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# Defluoridation of Water Using Low Cost Adsorbent

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### Abstract

Fluoride is often called a two- edge sword. In the human system this fluoride has a dual personality, a destructive effect (greater than 1.5 ppm – dental & skeletal fluorosis) and a beneficial effect (upto 1.0 ppm – caries prevention and health promotion). World Health Organization (WHO) and IS: 10500 recommend that the fluoride content in drinking water should be in the range of 1.0 to 1.5 ppm. Fluoride concentrations beyond the standards cause dental and skeletal fluorosis.

This paper presents the findings of an investigation on the use of leaf powder from various trees for the defluoridation of water. Here the study presents the suitability of inexpensive leaf adsorbents to effectively remediate fluoride-contaminated water. The efficiency of the sorption of fluoride ion is affected by contact time, pH and particle size of adsorbents. Treated leaf powder was studied at various pHs & contact time with aqueous solutions containing 10mg F<sup>-</sup> /l.

**Keywords:** Fluoride, Low cost adsorbent, Dental Fluorosis, Skeletal Fluorosis.

### 1. Introduction

Water is the major medium of fluoride intake by humans [1]. Fluoride in drinking water can be either beneficial or detrimental to health, depending on its concentration [2]. Ground water is the major source of freshwater on the earth. Groundwater containing dissolved ions beyond the permissible limit is harmful and not suitable for domestic use. Fluoride beyond desirable amounts (0.6 to 1.5mg/l) in groundwater is a major problem in many parts of the world [3]. The fluorides belong to the halogen group of minerals and are natural constituents of the environment. Fluorides are mainly found in ground water when derived by the solvent action of water on the rocks and the soil of the earth's crust. Fluoride is the most electronegative of all chemical elements and is never encountered in nature in the element form. It is seventeenth in the order of frequency of occurrence of the elements and represents about 0.06% to 0.09% of the earth's crust [4]. Many workers and reviewers used the word 'Fluoride' to denote the ionized, physiologically available form of the element. The chemical activity of the fluoride ion ( $E_0 = -2.8$  Volts) makes it physiologically more active than other elemental ion. Therefore, fluoride ions play an important role in human physiology. Its presence in low concentration may either inhibit or stimulates enzymatic process and its interaction with other organic and inorganic body components may cause disruption in normal physiological functions of human body.

The popular technologies for the removal of fluoride from water include: coagulation followed by precipitation, membrane processes, ion exchange and adsorption [5, 6, 7, 8, 9]. In coagulation, trace amounts of fluoride ions tend to remain in solution due to solubility restriction. Other shortcomings include the resulting high pH of the treated water and the generation of large amount of wet bulky sludge [10, 11].

The Nalgonda technique, based on precipitation processes, is also a common defluoridation technique. The limitations of the process are: daily addition of chemicals, large amount of sludge production, and low effectiveness for water having high total dissolved solids and hardness. Further, increase in residual aluminum in the treated water has been reported [12]. This may endanger human health as concentrations of aluminum, a neurotoxin, as low as  $8.0 \times 10^{-2}$  mg/l in drinking water have been associated with Alzheimer's disease [13, 14]. Membrane processes, though effective in fluoride removal, demineralise water completely, besides the high initial and maintenance costs.

Ion exchange methods are efficient for fluoride removal, but a tedious and difficult process of preparation of resins as well as the high cost necessitates a search for an alternative technique.

Adsorption techniques have been quite popular in recent years due to their simplicity, as well as the availability of wide range of adsorbents.

Research has focused on various types of inexpensive and effective adsorption media, such as different clays [15, 16], solid industrial wastes like red mud, spent bleaching earths, spent catalysts and fly ash [18, 19, 20, 21, 22], activated alumina, carbonaceous materials [23, 24, 25], bone charcoal [26], natural and synthetic zeolites and other low-cost adsorbents, with various degrees of success [27].

Here, this communication presents an investigation on the use of leaf powder from various trees for the defluoridation of water.

## 2. Materials and Methods

All the reagents used were of AR grade. Fluoride stock solution was prepared by dissolving 221 mg anhydrous sodium fluoride in 1000 ml distilled water in volumetric flask. Fluoride standard solution was prepared by diluting 100 ml stock solution to 1000 ml distilled water in volumetric flask. This 1 ml solution has 0.1 mg of fluoride.

### 2.1. Equipment

Fluoride ion was estimated by Orion ion meter as per standard methods. PH meter, and Remi shaking machine for agitating the samples for the required period at a speed of 200 strokes/minute were used. The surface area of the adsorbent particle, porosity and density were measured by using surface area analyzer, mercury porosimetry and specific gravity bottles, respectively.

### 2.2. Material development

Fresh leaves chosen based on their crude fiber content and tress were obtained from neem (*Azadirachta indica*) trees. The fresh leaves were sun-dried for 3–4 days, put in a cotton jute bag and crushed manually. (This process can save the energy expended in hot air oven drying and mechanical crushing.) The powder was sieved to get various particle sizes, viz. 600, 710, and 850 m, 1 mm, and 1.4 mm. Leaf powder biomass was further digested by chemical methods.

