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The Novel Use of Saraca thaipingensis Leaf Powder as Potential Biosorbent Material for the Removal of Chromium(VI)

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Abstract

Saraca thaipingensis or ‘Gapis’ tree, classified under the Fabaceae family is a native of Taiping; copious over Peninsular Malaysia and Southeast Asia. The withered and fallen dead leaves were collected from INTI International University’s garden walkway. To date, literature has yet to capture the use of *S. thaipingensis* tree parts or refuse as potential biosorbent material for the removal of heavy metals thus verifying the novelty of this study. Batch experiments were carried out with the leaf powder to study the effects of dosage, particle size and contact time towards Cr(VI) removal (%) at 1-100 mg/L. Results showed that Cr(VI) removal increased from 52.22% to 99.31% ($p < 0.05$) with increase in biosorbent dosage (0.005, 0.010, 0.015, 0.020, 0.025 and 0.050 g). The different particle size ranges tested were: 107-125, 126-150, 151-250, 251-500, and 501-1000 µm. Highest Cr(VI) removal of 99.53% was obtained with the 151-250 µm particle size; further size decrease did not yield more removal ($p > 0.05$). The optimal Cr(VI) removal was recorded after 45 min (99.62%) and 90 min (99.76%) contact time ($p > 0.05$). Further characterization and optimization studies are being carried out to develop a novel, sustainable, low cost yet effective leaf powder based biosorbent material.

1. Introduction

Water pollution arising from heavy metals has become one of the most important environmental concerns and pose detrimental hazard to man and nature [1]. Many industries use and generate chromium containing waste such as the metal finishing and electroplating industries, metallurgical works, film processing, tannery, chemical manufacturers, electronics, and petrochemicals, amongst others [2]. Chromium(VI) can occur as anionic chromate (CrO_4^{2-}), dichromate ($Cr_2O_7^{2-}$), hydrogen chromate ($HCrO_4^-$) or chromic acid (H_2CrO_4) and are very soluble and known to be carcinogenic and mutagenic to living organisms [3]. In many instances, Cr is released into the ecosystem by poor storage, leakage, inadequate disposal practices.

Current conventional physico-chemical treatment methods include chemical precipitation, activated carbon adsorption, ion-exchange resins, electro dialysis reverse osmosis, floatation, and membrane filtration [1,3]. These processes are costly and economically ineffective, single-waste oriented, inconsistent in performance, and have many technical limitations. The continuous use of chemicals and the toxic chemical sludge generated further aggravates the pollution issue. Hence, biosorption is presented as the alternative option for a rapid, reversible, cost effective, and environmental friendly technology. Over the years, biomass of either living (active) or dead (inactive) such as microorganisms including bacteria and bacterial products [3,4,5], fungi, yeast, and actinomycetes, seaweeds and algae [6,7] have been used for the removal of heavy metal ions. Nowadays, the use of low cost natural products or biowaste materials have gained much attention with the advancement of biotechnology [8,9,10]. This will prove to be a much more economical, effective, efficient and safer approach.

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However, only systems that render broad selectivity, high metal uptake and removal capacity, low operational cost, void of waste sludge will be of significance for applications [2,10].

This research explores the feasibility of the leaf powder of a local tree species as potential novel biosorbent material. *Saraca thaipingensis* or ‘pokok Gapis’, also known as the handkerchief tree, is classified under the Fabaceae or Leguminosae family is a native of Taiping, copious over Peninsular Malaysia, and most parts of Southeast Asia. The withered and fallen dead leaves were sourced from the trees grown at the INTI International University’s garden walkway. Not many biosorption studies have reported the use of tree refuse biomaterial as biosorbents. To date, literature search has yet to capture the use of *S. thaipingensis* tree parts or refuse as potential biosorbent material for the removal of heavy metals thus verifying the novelty of this study. In this pilot study, batch experiments were carried out with the leaf powder to study the effects of dosage, particle size, contact time and varying initial Cr(VI) concentrations towards Cr(VI) removal and uptake. The data gathered will be useful to follow-up with more application based research in view of developing a novel, sustainable, low cost yet effective leaf powder based biosorbent material.

2. Materials and Methods

2.1. Chemicals and Reagents.

All reagents used were of analytical grade. The Cr(VI) stock solution (1000 mg/L) was prepared using potassium dichromate ($K_2Cr_2O_7$) salt in double distilled water. The working and test solutions of differing Cr(VI) concentrations were prepared by diluting the stock solution to the desired volumes.

2.2. Sample Sourcing and Preparation

The withered and fallen dead leaves of *S. thaipingensis* were sourced from the many trees grown at the INTI International University’s garden walkway. The leaves were torn into smaller pieces washed repeatedly with distilled water and dried overnight at 60 °C. The dried leaf pieces were ground with a high speed grinder at 26 000 rpm for 2-3 min (Shanghai Jingke Scientific Instrument Co., Ltd.) in batches. The leaf powder was sieved (laboratory test sieves; Endecotts Ltd., London, England) to obtain particle size ranges of 501-1000, 251-500, 151-250, 126-150 and, 107-125 µm. The biosorbent leaf powder was stored in air-tight bottles prior to use.

