removal of Cr(VI) got increased. The result shows that the percentage removal of Cr(VI) increases with up to 20 mg adsorbent doses and remains almost constant for the studied adsorbent doses as shown in figure 4. The highest adsorption for 50 ppm adsorbate of Cr(VI) is found to be 20 mg.

3.5 Effect of concentration of Cr(VI) solution

The adsorption capacity of CTW can be evaluated with the help of an adsorption isotherm for the adsorption of Cr(VI). As the concentration of Cr(VI) ions increases, the adsorption of metal ions increases. At low Cr(VI) ions concentration, the concentration of Cr(VI) ions are limiting and get sufficient sites for their adsorption whereas at high concentration, the active sites on adsorbent are limiting compared to the Cr(VI) concentration and hence become saturated. The absorbance of the supernatant clear liquid is measured spectrophotometrically at the maximum wavelength of after 4h equilibrium interval. A graph was plotted and used to calculate the amount of Cr(VI) adsorbed. The maximum adsorption capacities of experimental results shown in fig.5 found to be 85.32 mg/g onto CTW at optimum pH2.

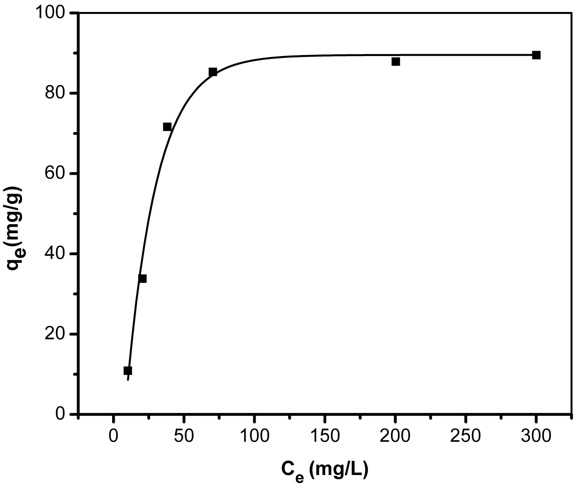

Figure 5: Effect of concentration for adsorption of Cr(VI) onto CTW.

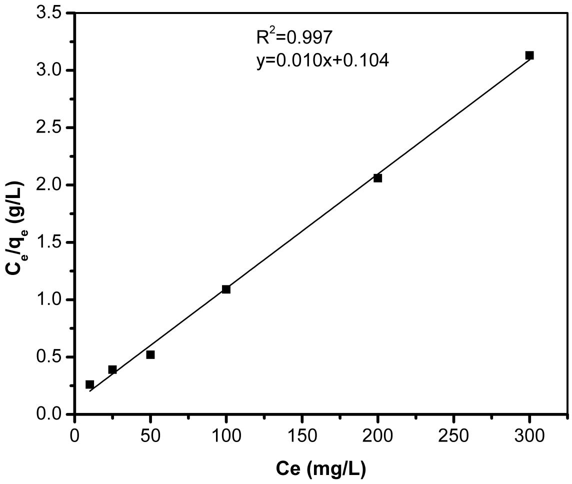

Figure 6: Langmuir isotherm plot for biosorption of Cr (VI) onto CTW.

3.6 Adsorption Isotherm

The correlation coefficient (goodness of fit) values for Freundlich and Langmuir isotherms were used to determine the applicability of the isotherms. Langmuir and Freundlich's relation, Equation (3) and (4), respectively, were used to calculate adsorption isotherms.

$$\frac{C_e}{q_e} = \frac{1}{q_m b} + \frac{C_e}{q_m} \tag{3}$$

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

In these equations, Ce(ppm) is the equilibrium concertation of adsorbate, q_e (mg/g) is the amount of Cr(VI) ions adsorbed per gram of adsorbent at equilibrium, qm (mg/g) is the maximum adsorption capacity, b (L/mg) is the constant. By plotting a graph of Ce/q_e versus Ce, the value of qm and b can be evaluated. The K_F (L/mg) is adsorption capacity and 1/n is adsorption intensity which indicates the relative distribution of energy and adsorption heterogeneity site. The result shows that the maximum adsorption capacity is found to be 85.32 mg/g onto CTW at the pH of 2. Higher initial concentration can increase the higher adsorption of metal ions. With the increase of concentration, the adsorption process get saturated. The higher value of Langmuir correlation coefficient (R^2 = 0.9974) indicates the better fitness of Langmuir model compared to the Freundlich model. It shows that the metal ion adsorption is predominantly a unimolecular. Additionally, in Langmuir's isotherm, the equilibrium parameter, the dimensionless equilibrium

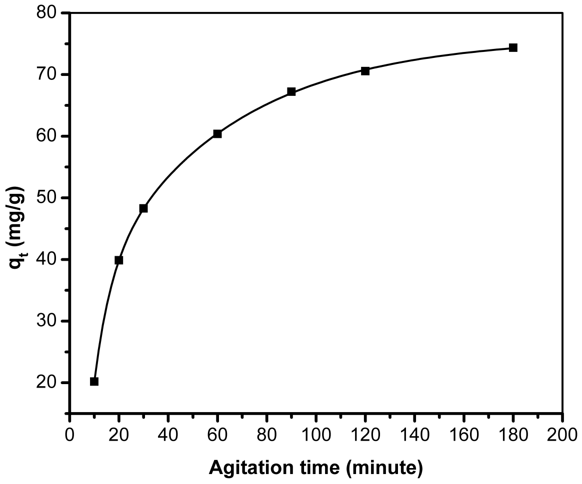

Figure 7: Time on adsorption of Cr(VI) onto CTW.

