

Defluoridation of Water Using Low Cost Bioadsorbents

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ABSTRACT

To meet drinking water standards, fluoride levels must be kept below 1 mg/L to prevent fluorosis. This study evaluates low-cost bio adsorbents; cashew shell powder, coconut shell powder, coconut shell crystal, and hydrothermally treated bamboo shoot powder; using batch adsorption at pH 4.4-4.9 and contact times of 1.5–3 hours. 3.5 g coconut shell powder and a mixture of 1.5 g coconut shell powder and 1.5g of cashew shell powder achieved 10 % removal, reducing fluoride from 10 ppm to 9 ppm. Both 2 g coconut shell crystal with 1 g cashew shell powder and bamboo shoot powder dosages of 2–6 g achieved 30 % removal (10 ppm to 7 ppm), while higher bamboo shoot dosages (8–10 g) showed lower efficiency due to supersaturation. The crystal form of coconut shell and bamboo shoot carbon are the most effective adsorbents, and it was found that dosage, pH, and contact time influence the Defluoridation efficiency. These bioadsorbents can be used for community level defluoridation using affordable, locally available materials.

Keywords: Coconut shell, Cashew shell, Bamboo shoot

I. INTRODUCTION

Water is necessary for the sustenance of life. However, most countries now face drinking water problems. Due to the presence of various pollutants like nitrates, iron and fluoride drinking water has become contaminated. 17 out of the 29 states of India report issues due to such contaminated water. Fluoride is considered a "double-edged sword" because while its deficiency can lead to dental cavities its excessive intake can result in dental and skeletal fluorosis [5]. Fluoride ions bond with the calcium ions in teeth and bones due to their strong electronegativity which can lead to fluorosis of bones and teeth. The Bureau of Indian Standards (BIS) specifies that the desirable fluoride concentration in drinking water is 0.6 mg/L to 1 mg/L [3]. In Kerala, endemic fluoride contamination has been reported especially in the districts of Palakkad and Alappuzha. In India, fluoride contamination is common in states such as Jammu & Kashmir, Punjab, Uttar Pradesh, Rajasthan, Gujarat, Odisha, Karnataka, Andhra Pradesh, Kerala, and Tamil Nadu [8]. On average fluoride content found in the Earth's crust is about 300 mg/kg [6]. It is found in a variety of minerals such as fluorspar, cryolite, rock phosphate, apatite, hornblende, mica, and others. Groundwater is also contaminated by Industrial wastewater discharges from semiconductor, aluminum, and glass manufacturing industries [7]. Recently, biosorption has emerged as an effective and attractive technique for fluoride removal [4]. This method uses low-cost adsorbents such as rice husk, sawdust, Moringa oleifera extract, coconut shell, bamboo shoot, etc. The objective of this study is to investigate the effectiveness of naturally occurring and low-cost materials like coconut shell, cashew shell, and bamboo shoot for defluoridation of water. The efficiency and sustainability of fluoride removal using these bioadsorbent were also investigated. The effect of controlling factors like pH, contact time, and adsorbent dosage on fluoride removal efficiency was also studied.

II. DEFLUORIDATION

Fluoride is a form of fluorine which resemble hydroxide ion in terms of charge and size. Fluoride salts and minerals are used in the production of hydrogen fluoride for fluorocarbons. Fluoride is found on Earth in several mineral forms, particularly fluorite. Fluoride salts are colourless but has a distinctive bitter taste.



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A. OCCURRENCE

Fluoride is the 13th most abundant element in the Earth's crust and is widely dispersed in nature in its mineral form, almost entirely in the form of fluorides in rocks and soil. Many fluoride-bearing minerals are known, but fluorite is of the most commercial importance due to its abundance as it is 49% fluoride by mass. It is naturally found in water around the globe.

B. REMOVAL OF FLUORIDE

When fluoride comes in contact with mineralizing tissues it disrupts the normal enamel formation in teeth and causes fluorosis [5]. If left untreated it can extend to the junction between dentin and enamel particularly during the early stages of tooth development. Providing water with an optimal fluoride concentration is the only way to fully protect future generations from the disease [3]. This can be achieved by removing fluoride from water (defluoridation), identifying alternative safe water sources, or transporting safe water from distant locations. Researchers at the University of Bordeaux in France studied 3,578 people aged 65 and above and found an increased risk of hip fractures when drinking water fluoride levels exceeded 1.8 mg/L.

