Assessing The Efficacy Of Azadirachta Indica And Millettia Pinnata Leaf Powders As Low-Cost Biosorbents For Chromium (VI) Removal From Aqueous Solutions

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abstract

This study examines the efficiency of Azadirachta indica (Neem) and Millettia pinnata (Karanj) leaf powders as low-cost, eco-friendly adsorbents for removing hexavalent chromium from aqueous solutions. Batch adsorption experiments were conducted to evaluate the effect of adsorbent dosage (2–6 g/L) and initial Cr (VI) concentrations (50, 75, and 100 ppm) on removal efficiency and adsorption capacity. Results showed that removal efficiency increased with rising adsorbent doses and higher initial Cr (VI) concentrations for both biosorbents. Neem leaf powder consistently outperformed Karanj in Cr (VI) removal, achieving a maximum efficiency of 74.81% at 6 g/L for 100 ppm Cr (VI). Adsorption capacity (q_e), however, declined with increasing adsorbent dosage due to site saturation and particle aggregation effects, with maximum values observed at the lowest dose (30.08 mg/g for Neem, 27.45 mg/g for Karanj). Overall, the findings highlight the potential of using Neem and Karanj leaf powders as sustainable adsorbents for the removal of Cr (VI) from contaminated water, with implications for scalable wastewater treatment technologies for heavy metal remediation.

Keywords: *Cr (VI) removal, biosorption, Neem leaf powder, Karanj leaf powder, Adsorbent dose, batch adsorption, low-cost adsorbents.*

INTRODUCTION

Hexavalent chromium [Cr (VI)] is recognized as one of the most hazardous and persistent heavy metals present in industrial effluents, posing significant risks to both environmental integrity and human health. Widely used in industries such as electroplating, leather tanning, and textile manufacturing, Cr (VI) enters natural water bodies through runoff and leaching, often contaminating surface and groundwater systems [1,2]. Its high solubility and mobility in aqueous environments enable it to easily penetrate biological membranes, leading to adverse health consequences, including lung cancer, skin lesions, kidney and liver damage, and reproductive and developmental toxicity [3-5]. Cr (VI) is known to be both mutagenic and carcinogenic, with prolonged exposure associated with cardiovascular issues and genetic damage, which makes it a significant global concern [5,6]. Aside from its direct effects on human health, Cr (VI) contamination disturbs both aquatic and terrestrial ecosystems. It alters the microbial communities [3,5] that are essential for nutrient cycling [7,8], which in turn diminishes soil fertility and reduces plant productivity [9]. The presence of these adverse effects underscores the critical need for the expedited development and implementation of robust remediation strategies.

Conventional methods for Cr (VI) removal include chemical reduction, precipitation, ion exchange, and membrane filtration [10,11]. Despite the efficacy of these methods, they exhibit significant limitations, including elevated operational and maintenance expenditures, the generation of secondary pollutants such as hazardous sludge, and diminished performance under low-concentration scenarios. These challenges underscore the necessity for economically viable and environmentally sustainable alternatives. In this context, biosorption has emerged as a compelling, eco-friendly approach for the remediation of Cr (VI) from aqueous environments. This process leverages naturally abundant biological materials, such as plant biomass, agricultural byproducts, algae, and microbial biomass, which can adsorb heavy metals through various mechanisms, including ion exchange, surface complexation, and electrostatic interactions [12-14]. The primary benefits of biosorption are its affordability, reduced sludge generation, the ability to reuse biosorbents, and its suitability for decentralized water treatment systems [15,16].

Recent research has focused on identifying suitable biosorbents from abundant and renewable resources. In this context, leaf powders from plants such as Neem and Karanj have shown considerable potential. Neem leaves, widely known for their therapeutic qualities, are rich in functional groups like hydroxyl, carboxyl, and amino groups, which facilitate metal ion binding [17,18]. Studies have reported Cr (VI) removal efficiencies of up to 85–87% using Neem leaf powder under optimized conditions [11,19]. Similarly, Karanj leaves may have significant biosorbent potential due to their high lignin and cellulose contents, although they have been comparatively less studied.

