

Defluoridation of Water Using Low Cost Adsorbents

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Abstract

The aim of this study was to develop a low-cost and safe method of fluoride removal. The adsorbents used in this investigation were medicinal plants like Ramacham (*Vetiveria zizaniode*), Chukku (*Zingiber officinale*) Pathimugam (*Caesalpinia sappan*) and Clove (*Syzygium aromaticum*). All adsorbents used were cheap and easily available. All adsorbents are medicinal herbs. The adsorbents were effective in removal of fluoride. Batch experiments were conducted by varying the pH, contact time, adsorbent dose and heating time. Finally, it has been observed that adsorbents were definitely having satisfactory adsorbent capacity.

KEYWORDS: Fluoride Removal, Ramacham (*Vetiveria zizaniode*) chukku (*Zingiber officinale*) Pathimugam (*caesalpinia sappan*) and Clove (*syzygium aromaticum*).

1.INTRODUCTION

Water is the most abundant and is essential component of our life supporting system. But today most of the countries are facing drinking water problems. In India, drinking water is contaminated at many places by various pollutants such as fluorides, nitrates, iron and so on.

Fluoride is considered as a “two edged sword” because deficiency of fluoride intake leads to dental caries while excess consumption leads to dental and skeletal fluorosis. Fluorine, the most electro negative of all compounds has not only notable chemical properties but also physiological properties of great importance to human health and well being. Fluoride ion is attracted by positively charged calcium ion in teeth and bones due to strong electro negativity which results in dental and skeletal and non skeletal fluorosis. Fluoride, the ionic form of fluorine is widely distributed in nature. It is a common constituent of most soil and rocks. The industrial effluent and sewage discharged from domestic water supplies supplemented with fluoride contribute to the fluoride level in aquatic systems. Combustion of coal and volcanic activity also contribute fluoride containing dusts and gases to the atmosphere.

During rainfall, these are being dissolved in water and contaminate water bodies. In India, most of the water bodies are highly contaminated by fluoride with varying concentration in the range of 0.5-20 mg/litre. The presence of fluoride in drinking water, with in the permissible limits of 0.5-1.0 mg /litre, is beneficial for the production and maintenance of healthy bones and teeth. However when consumed in higher doses (>1.5 mg / litre), it leads to dental fluorosis or mottled enamel and excessively higher concentration (>3.0 mg/litre) of fluoride may lead to skeletal fluorosis. In India, endemic fluorosis affects more than one million populations and is a major problem in 17 of 29 states.

Defluoridation of drinking water is the best practicable option to overcome the problem of excessive fluoride in drinking water. Among all available methods adsorption is considered to be simple, economical and globally pursued technique.

Adsorption is a surface phenomenon. Greater the surface area per unit mass of the adsorbent, greater will be the capacity for adsorption under the given conditions of temperature and pressure. Various adsorbents can be used for defluoridation. This study emphasizes on biosorbents. Sorption by using biosorbents is called biosorption. Sorption is a process in which adsorption and absorption takes place simultaneously. Therefore, sorption refers to surface and bulk phenomena of solids.

II. REVIEW OF RELATED LITERATURE

Studies on removal of fluoride by rice husk by **Waheed et al., (2009)** revealed that for de fluoridation optimum dose was found to be 10 mg /lit and optimum pH was found to be 2.