III. METHODOLOGY

A. SAMPLE COLLECTION

To determine the fluoride content in various water sources such as lake water, seawater, bottled water, and tap water, the samples were collected from various locations. Lake water sample was obtained from Thanni Lake, Fig. 1 shows the collection of a water sample from the lake. Seawater from Varkala Beach, tap water from Kollam Municipality and a bottled water was also obtained for the study.



Fig. 1. Collection of water sample from lake

B. FLUORIDE TEST

Fluoride determination tests were conducted on these collected samples. The principle used in the test is visual analysis. The detailed procedure is given below:

1) Materials

The equipment's needed include Nessler tubes, hot plates and a UV-Visible spectrometer. The glassware needed include standard flasks and measuring jars. Several chemicals like Sodium Thiosulphate Solution (0.1 N), Standard Sodium Fluoride Solution and Zirconium Alizarin Reagent are also needed and their preparation is given below.

Sodium Thiosulphate Solution (0.1 N) is made by dissolving 12.4 g of sodium thiosulphate in distilled water and made up the solution to 500 ml. Standard Sodium Fluoride Solution is made by Dissolve 0.221 g of sodium fluoride in distilled water and making up to 1000 ml solution. Then 100 ml of this is diluted to 1000 ml. Each ml of the final solution contains 0.01 mg fluoride. Store in plastic bottles. Zirconium Alizarin Reagent is made by dissolve 0.3 g zirconium oxychloride (or 0.25 g zirconium oxynitrate) in 50 ml water. Dissolve 0.07 g alizarin S and add it slowly to the zirconium solution with stirring until clear in 50 ml water. Dilute 1.12 ml concentrated HCl to 500 ml. Separately, add 37 ml concentrated H₂SO₄ to 400 ml water and make up to 500 ml. Cool both and mix. Add the clear zirconium solution to the acid mixture and make up to 1000 ml with water. The reagent turns orange-yellow in an hour and is ready to use. The mixture can be kept in a dark bottle in the fridge. It will stay good for 2-3 months. When 5 ml of this reagent is added to 100 ml of fluoride free water, it turns pink. Fluoride changes the colour to yellowish.

2) PROCEDURE

Take 100 ml of the clear sample and prepare standard fluoride solutions in 100 ml of distilled water using Nessler tubes. Then add 5.0 ml of zirconium alizarin reagent to each tube Fig. 2 shows Zirconium reagent. Ensure all tubes are at the same temperature (around 1–2°C range) during the test. After 1 hour, compare the colour of the test solution with the standards. If fluoride is above 1.0 mg/l, dilute the sample and repeat. Colour can also be checked at 570 nm using a spectrophotometer.



Fig. 2. Zirconium reagent

a) Observation to be recorded

Compare the colour of the sample with that of the standard fluoride solutions visually. Fig. 3 shows the visual analysis of samples.



Fig. 3. visual analysis of samples.

3) DEFLUORIDATION USING LOW-COST ADSORBENTS

Defluoridation can be defined as bringing down the fluoride level in water to optimal level. Using adsorbents is a conventional, cost effective and widely tested method to provide safe water to communities affected by fluorosis. Providing water with optimal fluoride concentration is the only effective way to protect future generations from this disease. This can be achieved through removing fluoride using suitable techniques, identifying alternative safe water sources or transporting water from safe, distant sources. Out of these, using adsorbent for fluoride removal is a widely tested cost-effective method for providing safe drinking water. The six main categories of defluoridation techniques are precipitation methods, ion exchange processes, membrane filtration processes, nano-filtration, electro-coagulation and adsorption [8].

Our study selected adsorption from available techniques due to its high efficiency, cost-effectiveness, and availability. We found that fluoride adsorption increased with greater bed depth, with optimal removal efficiency at 4 cm, where treated water turbidity was within drinking water limits. This was due to the increased surface area and active sites when the adsorbent bed depth was increased. The effect of adsorbent bed depth on fluoride removal was studied with a constant initial fluoride concentration of 10 mg/l. Beyond our findings, the adsorption method offers multiple advantages over others. It provides an efficiency ranging from 50% to 96%, compared to a maximum of around 70% in other processes [1]. It is also the most economical, requires minimal maintenance, and allows safe disposal, used adsorbents can be repurposed as filler material.

