

Defluoridation of Water by a Biomass: *Tinospora cordifolia*

Piyush Kant Pandey¹, Madhurima Pandey², Rekha Sharma²

¹Bhilai Institute of Technology, New Raipur, India; ²Centre for Environmental Science & Engineering, Department of Engineering Chemistry, Bhilai Institute of Technology, Durg, India.

Email: drpiyush_pandey@yahoo.com, drmadhurima_pandey@yahoo.com, rekha.sharma.ssitm@gmail.com

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ABSTRACT

This research is focused on the search of a biomass for the sequestration of fluoride from drinking water. Defluoridation of water was studied by batch experiments in biosorption process. The biomass was found to reduce fluoride to permissible limit 1.5 mg/L as prescribed by WHO. The efficiency of the sorption process was investigated under different experimental parameters such as pH 7, standing time 120 min and biomass doses 7.0 g with 5 mg/L concentration of fluoride. Neutral pH was identified as the optimum condition of the medium and 120 minutes was the best contact time for maximum fluoride adsorption. The experimental data was found good fitting to Langmuir and Freundlich isotherm models. In interference study tolerable effect was found with 50 mg/L concentration of co-ions, whereas increasing the concentration of co-ions retarded the fluoride removal capacity in some extent. FT-IR spectrum analysis showed fluoride binding in the different frequency ranges of the biomass. Eventually, this plant biomass is recommended as a suitable and low cost adsorbent to reduce fluoride into standard permissible limit.

Keywords: Sequestration; Defluoridation; Biosorption; FT-IR Spectrum; Permissible

1. Introduction

The occurrence of high fluoride in groundwater has drawn considerable attention the world over, since groundwater is the main source of fluoride intake [1]. Mineral sediments as well as discharge of industrial wastewater containing fluoride is a key contributor to groundwater and surface water pollution [2]. According to World Health Organization (WHO) guidelines, the optimum concentration of fluoride in drinking water is 1.5 mg/L [3]. According to the Indian standards for drinking water [4] the desirable limit of fluoride in drinking water is 1.0 mg/L; however, in the absence of any alternate source, maximum permissible limit is 1.5 mg/L. Fluoride ion in water exhibits unique properties, as its concentration in optimum dose in drinking water is advantageous to health and excess concentration beyond the prescribed limits affects the health [5]. Fluoride in drinking water has both beneficial and harmful effects on human health [6]. Fluoride in minute quantity is an essential component for normal mineralization of bones and formation of dental enamel [7]. When the concentration of fluoride is more than 4 mg/L, it causes softening of bones, ossification of tendons and neurological damage in several cases [8]. Whereas low fluoride content (<0.60 mg/l) in drinking water can cause dental caries and poor development of bones [9], high fluoride content (>1.0 mg/L) can lead to dental and skeletal fluorosis [10].

Excessive consumption of fluoride for a long term can lead to skeletal damage, dental fluorosis, mental disorders in children, osteosclerosis and structural changes in DNA [11]. The most common health problems due to intake of fluoride are dental and skeletal fluorosis [12]. Though fluoride enters the body mainly through water, food, industrial exposure, drugs, cosmetics etc. drinking water is the major source (75%) of daily intake [13].

High fluoride concentration in the ground water and surface water in many parts of the world is a cause of great concern. The occurrence of high fluoride concentration beyond permissible limit in groundwater is a problem faced by many countries notably India, Sri Lanka, Pakistan, China and parts of East Africa. The problem of excessive fluoride in ground water in India was first detected in Nellore of Andhra Pradesh in 1937 [14]. More than 60 million people including children in India are suffering with dental, skeletal and non skeletal forms of fluorosis [15].

The high fluoride levels in drinking water and its impacts on human health have increased the importance of defluoridation studies [16]. Defluoridation was reported by adsorption [17], chemical treatment [18], ion exchange [19], membrane separation [20], electrolytic de-fluoridation [21] and electro dialysis [22] etc. Among various processes, adsorption was reported to be an effective, environmentally friendly and economical one

[23]. The advantages of biosorption are very well known, the contaminants in water are removed by getting concentrated onto a disposed of [24]. Biosorption offers advantages of high efficiency in dilute effluents and no nutrient requirements. Recently considerable interest was observed on the application of biosorbent materials for removal of various pollutants. It provides a cost-effective solution for water management [25]. Adsorption is a mass transfer process in which a constituent in the liquid or gas phase is accumulated on solid or liquid phase and separated from its original environment [26]. Due to more simple design of adsorption unit, lack of sludge production and low investment costs, the adsorption process has more advantages than other methods in removal of pollutants from water and wastewater [27]. The uptake of anions has become a growing concern in the field of biosorption [28]. Investigators reported various types of adsorbents namely activated alumina [29], titanium-rich bauxite [30], synthetic resins [31], manganese oxide-coated alumina [32], carbon nanotubes [33], fish bone charcoal [34], rice husk carbon [35], with different degrees of success.

