Fluoride Adsorption Behavior in Cost-effective Plant-based Bio-sorbents

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Editor's note: The detrimental effects of excessive fluoride on the health of both humans and animals through drinking-water sources are evident, particularly regarding dental and skeletal fluorosis. This underscores the critical need for stringent control of fluoride concentration levels in drinking water. In their study, Visarapu et al. presented various plant-based biosorbents for fluoride removal, and their results demonstrated that Hibiscus leaves achieved a high fluoride removal efficiency with an impressively low dosage of just 1 g.

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Communication

Fluoride Adsorption Behavior in Cost-effective Plant-based Bio-sorbents

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Abstract

Concerns regarding the effects of excessive fluoride in water sources on human health have been raised globally. Conventional fluoride remediation approaches generally involve chemical treatment or expensive filtration systems that can introduce additional pollutants, leading to environmental implications. The utilization of plant-based biosorbent materials for fluoride removal holds promise as a sustainable approach to tackle the issue of fluoride in water sources. Utilizing these materials can lead to significant environmental benefits, including reduced toxicity and improved human health outcomes, ultimately contributing to a greener and more sustainable future. This work explores the potential of various plant-based biosorbents for fluoride removal, presenting a sustainable and environmentally friendly approach. The biosorption tests were executed and measured the factors of initial fluoride concentration, contact time, pH levels, and biosorbent dose on fluoride removal bio-sorption efficiencies. At a fluoride concentration of 10 mg/L, all the leaf powder biosorbents demonstrated remarkable sorption efficiencies, achieving removal rates ranging from 90% to 100% at pH 6 and a contact time of 60 minutes. Notably, Hibiscus leaves achieved high fluoride removal efficiency with a remarkably low dose of just 1 g. This study highlights the benefits of exploring plant-based biosorbents as more environmentally sustainable solutions to removing fluoride and other contaminants, which will result in improving human health while also being more environmentally friendly.

Keywords: Fluoride adsorption; Plant-based biosorbents; Cost-effectiveness, Sorption efficiency; Sustainable approach.

1. Introduction

Fluoride is an essential trace element that plays a crucial role in maintaining dental health and preventing tooth decay. Excessive fluoride impacts on the health of humans and animals via drinking-water sources demonstrated negative health impacts, most particularly dental and skeletal fluorosis, underscoring the necessity for tight control of fluoride concentration levels in drinking-water sources [1].

Continuous access to excessive fluoride can lead to skeletal fluorosis, a severely debilitating condition characterized by abnormalities of bone and joints, reinforcing the importance of fluoride being removed from drinking water on a global scale [2-4]. While techniques of coagulation, precipitation, and ion

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exchange can significantly reduce fluoride concentrations, these techniques employ chemicals and depict complicated techniques that are potentially costly and, more importantly, unsustainable from an environmental context, especially in environments with resource limitations [5, 6]. This highlights the urgent requirement for innovative, cost-effective, and eco-friendly fluoride removal strategies. Adsorption technology stands out due to its affordability, design simplicity, and ease of operation and maintenance. There are a number of factors that influence the efficiency of adsorption, which include contaminants and absorbent concentration, the surface characteristics of the adsorbent, pH of solution, and contact time [7].

In recent years, plant-based biosorbents have emerged as favorable candidates for fluoride remediation in water. Natural adsorbents, derived from various plant materials, agricultural waste, and naturally occurring minerals, possess inherent adsorption properties that make them attractive for water treatment applications [8, 9]. These adsorbents demonstrate notable selectivity, substantial adsorption capacities, and facile regenerability, rendering them viable candidates for efficient fluoride removal applications. Furthermore, these adsorbents are abundant, renewable, biodegradable, and present minimal risks to human health and the environment. Adsorption of fluoride ions on solid adsorbent occurs via diffusion of ions from solution across the boundary layer, followed by transfer of these ions to the internal surface for porous materials [10-15].

This manuscript aims for fluoride removal by plant-based biosorbents. It will explore the underlying mechanisms of fluoride adsorption onto these materials and highlight their potential advantages over traditional methods. Various plant-derived biosorbents, including tamarind leaves, custard apple leaves, betel leaves, orange peels, hibiscus leaves, mango leaves, curry leaves, moringa leaves, guava leaves, insulin leaves, grape tree leaves and rose petals have been explored for fluoride removal at pH 6, as reported in previous research [16]. Furthermore, this manuscript will comprehensively examine the key parameters that modulate the adsorption capacity and efficacy of these materials, including pH levels, contact duration, and adsorbent concentration.