IV. METHODS AND MATERIALS USED

In this study naturally occurring and vastly available materials such as cashew shell, coconut shell and bamboo shoot were tested for their ability to act as an effective bioadsorbent for defluoridation.

A. COCONUT SHELL POWDER

1) Preparation of Coconut Shell Powder

Coconuts were collected from Mayyanad, Kollam. The coconut fiber was removed manually; the shells were dried for one month and then burned until carbon crystals were formed. The resulting char was powdered and passed through an IS sieve of 600 microns. Fig. 4 shows the activated coconut shell.



Fig. 4. Activated coconut shell

2) Preparation of Standard Fluoride Solution

From a 1000 ppm standard fluoride solution, a 10 ppm standard fluoride solution was prepared for the experiment. Then, 3.5 g of coconut shell powder was added under a pH of 4.4 and kept for a contact

period of 1.5–3 hours. After the contact time, the solution was filtered, and the fluoride content was determined.

3) *Test to Determine the Fluoride Content*

Take 100 ml of the clear sample and prepare standard fluoride solutions in 100 ml of distilled water using Nessler tubes. Add 5.0 ml of zirconium alizarin reagent to each tube. Ensure all tubes are at the same temperature (around 1–2°C range) during the test. After 1 hour, compare the colour of the test solution with the standards. If fluoride is above 1.0 mg/l, dilute the sample and repeat. Colour can also be checked at 570 nm using a spectrophotometer.

B. *CASHEW SHELL POWDER AND COCONUT SHELL POWDER*

1) *Preparation of Coconut Shell Powder*

Coconuts were collected from Mayyanad, Kollam. The coconut fiber was removed manually; the shells were dried for one month and then burned until carbon crystals were formed. The resulting char was powdered and passed through an IS sieve of 600 microns. Fig 4. shows the activated coconut shell.

2) *Preparation of Cashew Shell Powder*

Cashews were collected from Mayyanad, Kollam. The nuts were removed manually, and the shells were dried for one month, then burned until carbon crystals were formed. The char was powdered and passed through an IS sieve of 450 microns. Fig. 5 shows the activated cashew shell powder.



Fig. 5. Activated cashew shell powder

3) *Preparation of Standard Fluoride Solution*

A 10 ppm solution was prepared using the 1000 ppm standard fluoride solution. Then, 1.5 g of the coconut shell powder and 1.5 g of the cashew shell powder were added under a pH of 4.4 and kept for a contact period of 1.5–3 hours. The solution was filtered and tested for fluoride content after the contact time was over.

4) *Test to Determine the Fluoride Content*

Take 100 ml of the clear sample and prepare standard fluoride solutions in 100 ml of distilled water using Nessler tubes. Add 5.0 ml of zirconium alizarin reagent to each tube. Ensure all tubes are at the same temperature (around 1–2°C range) during the test. After 1 hour, compare the colour of the test solution with the standards. If fluoride is above 1.0 mg/l, dilute the sample and repeat. Colour can also be checked at 570 nm using a spectrophotometer.

C. COCONUT SHELL CRYSTAL AND CASHEW SHELL POWDER

1) Preparation of Coconut Shell Crystal

Coconuts were collected from Mayyanad, Kollam. The fiber was manually removed, and the shell was dried for one month. It was then burned to form carbon crystals and passed through a 4.75 mm IS sieve.

2) Preparation of Cashew Shell Powder

Cashews were collected from Mayyanad, Kollam. The nuts were removed manually, and the shells were dried for one month, then burned until carbon crystals were formed. The char was powdered and passed through an IS sieve of 450 microns. Fig. 6 shows the adsorbents used.



Fig. 6. shows the adsorbents used.

3) Preparation of Standard Fluoride Solution

A 10 ppm solution was prepared from a 1000 ppm standard fluoride solution. Then, 2 g of coconut shell crystal and 1 g of cashew shell powder were added under a pH of 4.8 and kept for a contact period of 1.5–3 hours. The solution was filtered and tested for fluoride content After the contact time.