2. Materials and Methods

2.1. Plant Collection and Preparation of the Biomass

Plant was collected from different localities of Bhilai town ship C. G. in district Durg. The plant biomass used for the removal of fluoride ions from aqueous solutions. The plant was washed with deionised water to remove dirt and other particulate matter then cut into small pieces, sundried for 7 days then kept in a hot airoven at 60°C till dryness. It was then powdered and sieved through 1.18 μM sieve for getting uniform size biosorbent.

2.2. Biosorption Experiment

The biosorption capacity of biomass was determined by contacting various concentrations (1 - 7 mg/L) of 50-ml fluoride solution in 250-ml plastic beakers with 1 - 9 g of biomass. The filtrate containing the residual concentration of fluoride was determined by Orion ion meter (290A). For the determination of rate of fluoride biosorption, the supernatant was analysed for residual fluoride after the contact time of 10 - 1440 minutes. The effect of pH on fluoride sorption by biomass was determined at pH values of 3 - 8. Values of experimental operating parameters investigated are given in **Table 1**. Adsorption isotherm studies were carried out with different initial concentrations of fluoride while maintaining the adsorbent dosage. Pseudo first order model were fitted better when compared with the second order kinetic model. Langmuir and Freundlich models were applied to the

Table 1. Variation of experimental operating parameters of fluoride.

Parameter (permissible limit in mg/L)	Values investigated (1.5)
Initial fluoride concentration in mg/L	1, 2, 3, 4, 5, 6, 7
Amount of adsorbent dosages in g	1, 2, 3, 4, 5, 6, 7, 8, 9
Agitation time in minutes	10, 30, 60, 120, 180, 1440
pH	3, 4, 5, 6, 7, 8
Interference study of co-ions in mg/L	50, 100, 150, 200
Uptake capacity in mg/g	25

adsorption isotherm and different constants were generated. The Langmuir and Freundlich adsorption parameters and correlation coefficient were also calculated from the adsorption isotherm data.

3. Result and Discussion

3.1. Effect of Adsorbent Dose on Fluoride Adsorption

Biomass dose is an important parameter owing to its effect on efficiency and on the amount of fluoride removed per unit weight of biomass. The effect of biosorbent dosage (1 - 9 g/50ml) on the percentage removal of fluoride was studied. The fluoride adsorption increased with the increasing amount of the adsorbent as higher doses of the adsorbent provided more active sites. The dose of adsorbent having the optimum fluoride removal efficiency was found to be 7 g/50ml. Further increase in doses did not resulted in the considerable increase in defluoridation. This is due to the overlapping of the active sites at higher concentrations of the adsorbents, thus reducing the net surface area. Hence 7 g of the biomass was taken as the dose possessing the optimum fluoride removal efficiency and this was fixed as the dose of the material for further experiments.

3.2. Effect of pH on Biosorption

The pH of the aqueous solution is an important controlling parameter in the adsorption process. Thus, the role of hydrogen ion concentration was examined at pH values of 3 to 8. This was adjusted by adding 0.5 N HCl or 0.1 M NaOH. The effect of pH (3 - 8) on the removal of fluoride for a constant biosorbant dosage of 7 g/50 ml, and fluoride ion concentration of 5 mg/L was studied. It was found that pH had a marked effect on the fluoride uptake in this experiment. The percentage adsorption of fluoride ion was found to maximum at pH 7 (**Figure 1**) and then it decreased with a further increase of pH. The optimum pH for the removal of fluoride was found to be 7.