### 2.3. Alkali treatment

Leaf biomass powder sample (40 gm) and 400 ml 0.5 N NaOH was taken in 1000-ml conical flask. Then mixture was gently heated on burner for 20 min after boiling started. Using distilled water, the treated biomass was washed which continued until maximum colour was removed and clear water obtained.

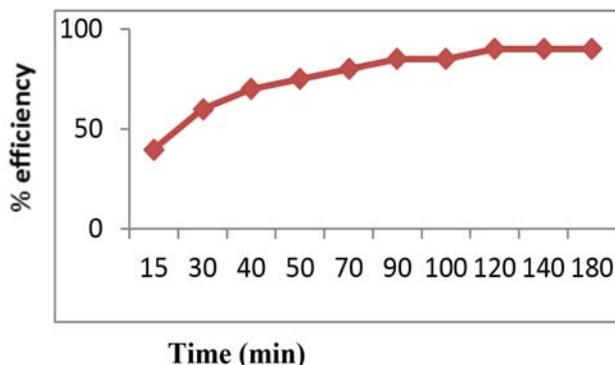
## 3. Results and Discussion

Successful application of the adsorption technique demands innovation of cheap, nontoxic, easily and locally available material. Bio adsorbents meet these requirements. Knowledge of the optimal conditions would herald a better design and modeling process. Thus, the effect of some major parameters like pH, contact time, and particle size of adsorbent and materials was investigated from kinetic viewpoint. Adsorption studies were performed by batch technique to obtain the rate and equilibrium data. Experiments were carried out by shaking 10 g/l of adsorbent dose with 100 ml of aqueous solution containing known concentration of fluoride ions and by agitating the samples on Remi shaking machine at a speed of 200 strokes/min. Samples containing fluoride ions were maintained at a desired pH by adding 0.5 N HNO<sub>3</sub>. All the

experiments were conducted at room temperature ( $27 \pm 0.5$  °C).

### 3.1. Effect of contact time

It is found that the removal of fluoride ions increases with increase in contact time to some extent. Further increase in contact time does not increase the uptake due to deposition of fluoride ions on the available adsorption sites on adsorbent material. Preliminary investigations on the uptake of fluoride ions on the adsorbent material at their optimum pH values indicate that the processes are quite rapid.

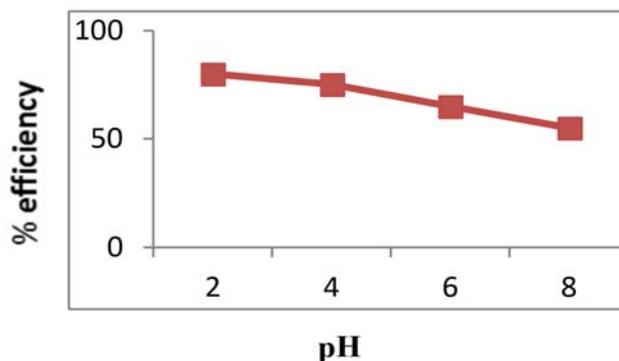


**FIG 1:** Effect of agitation time on the removal of fluoride ion. Adsorbent dose: 10 g/l, adsorbent size: mixed, volume of sample: 100 ml, temp.: 27°C, initial adsorbate conc.: 10 mg/l, pH: 2.

Typically, 80% of the adsorption occurs within the first hour of the contact for fluoride ions with an initial concentration and adsorbent dose of 10 mg/l for treated biosorbents (Fig.1). This initial rapid adsorption subsequently gives way to a very slow approach to equilibrium and saturation is reached in 1.5 to 3 h. For further optimization of other parameters, this contact time was considered as the equilibrium time.

### 3.2. Effect of pH

The pH of the aqueous solution is a controlling factor in the adsorption process. Thus, the role of hydrogen ion concentration was examined at pH values of 2, 4, 6, 8. The pH of the aqueous solution is a controlling factor in the adsorption process. Thus, the role of hydrogen ion concentration was examined at pH values of 2, 4, 6, 8, and 10. This was adjusted by adding 0.5N HNO<sub>3</sub> with 100 ml of standard solution of 10 mg/l of fluoride for a contact time of 60 min with a dose of 10 g/l of treated bioadsorbent. The influence of pH on the sorption rate is shown in Fig2.