4) Test to Determine the Fluoride Content

Take 100 ml of the clear sample and prepare standard fluoride solutions in 100 ml of distilled water using Nessler tubes. Add 5.0 ml of zirconium alizarin reagent to each tube. Ensure all tubes are at the same temperature (around 1–2°C range) during the test. After 1 hour, compare the colour of the test solution with the standards. If fluoride is above 1.0 mg/l, dilute the sample and repeat. Colour can also be checked at 570 nm using a spectrophotometer.

D. BAMBOO SHOOT POWDER

1) Preparation of Bamboo Shoot Powder

Fresh bamboo shoots were obtained from Chathanoor, Kollam. The shoots were peeled then cut into pieces and dried at 70°C overnight. The dried material was then ground into powder and used as the base for preparing porous N-doped carbon via hydrothermal treatment. In a typical procedure, 2 g of dried bamboo shoot powder was combined with 18 ml deionized water and 0.5 ml concentrated sulfuric acid and then the mixture was stirred for 30 minutes. The mixture was transferred to a Teflon-lined Muffle Furnace and treated at 180°C for 5.5 hours to form hydrochars. The dark brown solids obtained were first filtered and then washed with distilled water, after that it was dried at 25°C. These were post-carbonized at 750°C, 850°C, and 950°C in a nitrogen atmosphere for 2 hours, then cooled under vacuum for 24 hours. The dried bamboo shoots were also calcined at 850°C under nitrogen for comparison. Fig.

7 shows the Steps involved in preparation of bamboo shoot powder and Fig. 8 to Fig. 11 show the different stages of bamboo shoot preparation and Fig. 12 shows the muffle furnace.

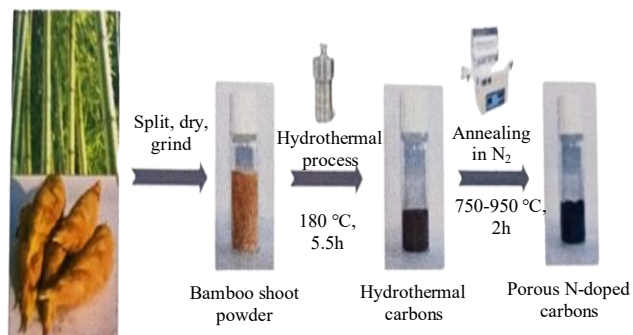


Fig. 7. Steps involved in preparation of bamboo shoot powder.

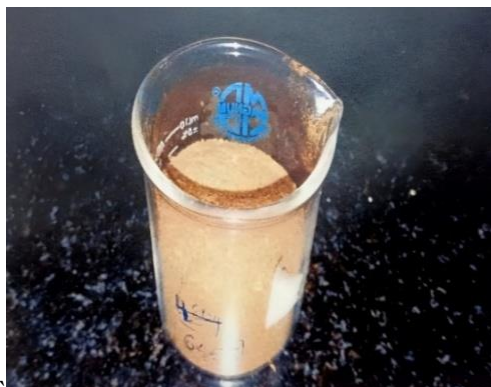


Fig. 8. Powder form oven dried bamboo shoot.



Fig. 9. Bamboo shoot in Con. Sulphuric acid



Fig. 10. Filtering the bamboo shoot after acid treatment



Fig. 11. Bamboo shoot after acid treatment



Fig. 12. Muffle Furnace

2) Preparation of Standard Fluoride Solution

A 10 ppm solution was prepared using the 1000 ppm standard fluoride solution. Then, 2 g, 4 g, 6 g, 8 g, and 10 g of bamboo shoot powder were added under a pH of 4.8 and kept for a contact period of 1.5–3 hours. The solution was filtered and tested for fluoride content after the contact time was over.

3) Test to Determine the Fluoride Content

Take 100 ml of the clear sample and prepare standard fluoride solutions in 100 ml of distilled water using Nessler tubes. Add 5.0 ml of zirconium alizarin reagent to each tube. Ensure all tubes are at the

same temperature (around 1–2°C range) during the test. After 1 hour, compare the colour of the test solution with the standards. If fluoride is above 1.0 mg/l, dilute the sample and repeat. Colour can also be checked at 570 nm using a spectrophotometer. Fig. 13 shows the activated bamboo shoot adsorbents in the samples.