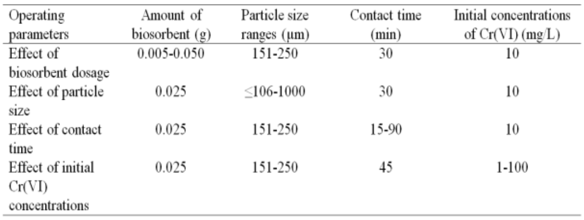
2.3. Chromium(VI) Analysis

The standard analysis protocol using the Cr(VI)-specific colorimetric reagent 1,5-diphenylcarbazide (DPC) was followed [3,7]. The % Cr(VI) removal is given as = $C_0 - C_e/C_0 \times 100$, and Cr(VI) uptake (q_e, mg/g) = $C_0 - C_e/w \times$ volume of solution (L); where C_0 (mg/L) is the initial concentration of Cr(VI), C_e (mg/L) is the residual concentration of Cr(VI) at equilibrium, and *w* is dry weight of leaf powder (g) [3,7].

2.4. The Effect of Biosorbent Dosage, Particle Size, Contact Time, and Initial Concentration

The experimental design is as presented in Table 1. All the operating parameters were evaluated in batch mode. Final volume of flask mixture was 25 mL and initial pH was adjusted to pH 2 (optimal pH as determined earlier; data not shown). The flask mixture was agitated at 250 rpm (orbital shaker; Smith) in ambient temperature (26 ± 2°C).

Table 1: Experimental design and parameters investigated



Operating parameters	Amount of biosorbent (g)	Particle size ranges (µm)	Contact time (min)	Initial concentrations of Cr(VI) (mg/L)
Effect of biosorbent dosage	0.005-0.050	151-250	30	10
Effect of particle size	0.025	≤106-1000	30	10
Effect of contact time	0.025	151-250	15-90	10
Effect of initial Cr(VI) concentrations	0.025	151-250	45	1-100

Statistical Analysis. All tests were carried out in triplicates and the mean recorded. The data were subjected to appropriate tests (IBM SPSS version 21). Significance level was set at 95% (α = 0.05).

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3. Results and Discussion

3.1 The Effect of Biosorbent Dosage, Particle Size and Contact Time

The effect of biosorbent dose on Cr(VI) removal is presented in Table 2. The mean percentage of Cr(VI) removal increased from 52.22% to 99.31% with an increase of biosorbent dose from 0.005 g to 0.050 g. This was due to the increase of available binding sites and active surface area as the biosorbent amount increased. There was a significant difference ($p < 0.05$) between mean percentage of Cr(VI) removal for each group of biosorbent dose. The lowest removal was observed with 0.005 g powder followed by 0.010 g. Although the highest removal was observed at 0.050 g, but multiple comparisons test revealed that there was no significant difference ($p > 0.05$) in the % Cr(VI) removal between the 0.020 g, 0.025 g and 0.050 g dosage range. Although more leaf powder amount increases the surface area, but the non-significant removal % may be due to competition between the Cr(VI) ions for the available sites [2,4]. In view of this, mass of 0.025 g (0.1% w/v) was identified as optimal.

Table 2: The effect of biosorbent dosage towards Cr(VI) removal at C_0 = 10 mg/L after 30 min with particle size of 151-250 µm.

Biosorbent dosage (g)	Cr(VI) removal (%)
0.005	52.22 ± 4.61
0.010	70. 24 ± 3.41
0.015	89.01 ± 0.68
0.020	94.68 ± 0.86
0.025	95.90 ± 0.95
0.050	99.31 ± 0.39

Note: readings are mean ± standard error (n = 3).

In terms of particle size, based on Table 3, there is little effect towards Cr(VI) removal below the 500 µm particle size ranges ($p > 0.05$). The highest Cr(VI) removal was seen at 99.53% with the 151-250 µm particle size leaf powder, and the lowest with the 501-1000 µm (79.33%). However, the % Cr(VI) removal increased with a decrease in particle size only up to the 151-250 µm particle size range beyond which Cr(VI) removal started to decrease with a decrease in particle size although the magnitude was insignificant. Hence the optimum particle size range was determined as 151-250 µm.

Table 3: The effect of particle size towards Cr(VI) removal at C_0 = 10 mg/L after 30 min with biosorbent dose of 0.025 g.

Biosorbent particle size range (µm)	Cr(VI) removal (%)
501-1000	79.33 ± 2.52
251-500	95.93 ± 1.37
151-250	99.53 ± 0.12
126-150	99.46 ± 0.18
107-125	98.15 ± 0.50
≤ 106	98.89 ± 0.22

Note: readings are mean ± standard error (n = 3).