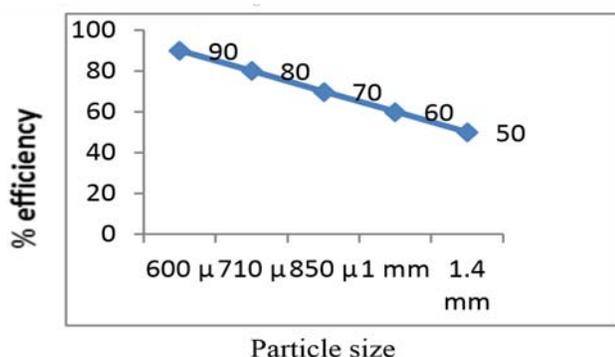
**FIG 2:** Effect of pH on the percentage removal of fluoride ion. Adsorbent dose: 10 g/l, adsorbent size: mixed, volume of sample: 100 ml, temp.: 27 °C, initial adsorbate conc.: 10 mg/l, time of contact: 60 min.

We have observed decrease in the extent of removal of fluoride ions with increase in the pH of the solution. This was investigated as 80% at pH 2 and 75% in the case of pH 4. Hence further studies were conducted within these pH values. In the case of treated biosorbents, the percentage of adsorption increased almost linearly between 2.0 and 8.0, attaining a maximum removal at pH 2.0 in 60 min of contact time.

In this case, the result may be due to neutralization of the negative charges at the surface of the treated biosorbents by greater hydrogen ion concentration at lower pH values. This reduces hindrance to diffusion of the negatively charged fluoride ions on to the increased active surface of treated biosorbents.

### 3.3. Effect of adsorbent particle size

Experiments were conducted to evaluate the influence of adsorbent particle size for a constant weight on the removal of fluoride ions. Particle size analysis was conducted on treated bioadsorbents and the percentage composition of particle size was investigated. The results obtained with the variation of adsorbent particle size and the percent of the fluoride ions are graphically represented in Fig. 3.



**FIG 3:** Effect of particle size on removal of fluoride ion. Temp.:  $27 \pm 5$  °C, initial adsorbate conc.: 10 mg/l, volume of sample: 50 ml, adsorbent dose: 10 g/l, time of contact: 120 min, pH: 2.

The uptake of fluoride ions at different adsorbent particle sizes increased with decrease in sorbent particle diameter. The presence of large number of smaller particles provides the sorption system with a larger surface area available for fluoride ion removal and it also reduces the external mass transfer resistance. Also, the time required for 50% of the total adsorption is less with the particles of smaller size. This also gives some idea of rate-limiting step of the adsorption process. The removal of fluoride ions has been studied at a room temperature of  $27 \pm 0.5$  °C. With the largest particle size of 1.4 mm the amount of fluoride ions adsorbed was found to be 50-60% treated biosorbents respectively, and 90-95% with smallest particle size of 600μ for an initial fluoride ion concentration of 10 mg/l, respectively. Small particle size provides more active surface area and hence such results were observed.

### 3.4. Disposal of exhausted adsorbent materials

It is necessary to separate fluoride ion before its disposal. The exhausted adsorbent materials need to be dried and burned. The resultant product can be used in the manufacturing of bricks. Using various chemicals such as  $H_2SO_4$ , HCL,  $HNO_3$ , NaOH, EDTA, etc. desorption is possible.

### 3.5. Advantage of low-cost adsorbents over conventional adsorbents

The efficiencies of removal of fluoride ions of various nonconventional adsorbents vary between 50 and 90% depending upon the characteristics and particle size of adsorbent(s). A combination of adsorbents can also be used effectively in defluoridation treatment.

- Nonconventional adsorbents are relatively cheaper compared to conventional ones and are easily available resulting in savings in cost.
- Nonconventional adsorbents require simple alkali or acid treatment for the removal of lignin before their application and to increase efficiency.
- Since the cost of these adsorbents is relatively low they can be used once and discarded.
- Nonconventional adsorbents require less maintenance and supervision. Separation is possible to segregate the nonconventional adsorbents from the effluents before their disposal.
- These nonconventional adsorbents can be disposed off easily and safely. Used adsorbents can be reused as a filler material in low-lying areas and hence their disposal does not pose any serious problem.

### 4. Conclusions

Results show that these low-cost bio adsorbent could be fruitfully used for the removal of fluoride over a wide range of concentrations. Treated bio adsorbents were observed to be efficient for the uptake of fluoride ions between 2.0 and 8.0 pH. Fluoride removal for a given bio adsorbent size increased with time attaining equilibrium within 1.5 h.

The percentage of fluoride removal was found to be a function of adsorbent particle size and time at a given initial solute concentration. It increased with time, and higher initial solute concentration decreased with time.

The adsorption capacity of treated biosorbents was studied by varying the particle size. With the largest particle size of 1.4 mm, the amount of fluoride ions adsorbed was found to be 50%. With smallest particle size of 600μ for an initial fluoride ion concentration of 10 mg/l, 90% adsorption was observed. Small particle size provides more active surface area and hence such results. Treated biosorbents can be disposed off safely by burning after use.

Treated biosorbents are locally available and hence involve no expenditure on transportation and have a very low cost for pretreatment. There is no need to regenerate the exhausted treated biosorbents as they are available abundantly, easily, cheaply and locally. Our future work deals with the evaluation and performance of various biomasses for the removal of fluoride.

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