Economically, utilizing Neem and Karanj leaf powders provides multiple benefits. These materials are readily available as agricultural by-products, require minimal processing, and reduce the dependence on expensive synthetic resins or activated carbon [20,21]. Their application additionally aids in achieving circular economy objectives by utilizing agricultural waste, thereby promoting sustainable development and local environmental sanitation initiatives [22,23]. Despite the increasing attention towards plant-based biosorbents, there is a paucity of comparative analyses that assess their performance under standardized experimental conditions. This study seeks to address this research gap by rigorously evaluating and comparing the Cr (VI) removal efficiency of leaf powders derived from *Azadirachta indica* and *Millettia pinnata*. Key parameters such as adsorbent dosage, initial metal concentration, and underlying removal mechanisms are investigated to assess their biosorptive potential. The research seeks to advance the development of low-cost, sustainable, and eco-friendly water treatment technologies for mitigating heavy metal contamination in aquatic environments.

MATERIALS AND METHODS

Collection and Preparation of Adsorbents

Azadirachta indica and *Millettia pinnata* leaves were collected from the vicinity of the Forestry Department, GGV Campus. The leaves were thoroughly washed with tap water to remove surface dust and impurities, followed by rinsing with distilled water. The cleaned leaves were sun-dried for 2 days, ground using an electric grinder, and sieved to obtain a uniform particle size of 212 µm.

The powdered material was washed 3–4 times with distilled water to remove residual color and turbidity, then oven-dried at 60 °C for 30 hours. The final adsorbent powder was stored in airtight zip-lock bags for further use without any chemical or physical modification.

Preparation of Cr (VI) Stock Solution

A stock solution of 1000 mg/L of Cr (VI) was prepared by dissolving 2.8287 g of analytical-grade potassium dichromate ($K_2Cr_2O_7$, ≥99.9% purity) in 1000 mL of distilled water. Working solutions of desired concentrations (50, 75, and 100 mg/L) were prepared by appropriate dilution with distilled water.

Batch Adsorption Experiments

Batch adsorption studies were conducted to evaluate the Cr (VI) removal efficiency. For each experiment, 100 mL of Cr (VI) solution (50, 75, and 100 mg/L) was placed in 250 mL conical flasks. Varying doses of adsorbents (2, 4, and 6 g/L) were added. The flasks were agitated using a Rotex shaker at 160 rpm for 60 minutes at room temperature. After shaking, samples were filtered through Whatman No. 42 filter paper, and the residual Cr (VI) concentration was determined spectrophotometrically at 540 nm using a UV-Visible spectrophotometer after color development (explained below).

The removal efficiency (%) was calculated using the following equation:

Removal Efficiency (%)

$$\text{Removal} = \frac{C_0 - C_e}{C_0} \times 100 \tag{1}$$

The adsorption capacity at equilibrium (q_e) was calculated using:

$$q_e = \frac{(C_0 - C_e) \times V}{m} \times 100 \tag{2}$$

Where C_o = Initial Cr (VI) concentration (mg/L), C_e = Equilibrium Cr (VI) concentration, (mg/L), V = Volume of solution (L), m = Mass of adsorbent (g), q_e = Adsorbed Cr (VI) per unit mass of adsorbent (mg/g)

RESULTS AND DISCUSSION

Effect of Adsorbent Dose on Cr (VI) Removal Efficiency

The results from batch adsorption experiments revealed a consistent increase in the removal efficiency of Cr (VI) with the rise in adsorbent doses for both Neem and Karanj leaf powders across all initial Cr (VI) concentrations (50, 75, and 100 ppm) (Figs. 1 & 2). For Neem leaf powder, at 50 ppm initial concentration, the removal efficiency increased from 9.58% at 2 g/L to 21.10% at 6 g/L. At 75 ppm, the removal efficiency improved from 49.49% to 65.91%, while at 100 ppm, it rose from 60.16% to 74.81% over the same dose range (Fig. 1). A similar trend was observed in the case of Karanj leaf powder, although the values remained comparatively lower than Neem. At 50 ppm, removal rose from 4.59% to 9.93%, at 75 ppm from 39.92% to 48.27%, and at 100 ppm from 54.90% to 60.05% as the dose increased from 2 to 6 g/L (Fig. 1).

The increase in removal efficiency of Cr (VI) with rising adsorbent doses can be attributed to multiple mechanisms. As the adsorbent concentration increases, the available surface area and active sites for adsorption also increase, enhancing the interaction between the adsorbent and Cr (VI) ions in solution [24,25]. This phenomenon is supported by the adsorption kinetics, which typically follow a pseudo-second-order model, indicating that the rate of adsorption is dependent on the number of available sites [25,26]. Additionally, the Freundlich and Langmuir isotherm models suggest that the adsorption process is favorable and can reach saturation at higher adsorbent doses, leading to improved removal efficiencies across varying initial concentrations of Cr (VI) [27,28].