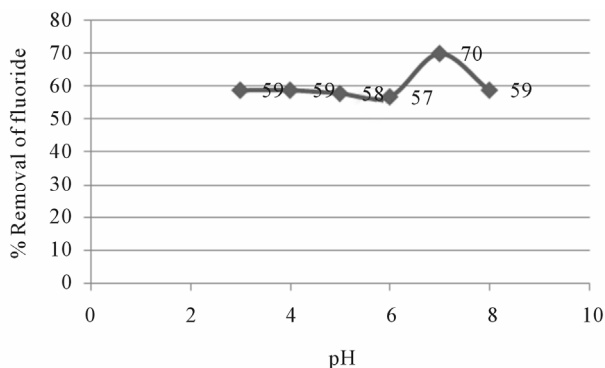


Figure 1. Showing effect of pH on % removal of fluoride.

3.3. Time Dependence Studies for Fluoride Binding

The variation in percentage removal of fluoride with time was studied using the solution of fluoride with initial concentration of 5 mg/L, adsorbent dosage 7 g at pH 7. The time was varied from 10 to 1440 min. On increasing the contact time, the percentage removal was found to gradually increase till 120 min. Further increase in time decreased the removal of fluoride. Hence, the optimum contact time for fluoride removal was 120 min (Figure 2) at pH 7. Further increase in contact time does not increase the uptake due to saturation of fluoride ions on the available sites present on adsorbent material. This means that the biomass requires contact time of 120 minutes for the removal of the maximum amount of fluoride. For the optimization of other parameters, the contact time of 120 minutes was considered as the equilibrium time.

3.4. Effect of Initial Fluoride Concentration

To achieve the maximum adsorption capacity of fluoride, the experiment was conducted under optimum conditions (adsorbent doses fixed at 7 g/50 ml, fixed contact time of 120 min at pH 7). The effect of initial fluoride concentration was studied by varying fluoride concentration from 1 to 7 mg/L. The results indicate that the percentage removal were remaining constant (70%) after the 5 mg/L concentration of fluoride solution. This is due to lack of available active sites on the adsorbent surface.

3.5. Study on the Adsorption Kinetics

In this kinetic study the experiment was conducted in which the concentration of fluoride was 5 mg/L (where time varied from 10 min to 1440 min at 25°C). The amount of fluoride adsorbed q (mg/g) increased and $\log(q_e - q)$ decreased from 10 min to 120 min, whereas from 120 min to 1440 min q (mg/g) decreased and $\log(q_e - q)$ increased. The value of co-relation coefficient R^2 is greater (0.99) than (0.82) showing the effect of time on adsorption process before and after equilibrium time.

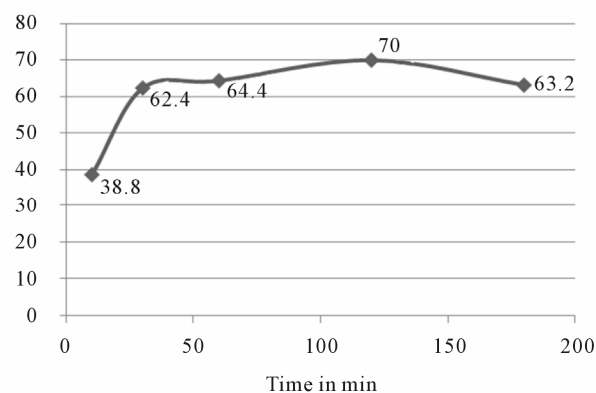


Figure 2. Showing effect of contact time on % removal of fluoride.

3.6. Sorption Equilibria Studies

The linear plots of C_{eq}/q vs. C_{eq} show that adsorption follows the Langmuir adsorption model. The essential characteristics of the Langmuir isotherms can be expressed in terms of a dimensionless constant separation factor or equilibrium parameter, R_L , which is defined as

$$R_L = 1/(1 + bC_0)$$

where b is the Langmuir constant and C_0 is the initial concentration of fluoride. The R_L value indicates the shape of isotherm. According to McKay *et al.* (1982), R_L values between 0 and 1 indicate favourable adsorption. $K_f(1.047)$ and $n(1.325)$ were calculated from the slopes of the Freundlich plots. According to Kadirvelu and Namasivayam (2000), n values between 1 and 10 represent beneficial adsorption. The magnitude of K_f and n shows easy separation of fluoride. The value of n , which is related to the distribution of bonded ions on the sorbent surface, is indicating that adsorption of fluoride is favorable.