The potential challenges and limitations associated with the implementation of plant-based biosorbents for large-scale fluoride removal will be addressed, along with strategies to overcome them. Additionally, the manuscript will shed light on the economic feasibility and sustainability aspects of utilizing these materials for water treatment purposes.

The insights provided in the study represent a key step forward in understanding fluoride removal technologies and provide an applicable reference for researchers, environmental engineers, and policy makers seeking effective, sustainable ways to remediate fluoride contamination of water sources and thereby provide access to safe drinking water on a global scale.

2. Experimental

2.1. Materials

Sodium Fluoride, SPADNS reagent, and Zirconyl Chloride were purchased from Merck. HCl, NaOH were purchased from Alfa Aesar. Naturally occurring and abundantly available low-cost materials such as Tamarind leaves, Custard apple leaves, betel leaves, orange peels, Hibiscus leaves, mango leaves, Curry leaves, Moringa leaves, Guava leaves, Insulin leaves, Grape tree leaves, rose petals were collected from the local market surrounding Hyderabad, India. All the leaf powders were dried until the water content was reduced to less than 5%. The dried leaf powders were then ground, and the powder smaller than 60 mesh was collected.

2.2. Fluoride stock preparation

A stock solution was made by dissolving 2.21 g of anhydrous sodium fluoride in 100 mL of double-distilled water and then diluting to 1000 mL. Fresh working solutions of 10 mg/L fluoride were prepared from the stock solution, as required. The pH was adjusted to 6 with 0.5 N HCl and 0.5 N NaOH solutions.

2.3. Batch biosorption studies

Biosorption investigations were performed by changing the biosorbent dosage to 1g, 2g, and 3g of 50 mL of 10 mg/L fluoride solutions with a contact time of 60 minutes at pH 6 and room temperature in a 100 mL Erlenmeyer flask under mixing conditions at 150 rpm on a rotary.

UV-vis spectrophotometric analysis of the fluoride ion in solution was carried out using the SPADNS reagent method. 958 mg of standard SPADNS reagent was prepared by dissolving the reagent in 500 mL of

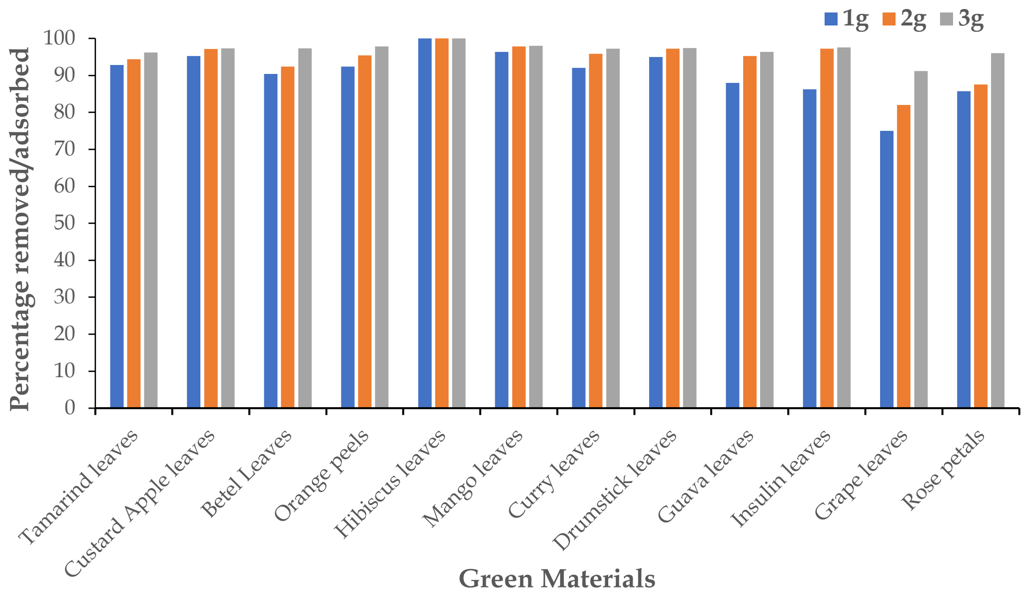

Figure 1. Percentage of fluoride removal/adsorption by various plant-based biosorbents determined by SPADNS method at 570 nm.

double-distilled water (DDW) and kept in the dark from sunlight. The zirconyl acid reagent was prepared by dissolving 133 mg of zirconium chloride in 25 mL of DDW and 350 mL of 1N HCl with distilled water made up to 500 mL. Then, equal volumes of the SPADNS reagent and the zirconyl acid reagent were combined to prepare a reference for the solution. Table 1 summarizes the operating conditions for Fluoride ion adsorption.