Furthermore, the superior performance of Neem leaf powder over Karanj leaf powder can be attributed to several factors. Neem leaf powder has been shown to effectively follow both the Freundlich and Langmuir adsorption isotherms, indicating its strong adsorption capacity and surface heterogeneity, which enhances its ability to adsorb Cr (VI) ions from aqueous solutions [24,29]. The adsorption mechanism of Neem leaf powder is described by pseudo-second-order kinetics, suggesting that chemisorption is the rate-limiting step, which is typically more effective for heavy metal removal [24]. Additionally, Neem leaf powder is effective across a wider pH range (6 to 8), which is more conducive to the removal of Cr (VI) compared to other natural adsorbents that may require more acidic conditions [17]. Furthermore, the adsorption process involves the transfer of Cr (VI) ions onto the surface and into the pores of the Neem leaf powder, enhancing its capacity to remove these ions even at higher doses [30].

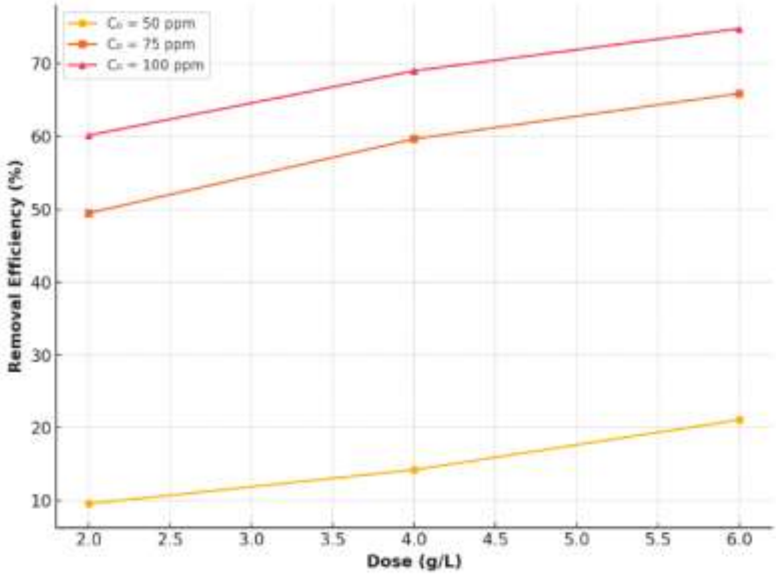

Figure 1: Effect of adsorbent dose and initial concentration on removal efficiency of Cr (VI) for Neem.

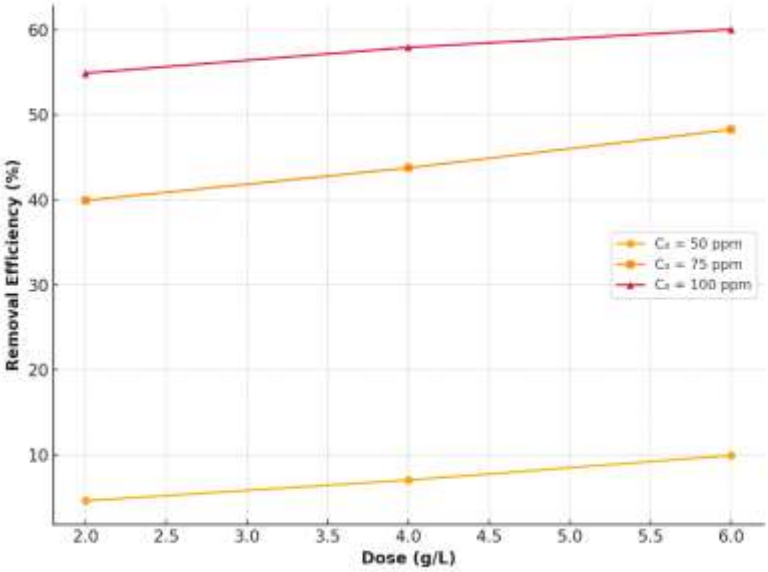

Figure 2: Effect of initial concentration and adsorbent dose on removal efficiency of Cr (VI) for Karanj.