3.7. Co-Ions Competition and Biosorption Interference

Co-ions competition and interference experiments are performed to find out the interference of other species with the binding of the target metal ion to the biomass. Competition between chemical species for the binding sites depends on factors such as the chemistry of the metal, pH of the solution, the nature of the binding sites, the amount of binding sites, the diversity of chemical species, metal ion concentration, and the selectivity of the biomass to bind certain species. The interference studies of co-ions (50 - 200 mg/L) with 5 mg/L solution of fluoride were investigated. In this study tolerable effect was found with 50 mg/L concentration of co-ions (Cl, NO₃, SO₄, PO₄, Cr, Cd, Ca, Mg, Mo, Ni, Cu, Fe, Mn), whereas increasing the concentration, retarded the fluoride removal capacity in some extent.

3.8. Interpretation of FT-IR Spectra

On comparing the IR spectra it was found that there are significant changes in the fresh and exhausted biomass spectra. The identification of absorption spectrum shows the range, type, vibration and functional groups of the biomass. FT-IR study of fresh biomass (**Figure 3**) and exhausted biomass (**Figure 4**) shows major differences in the region 500 - 1600 cm^{-1} range is assigned to $-\text{NH}_2$ groups in different parts of the biomass. Frequency

ranges of biomass indicating bonding of fluoride with the amines groups of biomass. Hence, based on FT-IR spectrum analysis it can be conclude that the fluoride binding in the biomass takes place by the substitution of amine groups.

3.9. Applicable to Treatment of Fluoride at Neutral pH

Permissible limit of fluoride contaminated water for use

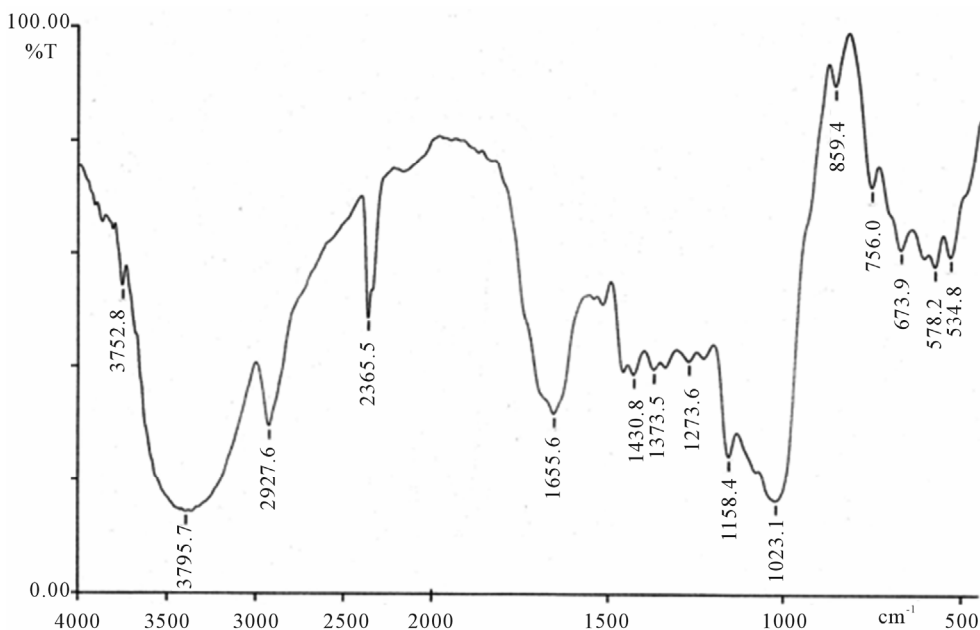


Figure 3. FT-IR spectra of fresh biomass.