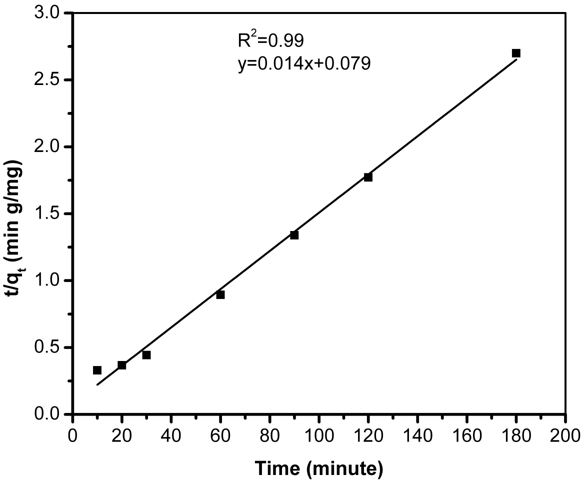

Figure 8: Pseudo second order adsorption kinetics of Cr(VI) onto CTW.

constant (K_L), given by equation (5):

$$K_L = \frac{1}{1 + bC_i} \tag{5}$$

where b is the Langmuir constant and C_i is the initial metal ion concentration. In this research work, the value of K_L is between 0 and 1 indicating the nature of favorable Langmuir model of adsorption isotherm.

3.7 Effect of contact time

For the study of effect of contact time in the adsorption process, 20 mg of adsorbent was put into the 20 mL of 50 ppm of Cr(VI) solution. The pH of the solution was maintained 2 for the study. After shaking the solution for the 24 h, the content was filtered and the clear solution was processed for its concentration measurement using double beam UV-Vis spectrophotometer. Adsorption of Cr(VI) was initially increased with increase of the contact time and remain constant. Figure 7 shows that the amount of adsorption increased from 5 minutes up to 120 minutes, and then remained constant thereafter.

3.8 Adsorption Kinetics

The experimental adsorption kinetic data can be evaluated following integrated form of pseudo-second-order kinetics model explained by Ho and Mckay [20] as given in equation (6).

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e} \tag{6}$$

Here, q_t (mg g^{-1}) is the amount of the adsorption at time (min), k_2 (gmg^{-1}min^{-1}) is the rate constant of pseudo-second order kinetic of adsorption. Using the plot of t/q_t versus time, we can calculate the values of K_2 and qe. Pseudo-second order kinetics plot gives the perfect straight line for the adsorption of Cr(VI) onto CTW shown in Fig.8. Correlation coefficient R^2 value is 0.992 which indicates the pseudo second order kinetic model for the adsorption. The correlation coefficient value is about unity i.e. 0.992 confirms that metal ions, Cr(VI) are chemisorbed onto CTW.

3.9 Biosorption Mechanism

The high value of q_m could be associated with high surface area resulted from ring opening of cellulose, hemicellulose or lignin mass due to acid treatment in charring process. The ring opening process of cellulose increased the active surface sites during the charring process. In the hexavalent chromium having ionization process at pH2, the concentration of $HCrO_4^-$ ions are maximum. Furthermore, at a low pH condition, protonation is possible due to high concentration of protons. As a result, $HCrO_4$ is electrostatically attracted to the protonated surfaces of the biosorbents, which helps it to bind to the surface of CTW (figure is not given). Similarly, Cr(VI) get adsorbed onto the protonated sites within the CTW by an ion-exchange mechanism [20, 21].

Table 1: Maximum Cr(VI) biosorption capacities (q_m) of CTW with other reported biosorbents.

SN	Biosorbent	pH	q_m (mg/g)	Reference
1	Arthrobacter viscous biomass	2.0	20.377	[21]
2	Waste newspaper	3.0	59.88	[22]
3	Coconut husk	2.0	29.00	[23]
4	Cacktus fruit (opuntia)	2.0	18.50	[24]
5	Carbon slurry	2.0	15.24	[25]
6	Nutshell powder	2.0	72.12	[26]
7	Coconut coir pith	2.0	76.3	[27]
8	Banana peel	1.5	10.42	[28]
9	Charred tea waste	2.0	85.32	Present work

4 Conclusion

The charred tea waste was prepared by the treatment concentrated sulphuric acid into tea waste. The maximum Cr(VI) uptake capacity was found to be 85.32 mg/g onto CTW at pH 2 reaching equilibrium in 120 min. The maximum percent Cr(VI) removal was found to be 81.42% at the optimum pH 2. The experimental data were in good agreement with the Langmuir isotherm. The adsorbent CTW follows pseudo-second-order kinetics with the correlation coefficient ($R^2 = 0.992$). The result of high adsorption capacity with fast kinetics concludes that CTW can be used as an efficient bio-adsorbent for the sequestration of Cr (VI) from contaminated water.

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