Effect of Initial Cr (VI) Concentration on Removal Efficiency

The removal efficiency of both adsorbents increased with the rise in initial Cr (VI) concentration at a given adsorbent dose. This pattern was consistently observed across all dose levels (2, 4, and 6 g/L) for both Neem and Karanj leaf powders (Figs. 1 & 2). For Neem leaf powder, at 2 g/L dose, Cr (VI) removal increased from 9.58% at 50 ppm to 60.16% at 100 ppm. At 6 g/L, removal rose from 21.10% to 74.81% as initial concentration increased from 50 to 100 ppm (Fig. 2). Similarly, for Karanj leaf powder, removal efficiency at 2 g/L improved from 4.59% (50 ppm) to 54.90% (100 ppm), and at 6 g/L, from 9.93% to 60.05% (Fig. 2).

The higher removal efficiency at elevated Cr (VI) concentrations may be attributed to several interrelated factors. Both Neem and Karaj leaf powders exhibit effective adsorption characteristics, primarily following the Freundlich and Langmuir isotherms, which indicate a heterogeneous surface with varying affinities for Cr (VI) ions [24,29]. Furthermore, the adsorption kinetics closely follow a pseudo-second-order model, indicating that the rate of Cr (VI) uptake is primarily dependent on the number of available active sites on the adsorbent surface [24,31]. At higher concentrations, the increased concentration gradient serves as a stronger driving force for mass transfer, thereby enhancing the interaction between Cr (VI) ions and the functional groups (such as hydroxyl, carboxyl, and phenolic groups) present on the adsorbent surface [30,32]. Additionally, the speciation of Cr (VI) in aqueous media into various oxyanionic forms (e.g., $HCrO_4^-$, $Cr_2O_7^{2-}$, CrO_4^{2-}) facilitates more effective electrostatic and ligand-exchange interactions with the adsorbent, further improving the overall removal efficiency [32].

Effect of Adsorbent Dose on Adsorption Capacity

A reverse trend was observed with an increasing dose of the adsorbent. As the dose escalated from 2 g/L to 6 g/L, the equilibrium adsorption capacity (q_e) exhibited a noticeable decline (Figs. 3 & 4). Specifically, for Neem powder, q_e decreased from 30.08 mg/g at 2 g/L to 12.05 mg/g at 6 g/L (Fig. 3). Similarly, Karanj's q_e values diminished from 27.45 mg/g to 10.01 mg/g across the same dose range (Fig. 4). The decrease in adsorption capacity with increasing biosorbent dose (2 g/L, 4 g/L, and 6 g/L) can be attributed to several interrelated mechanisms. As the biosorbent concentration increases, the available surface area for adsorption becomes saturated, reducing the effective interaction between the biosorbent and the metal ions due to overcrowding and steric hindrance [33]. Additionally, the presence of multilayer adsorption can hinder the accessibility of active sites, as the formation of hindered layers can limit the effective surface area available for further adsorption [34]. Furthermore, the size and characteristics of the biosorbent, such as the extracellular polymeric substances, can influence the biosorption mechanisms, with larger particles exhibiting predominantly physical adsorption, which is less effective than chemical binding [35]. Lastly, the physicochemical properties of both the biosorbent and the heavy metal ions, including their interactions and the conditions of the adsorption environment, play a crucial role in determining the overall adsorption efficiency [36].

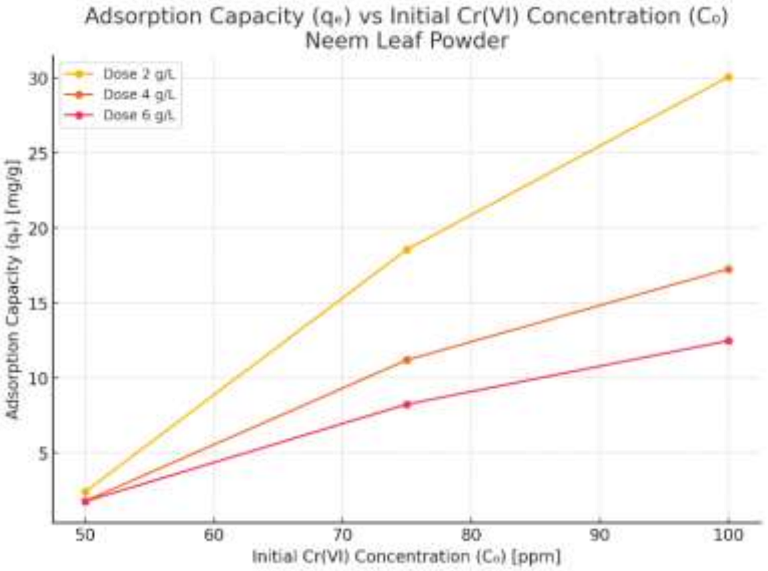

Figure 3: Effect of initial concentration and adsorbent dose on adsorption capacity of Neem