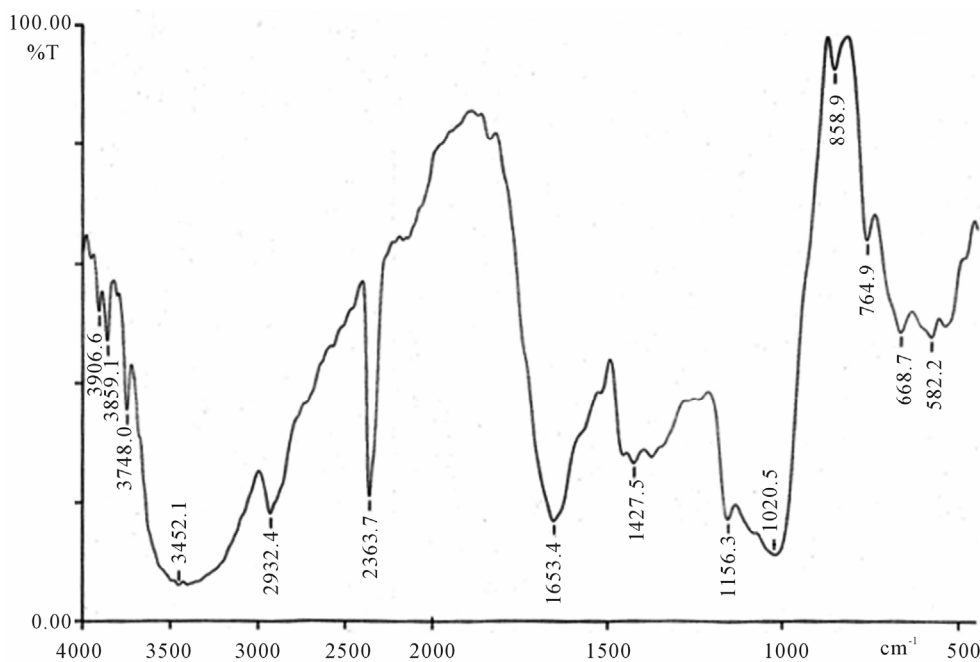


Figure 4. FT-IR spectra of exhausted biomass.

in irrigation and other purposes are higher than the limit for drinking water. This biomass was applicable to defluoridation treatment of drinking water within standard WHO permissible limit (1.5 mg/L) at neutral pH without changing the chemical nature of water. The appreciable results were being obtained without disturbing the pH of the water.

3.10. Significance of the Plant Biomass

Plant material was readily, locally available, rich in proteins and calcium, caused no change in the pH of the treated water, and gave good result at neutral pH, their residue after use was easy to dispose off without changing the pH of the disposed field. This plant material was used as a cost effective, potential natural biosorbent for defluoridation of water. Comparison of the adsorption capacities of different sorbents are shown in **Table 2**.

3.11. Mass Balance of Fluoride in the System

Desorption study was carried out to account for the mass balance of fluoride in the system as its concentration in optimum dose in drinking water is advantageous to health. For this the loaded biomass after a single exposure to the known amount of fluoride was carried out for

desorption. The result shows that about 55% of the adsorbed amount of fluoride could be leached easily after first attempt of desorption only. Remaining up to 99.9% of the fluoride was desorbed after sub sequential attempts of this study.

4. Conclusion

The biomass of the natural plant demonstrated a good capacity of fluoride biosorption, highlighting its potential for the drinking water treatment process. pH had a strong effect on biosorption capacity and the optimum pH found 7. The biosorption was rapid and equilibrium achieved within 120 min. The uptake capacity of fluoride was found to be 25 mg/g. The results indicate that Langmuir and Freundlich sorption models were good agreement with the experimental results. Further, the biosorbents are characterized by FT-IR spectral analyses. The biomass found to be very efficient, instantaneous and economical for removing fluoride from drinking water. This biosorptive material is very useful to reduce fluoride within standard WHO permissible limit (1.5 mg/L) at neutral pH. The significant results were being obtained without disturbing the pH of the water. The fluoride containing biomass can be dumped as a solid waste material in pits

Table 2. Comparison of the adsorption capacities of different sorbents for defluoridation.

Adsorbents	pH	Adsorption capacity (mg/g)	References
Alumina cement granules	-	34.36	[36]
Carbon nanotubes	7.0	33.7	[37]
Powdered biomass <i>Tinospora cordifolia</i>	7.0	25	[This plant]
Hydrous ferric oxide	4.0	16.5	[38]]
Ca-treated <i>Anabaena fertilissima</i>	-	7.0	[39]
Waste carbon slurry	7.58	4.86	[40]
Ca-treated <i>Chlorococcum humicola</i>	-	4.5	[39]
<i>Eichhornia crassipes</i>	-	4.4	[41]
Manganese oxide coated alumina	7.0	2.85	[42]
Activated alumina	7.0	1.78	[43]
<i>Spirodela polyrrhiza</i>	-	0.91	[44]
Used tea leaves	1.5	0.51	[45]
Moringa indica based activated carbon	2.0	0.23	[46]
Activated charcoal	6.8	0.15	[47]
Alum treated flyash	6.8	0.08	[47]
Powdered biomass (<i>Azadirachta indica</i> + <i>Ficus religiosa</i> + <i>Acacia catechu willd</i>)	2.0	0.04	[48]

without changing the chemical nature of soil because the whole experimental parameters were identified at the neutral pH 7.

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