Table 1. Summary of operating conditions for Fluoride ion adsorption

Parameter	Operating condition
pH	6
Contact time	60 min
Biosorbent dosage	1g, 2g, 3g
Initial Fluoride ion concentration	10 mg/L

Biosorption tests were conducted in the manner of shaking 50 mL of fluoride solution with several biosorbent doses (1g, 2g, and 3g) in an orbital shaker for 60 minutes. The solids were then separated from the solution by filtration through Whatman 42 filter paper, and the solution adjusted to 20 mL. The fluoride concentration was determined using a UV-Visible Spectrophotometer (Shimadzu) utilizing acid-zirconyl SPADNS reagent at 570 nm. Fluoride removal percentage and adsorption efficiency were then calculated. Three replicates were conducted, and the average values are shown. Control experiments without biosorbent were conducted to ensure that good detection was exclusive to biosorbent. The percentage removal and adsorption efficiency of biosorbents for fluoride were calculated according to the equations.

$$Q_e = ((C_i - C_e))/m\,V \quad (1)$$

$$\text{Sorption efficiency (\%)} = ((C_i - C_e))/C_i \times 100 \quad (2)$$

where Q_e represents the amount of fluoride ions adsorbed at equilibrium (mg/g), C_i and C_e denote the initial and equilibrium fluoride concentrations (mg/L), m is the mass of the adsorbent (mg), and V is the volume of the solution (mL).

3. Results and discussion

The equilibrium sorption results were determined at pH 6, a contact time of 60 minutes, and a fluoride concentration of 10 mg/L and are shown in Figure 1. As the biosorbent dose increased (1g, 2g, and 3g), fluoride removal efficiency also increased, with the highest removal efficiency obtained at 3g of biosorbent dose. This indicates that at 3g, the biosorbent had enough active sites for efficient fluoride binding. The increased

Table 2. Comparative Analysis of Fluoride Removal Percentages of Various Biosorbents

Biosorbent	Dose of Adsorbent (mg/L)	pH	Removal Percentage (%)	Reference
Lemon peel	20	5	48.36	[17]
H-modified tea biochar	500	2	93.31	[18]
Moringa oleifera seed powder	10	6.5	97.2	[19]
Grape pomace	10	3	96.13	[20]
Bivalve shell	3	5.5	97.26	[21]
Terminalia Chebula	4	7	98	[22]
Insulin leaves	10	6	97.6	Present work
Orange peels	10	6	97.8	Present work
Mango leaves	10	6	97.98	Present work
Hibiscus leaves	10	6	100	Present work

biosorption efficiency at higher biosorbent dosages may be attributed to less flocking and increased availability of pores, surface area, and active binding sites, resulting in more fluoride ions binding. Table 2 provides a comparative summary of fluoride removal percentages for different biosorbents.

These different leaf powders exhibit varying levels of removal performance when used for Fluoride removal at 10mg/L. The range of removal performance depends on the chemical constituents, properties, and characteristics of materials and the contaminant being targeted as well [23-25]. The results showed that all biosorbents used in the experiments have a good to high affinity for adsorbing/removal of Fluoride onto their surfaces. The results indicate that hibiscus leaves are highly effective for fluoride removal, with a remarkable 100% removal rate achieved at a low dose of 1g. The minimum removal performance was observed with 75% of grape tree leaves at 1g. The range of performance, i.e., low (85-90%), medium (90-95%), and high (95-100%), by these biosorbents was also presented here in Figure 2.

This study assessed the effectiveness of different plant-derived biosorbents for fluoride removal using UV-Vis spectrophotometry (SPADNS method) at 570 nm. There was a noticeable increase in removal efficiency; first with increasing biosorbent dosage (1g to 2g and 2g to 3g); second, in that 9 materials had a minimal increase (0-5%) in removal efficiency for each subsequent dosage level, and 2 materials had a moderate increase (5-10%). The comparison of the lowest and highest dosage level (1g-3g) highlighted differences: 5 materials had increases in removal efficiency in the range of 5-10%, 4 materials with 10-15%, and 2 materials in the range of 15-20%. Grape tree leaves had the highest