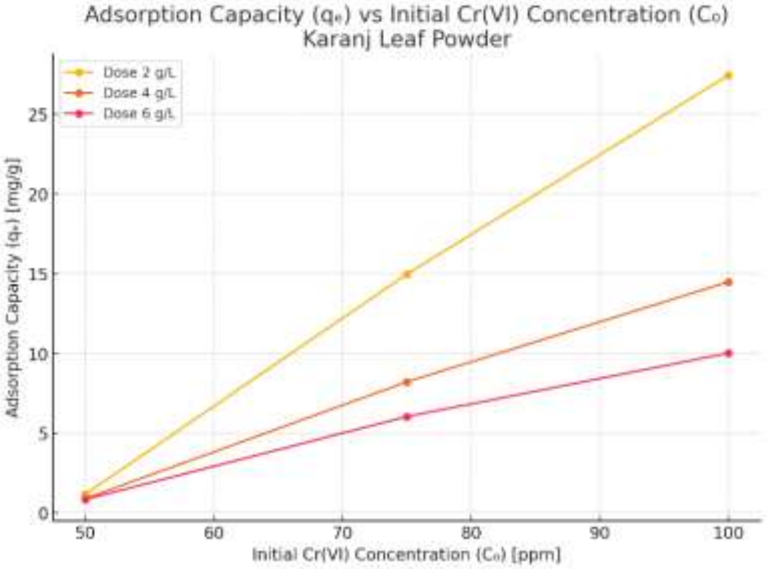

Figure 4: Effect of initial concentration and adsorbent dose on adsorption capacity of Karanj

Effect of Initial Cr (VI) Concentration on Adsorption Capacity

The adsorption capacity of both Neem and Karanj leaf powders was significantly affected by the initial Cr (VI) concentration (Figs. 3 & 4). Experimental data indicated a clear trend: increasing C_0 from 50 to 100 ppm resulted in an increase in adsorption capacity at all dose levels (2, 4, and 6 g/L). For instance, Neem leaf powder at 2 g/L dose exhibited an increase in q_e from 2.40 mg/g (C_0 = 50 ppm) to 30.08 mg/g (C_0 = 100 ppm) (Fig. 3). Similarly, Karanj leaf powder showed a rise in q_e from 1.15 mg/g to 27.45 mg/g over the same C_0 range (Fig. 4). The increasing trend in adsorption capacity at all dose levels with initial Cr (VI) concentration can be attributed to several underlying mechanisms. As the initial concentration of Cr (VI) rises, the driving force for mass transfer increases, enhancing the likelihood of Cr (VI) ions interacting with the adsorbent surfaces, which is supported by findings that show higher adsorption capacities with increased initial concentrations [37-39]. Additionally, the presence of more Cr (VI) ions allows for greater saturation of available active sites on the adsorbents, such as crosslinked cationic konjac glucomannan and carbon nanotubes, which exhibit a Langmuir isotherm behavior indicating monolayer adsorption on a surface with a finite number of identical sites [40,41]. Furthermore, the adsorption kinetics suggest that the process is rapid and can be influenced by both chemisorption and physisorption mechanisms, which also contribute to the observed increase in capacity with higher initial [39].

CONCLUSION

The present study demonstrates that Neem and Karanj leaf powders are effective biosorbents for Cr (VI) removal, with performance improving at higher adsorbent dosages and initial Cr (VI) concentrations. Neem leaf powder showed superior Cr (VI) adsorption, likely due to its higher density of active binding sites. These findings suggest that plant-based materials such as Neem and Karanj leaves can serve as cost-effective, sustainable adsorbents in the treatment of heavy metal-contaminated water, particularly in areas affected by industrial chromium pollution. Further studies involving kinetic and isotherm modeling are recommended to understand the underlying adsorption mechanisms better.

Conflict of Interest: There are no conflicts of interest declared by the authors.

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