Sutapa chakrapathy (2012) had studied fluoride removal in aqueous solution by Neem charcoal as adsorbent and showed that de fluoridation were highly influenced by temperature, pH of solution and initial fluoride concentration. **Veeraputhiran et al (2011)** showed that activated carbon prepared from *Phyllanthus emblica* as adsorbent shows enhanced removal of fluoride by 82% at equilibrium contact time of 75 minutes. **Ramanjanejulu et al., (2013)** showed that maximum adsorption of fluoride for tamarind shell powder 85% and papal leaf powder 79.5% was observed at pH 2. **Hanumantaraoe et al., (2013)** studied that fluoride removal using activated carbon prepared from biomaterials *Typha Angustata*, *Lagenariasiceraria*, *Acacia farrusiana* as adsorbent and revealed that percentage of fluoride removal has increased with increase of contact time and dose of adsorbent. **Keerthi.B.Gurani (2015)** explained the removal of fluoride using batch techniques by activated carbon prepared from *Phoenix dactylifera* seeds. It revealed that maximum removal of fluoride is obtained at pH 7+ or - 0.20. **Suman Mann et al., (2014)** studied the removal of fluoride using saw dust. The experimental data were fitted to the langmuirequation. **Jamode (2004)** removed fluoride using Neem, Peepal and Khair. Fluoride removal for the adsorbents increased with time attaining equilibrium with in 1.5 hrs. **Nabakumar Mandal, et al., (2012)** revealed that residual part of Tea dust waste was effectively used for removal of fluoride from aqueous medium. **Dinesh Mohan et al., (2012)** removed fluoride using Bio-char, a green waste and revealed that fluoride adsorption decreased with increase of temperature. **Getachew.A.Huseen et al., (2014)** studied defluoridation using carbon prepared from banana peel and coffee husk. Coffee husk was observed to be much better than banana peel. **Bhagwanyada.V (2014)** studied defluoridation of drinking water using stem charcoal of aralu (*ailanthus exelsa*) bisorption equilibrium was achieved within 180 minutes and maximum fluoride removal is 94%. **Harikumar et al., (2011)** showed that *Vetiveria zizanoide* a herbalplant of kerala is an effective adsorbent for the removal of fluoride from aqueous solution.

Sinha et al., (2000) studied defluoridation using *Hydrilla verticillata* and observed that the plant is effective for removing fluoride. **Shanti et al., (2005)** studied defluoridation using low cost adsorbents and found that *Eletaria cardamomum* is most effective.

III. MATERIALS AND METHODS

Adsorption is the change in concentration at the interfacial layer between the two phases of a system due to surface forces. Temperature, surface area, nature of adsorbent, pH, contact time, size of adsorbent particles, etc are the factors affecting the adsorption. In the present study synthetic water sample is prepared and used for experimental purpose of defluoridation of water.

IV. MATERIALS USED

For preparing synthetic fluoride water sample anhydrous sodium fluoride (NaF) and distilled water were used. Biosorbents in the present study were medicinal plants like Ramacham (*Vetiveria Zizaniode*), Chukku (*Zingiber Officinale*), Pathimugam (*Caesalpinia Sappan*) and Clove (*Syzygium aromaticum*).

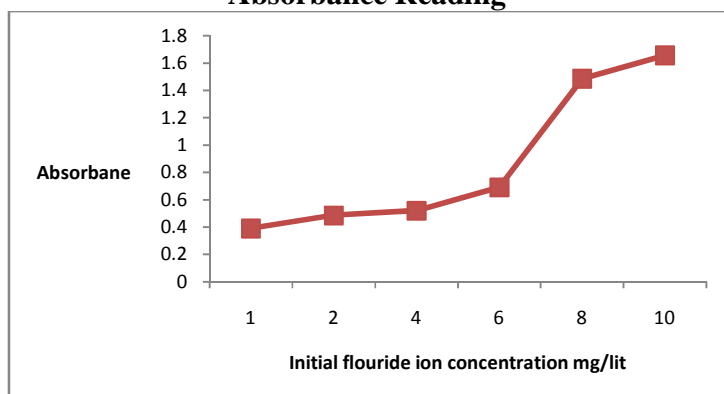
Adsorbents *Vetiveria Zizaniode* is a perennial grass. Its roots are used for making medicated drinking water. *Caesalpinia Sappan* is a flowering tree. Its heart wood is used for making herbal drinking water. Clove is the unopened flower bud obtained from the tree. The carminative and stimulant properties give clove high medicinal value. *Zingiber officinale* is a flowering plant. Dried ginger root is used as spice and as a folk medicine.

All the samples were brought from the local market, washed in tap water and double distilled water and dried and then grinded in motor and sieved to get fine powder particle of 75 micron. Adsorption of fluoride on biosorbents was done by batch sorption experiment. Synthetic fluoride solution having initial fluoride ion concentration of 10mg/lit is used. To this solution SPADNS and zirconyl acid solution of 5ml each was used. The sample was checked for fluoride detection in spectrophotometer at wavelength 570nm. Removal efficiency was found out by comparing with absorbance readings.