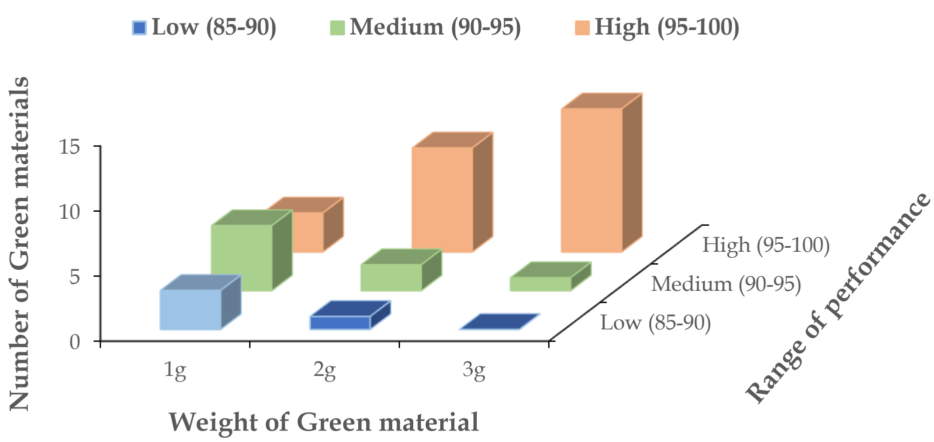

Figure 2. The range of performance, i.e., low (85-90%), medium (95-100%), and high (95-100%) by weight 1g, 2g, 3g of various leaf powders, with dose determined by SPADNS method with UV-Vis spectrophotometry at 570nm.

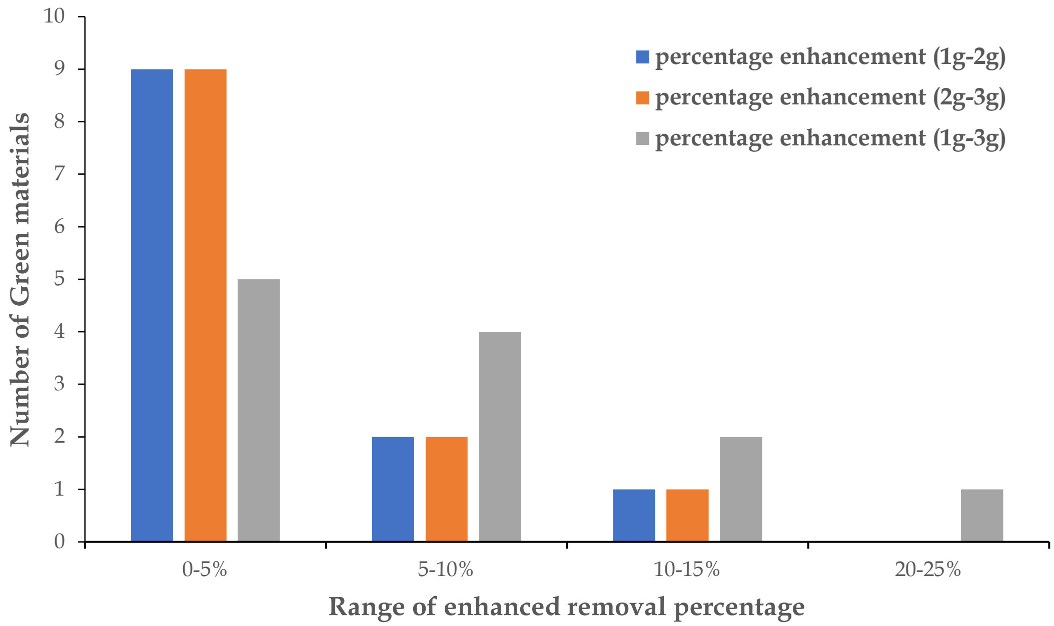

Figure 3. Enhancement in removal percentage from 1g-2g, 2g-3g, 1g-3g of fluoride ion by various leaf powder materials determined by SPADNS method with UV-Vis spectrophotometry at 570nm.

increase in removal efficiency, having a 22% increase in removal efficiency (75%-97%) from the increase of dosage of 1g to 3g; demonstrating the importance of biosorbent dosage on fluoride removal (Figure 3).

4. Conclusions

Batch experiments were conducted to explore the biosorption of fluoride ions using 12 distinct plant-based biosorbent materials, examining the effects of key operating parameters such as pH, contact time, adsorbate concentration, and biosorbent dosage. The studies evaluated the potential of diverse biosorbent materials in removing fluoride ions from water. This study has shown that biosorption is significantly affected by biosorbent dose. The data suggested that the optimum removal of fluoride ions was at pH 6, and a concentration of 10 mg/L of fluoride was achieved in 60 minutes of contact time. Nevertheless, hibiscus leaves displayed high removal efficiency at only 1 g biosorbent dose. Most of the plant-based biosorbent materials exhibited good to excellent removal efficiency, and some of them showed an up to 100% adsorption efficiency. The biosorbents were environmentally friendly, practical, and inexpensive, thereby presenting an efficient option for removing fluoride ions from water.

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Conflict of Interest

The authors declare no conflict of interest.

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