For the adsorbate and biosorbent to achieve equilibrium, an adequate amount of time is necessary for the reaction to complete. Table 4 indicates a rapid increase in Cr(VI) % removal by the leaf powder from 91.65% (after 15 min) to the ranges of 99% (after 30 min). The highest removal was observed after 90 min at 99.76% which was significantly higher when compared to Cr(VI) removal after 15 and 30 min. However, the difference in % removal between 45 min and 90 min was found to be insignificant ($p > 0.05$). The variation in the extent of adsorption may be due to the fact that initially all sites on the adsorbent surface were vacant and the solute concentration gradient was relatively high. This favors a higher rate of contact-collision and subsequent binding of the ions and to the leaf powder sorbent. The leaf powder is highly packed with Cr ions per unit of surface area [11]. After 45 min, the adsorption rate became virtually constant. Conversely, this reflects the fact that all the previously vacant sites are now occupied with the Cr ions and/or complexes. No further adsorption can occur after equilibrium as the biosorbent mass is constant in the batch mode. As such, 45 min was opted as the optimal contact time.

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Table 4: The effect of contact time towards Cr(VI) removal at C_0 = 10 mg/L with biosorbent dose of 0.025 g with particle size of 151-250 µm.

Contact time (min)	Cr(VI) removal (%)
15	91.65 ± 0.21
30	99.24 ± 0.14
45	99.62 ± 0.14
60	99.61 ± 0.07
75	99.45 ± 0.16
90	99.76 ± 0.01

Note: readings are mean ± standard error (*n* = 3).

3.2 The Effect of Initial Cr(VI) Concentrations

The study was performed at an initial Cr(VI) concentrations of 1-100 mg/L with optimal conditions ascertained earlier, i.e. biosorbent dosage of 0.025 g with particle size of 151-250 µm for 45 min. The other conditions remain the same. Fig. 1 shows that the % of Cr(VI) removal increased from 95.77% at 1 mg/L to 99.47% at 10 mg/L Cr(VI).

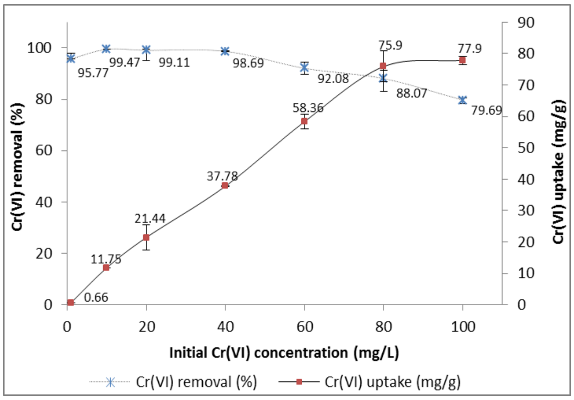

Figure 1: The effect of initial concentration of Cr(VI) towards Cr(VI) removal and uptake by *S. thaipingensis* leaf powder. Readings are mean ± standard error (*n* = 3).

This is due to the increasing number of Cr ions in the aqueous solution and the readily available binding sites on the leaf powder surface with higher contact-collision probability. However, the Cr(VI) % removal decreased from 99.11% to 79.69% with an increase in initial concentrations of Cr(VI) from 20 mg/L to 100 mg/L albeit increase in Cr(VI) uptake. It is to be noted that the number of binding sites is constant and beyond 1 mg/L, the efficiency and % removal decreases (Fig. 1) as active sites become saturated. This leads to limited surface area for further adsorption as the leaf powder dosage is kept constant. When the Cr(VI) concentration of a solution is low (i.e. 1-10 mg/L), the ratio of the available sorbent surface area to the initial number of Cr(VI) ions is larger than that in those of high Cr(VI) concentration (i.e. higher than 10 mg/L), hence at low Cr(VI) concentration, the % removal is independent of the initial concentration. On the other hand, Cr(VI) uptake is higher as the initial Cr(VI) concentration was increased from 10 mg/L to 100 mg/L, the Cr(VI) uptake also increased accordingly from 0.66 mg/g to 77.9 mg/g (79.69% removal). This was comparable or better than other reported biosorbent materials in the literature [2,3,8,11]. Morphological and elemental analysis by scanning electron microscopy (SEM) combined energy-dispersive X-ray (EDX) confirmed the uptake of Cr by the leaf powder (data not shown).

In conclusion, the leaf powder of *S. thaipingensis* is feasible to be investigated and developed further as a potential novel low-cost biosorbent material for the removal of Cr(VI) with added options for modifications such as turning into activated biocarbon, and investigating adsorption of other heavy metal ions. Besides, the tree is abundantly found, the process is environment-friendly and there is no cost involved in obtaining the tree leaf material thus favouring the economics of sustainable environmental remediation. More extensive work such as isotherm and kinetic modeling, and leaf powder characterization are being carried out in the laboratory to better understand the process and mechanisms involved.

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