Fig. 13. Adsorbents in samples

V. RESULTS AND DISCUSSION

The following results were obtained from our study,

A. Fluoride content in collected samples

The fluoride content in sea water is found to be 1.5 ppm. Since the fluoride content is within the permissible limit (0.5-1.5ppm), the fluoride concentration is sufficient in the sample. Similarly, the fluoride content in lake water is 0.5 ppm. Since the fluoride content is within the permissible limit (0.5-1.5ppm), the fluoride concentration is sufficient in the sample. But, the fluoride content in tap water is only 0.1 ppm- Since the sample is deficient in fluoride content it may cause fluorosis so it is very essential to add additional fluoride (fluoridation). The bottled water did not show any fluoride content. Since the sample is devoid of fluoride content it will cause fluorosis so it is very essential to add additional fluoride (fluoridation). Table 1 shows the fluoride level in different water samples.

Table 1. Fluoride level in different water samples

Sl. No	Sample	Fluoride content (ppm)
1	Sea water	1.5
2	Lake water	0.5
3	Tap water	0.1
4	Bottled water	Nil

B. Coconut shell powder

A water sample with an initial fluoride concentration of 10 ppm was treated using coconut shell powder as the adsorbent. The test was conducted at pH values between 2 and 6, using 3.5 grams of the adsorbent for a contact time of 1.5 to 3 hours. The final fluoride content was found to be 9ppm.

C. Cashew shell powder and Coconut shell powder

The same procedure was repeated using a mixture of cashew shell powder and coconut shell powder (1.5 g each). The pH values between 2 and 6, and contact time remained between 1.5 and 3 hours. The final fluoride content was found to be 9ppm.

D. Coconut shell crystal and Cashew shell powder

This time, a mixture of coconut shell crystal (2 g) and cashew shell powder (1 g) was used. The test conditions were similar: pH values between 2 and 6, initial fluoride concentration 10 ppm, and contact time = 1.5 to 3 hours. The final fluoride content was found to be 7ppm. Table 2 shows the Fluoride adsorption by coconut shell and cashew shell.

Table 2. Fluoride adsorption by coconut shell and cashew shell

Sl. No	Sample	Weight (g)	pH	Initial fluoride content (ppm)	Final fluoride content (ppm)
1	Coconut shell powder	3.5	4.4	10	9
2	Coconut shell powder + cashew shell powder	1.5 + 1.5	4.8	10	9
3	Coconut shell crystal + cashew shell powder	2 + 1	4.8	10	7

When coconut shell powder alone is used as the adsorption agent only 1 ppm of fluoride is adsorbed from the sample, even cashew shell powder is mixed with the same adsorption agent the result was same. So we conclude that cashew shell is a poor adsorption agent for fluoride. At the same time the coconut shell crystal along with cashew shell powder proved itself to be the more effective adsorbent for fluoride as comparatively it adsorbed 3 ppm of fluoride. Hence, we finalize that the crystal form of coconut shell is the most effective adsorption agent among the adsorbents used.

E. Bamboo shoot powder

A water sample with an initial fluoride concentration of 10 ppm was treated using bamboo shoot powder powder as the adsorbent. The test was conducted at pH values between 2 and 6, and amount of adsorbent bed of 3g for a contact time of 1.5 to 3 hours. Table 3 shows Fluoride adsorption by bamboo shoot and Fig. 14 shows graphically the fluoride adsorption by activated bamboo shoot. After the treatment of the samples, it was observed that fluoride concentration of samples treated with 2g, 4g, and 6g of the bamboo shoot powder all showed a final concentration of 7 ppm. But when the amount of bamboo shoot powder was increased to 8g and 10 g the fluoride concentration was 9 ppm.

Table 3. Fluoride adsorption by bamboo shoot

Sl. No.	Weight of bamboo shoot (g)	pH	Initial fluoride content (ppm)	Final fluoride content (ppm)	Adsorption (ppm)
1	2	4.9	10	7	3
2	4	4.9	10	7	3
3	6	4.9	10	7	3
4	8	4.9	10	9	1
5	10	4.9	10	9	1

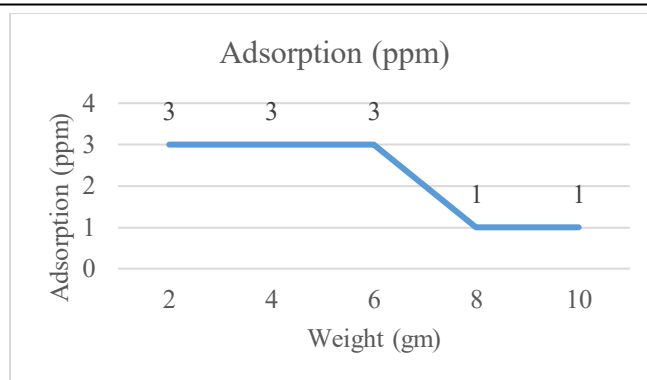


Fig. 14. Fluoride Adsorption by bamboo shoot.

From the above set of experiments, we observed that, from two to six grams of bamboo shoot powder, about 3 grams of fluoride is adsorbed within a pH range of 2-6. When the weightage of adsorbents is increased the adsorption rate fallen gradually, as eight gram and ten grams of adsorbent is used; only 1 ppm of fluoride is adsorbed.

F. Comparison with Previous Studies

Table 4 shows how different low-cost bioadsorbents remove fluoride from water sample with key parameters like pH, Contact Time, Ph, Dose and Initial F⁻ level.

Table 4. Comparison of Fluoride Removal Efficiencies for Low-Cost Bioadsorbents under Batch Conditions

Adsorbent & Study	Conditions (pH, Contact Time, Dose, Initial F ⁻)	Removal Efficiency	Source
crystals of coconut shell, cashew shell, bamboo shoot	pH 4.4–4.9, 1.5–3 h, 3.5 g C.S. powder; 2 g C.S. crystal+1 g C.Shell; 2–6 g bamboo shoot; initial 10 ppm	10 % (→9 ppm) with C.S. powder; 30 % (→7 ppm) with C.S. crystal + cashew; 30 % (→7 ppm) with 2–6 g bamboo shoot	This study
Coconut shell coal	pH not stated; contact time not stated; 1 g/L dose; initial 10 ppm	~13.7 % (1.37 mg F ⁻ /g capacity)	[2]
Mixed low-cost adsorbents	pH ~5; 2 h contact; doses not specified; initial 10 ppm	50–96 % (range across adsorbents)	[4]
Coconut shell + other bioadsorbents	pH 6; 2 h contact; 1 g adsorbent/100 mL; initial 5 ppm	83.5 % (→0.825 mg/L)	[1]
Review of various plant-based adsorbents	pH 5–6; 2 h contact; 1–4 g/L dose; initial 10 ppm	30–40 % (rice husk, sawdust)	[3]
Rice-husk biochar	pH 4–5; 2 h contact; 5 g/L dose; initial 10 ppm	35 % (→6.5 ppm)	[6]

VI. CONCLUSIONS

This study demonstrated that low-cost bioadsorbents; coconut shell (powder and crystal), cashew shell powder, and hydrothermally treated bamboo shoot powder; can remove up to 30 % of fluoride under batch conditions (pH 4.4–4.9, 1.5–3 hours contact), reducing fluoride from 10 ppm to 7 ppm. However, this level of fluoride removal is insufficient to meet the Bureau of Indian Standards permissible limit of 1 mg/L, indicating that these materials cannot serve as a standalone solution for safe drinking water defluoridation. To enhance practicality at the rural level, future work should include chemical activation of the carbons (acid/base or steam treatments) to increase surface area and functional binding sites; expanded optimization of pH and contact times including continuous flow column studies to maximize capacity; and development of hybrid treatment trains that combine bioadsorptive bulk removal with established polishing methods (e.g., activated alumina, ion exchange, or membrane filtration) to achieve the $\geq 75\%$ removal required. Pilot - scale trials with actual groundwater and evaluation of adsorbent regeneration and reuse will be critical to validating a cost-effective, community - deployable defluoridation approach.

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