V. DEVELOPMENT OF STANDARD CURVE

The standard fluoride sample in the range of 1 mg/lit to 10 mg/lit was prepared by taking appropriate quantities of standard fluoride solution with distilled water. Then pipette 5 ml each SPADNS solution and zirconyl acid solution to each standard samples and mixed well. The Spectrophotometer was set to zero absorbance with reference solution and absorbance readings of standard were obtained.

Absorbance Reading



VI. EXPERIMENTAL SETUP

The study was conducted in a conical flask using fluoride water sample containing different pH and initial fluoride ion concentration. In these conical flasks adsorbent dosages

were added and particular contact time was given. After the contact time the content of the flask was filtered using Whatmans' filterpaper41. The filtrate was used for fluoride ion estimation using SPADNS method. The same procedure is used for different pH, contact time, adsorbent doses and heating minutes. The pH was varied from 2 to 10. The contact time was varied from 30 to 120 minutes. The adsorbent dose varied from 1.5gm to 3gm. The heating time was varied from 5 to 20 minutes. The change in parameters was done to find the maximum fluoride efficiency.

VII. RESULTS AND DISCUSSIONS

A.OPTIMUM pH

The experiments were carried out using adsorbents Ramacham, Chukku, Pathimugam and Clove by varying pH. The pH was varied from 2 to 10. For these experiments initial fluoride concentration was (10 mg/lit) with adsorbent dose of 1.5 gm and a contact time of 1 hour. When the Ramacham powder is used it was found that maximum removal (47%) was observed at pH 7. When the Chukku powder is used it was found that maximum removal (49.2%) was observed at pH 6. When the Pathimugam powder is used it was found that maximum removal (25.5%) was observed at pH 8. When the Grambu powder is used it was found that maximum removal (36.91%) was observed at pH 2. The pH controls the water adsorbent interfaces. The lower de fluoridation efficiency at pH below 6 was possibly due to protonation of fluoride. This result could be attributed to the formation of weakly ionized HF. Above the pH of 7, there may be competition between the OH⁻ and F⁻ ions. Chukku shows maximum fluoride removal at pH 6.

By using the bio adsorbents, fluoride ions could be removed effectively from water. Biosorption increased with increase in pH and then decreased. In the present study it was observed that de fluoridation was high in the pH range from 5 to 8. This finding agrees with the findings of earlier works.

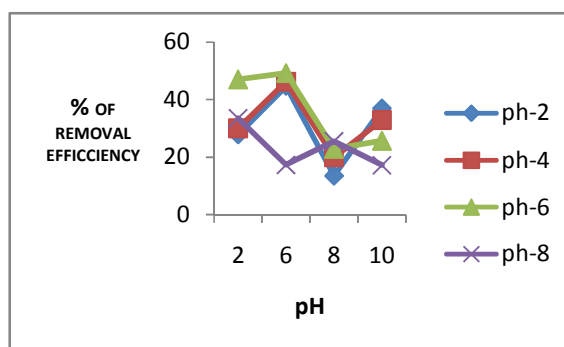


Figure:1

B.OPTIMAL CONTACT TIME

The adsorbent dose of 1.5gm was taken and kept constant throughout the experimental work. The contact time was varied from 30 minutes to 150 minutes for adsorbents. The maximum removal efficiency was 33.5% at a contact time of 60 minutes. The percentage of fluoride adsorbed in different intervals of time was studied. It was observed that on increasing the time by 30 minutes, the fluoride ion removal initially increased. After 60 minutes there is decrease in adsorption in all four biosorbents. When the Ramacham powder is used it was found that maximum removal (33.5%) was

observed at 60 minutes contact time. When the Chukku powder is used it was found that maximum removal (9.36%) was observed at 60 minutes contact time. When the Pathimugam powder is used it was found that maximum removal (24.95%) was observed at 60 minutes contact time. When the Grambu powder is used it was found that maximum removal (33.53%) was observed at 60 minutes contact time. The perusal of results show higher removal rate initially and slower rate after lapse of time Vacant adsorption sites, high solute concentration gradient, electrostatic affinity

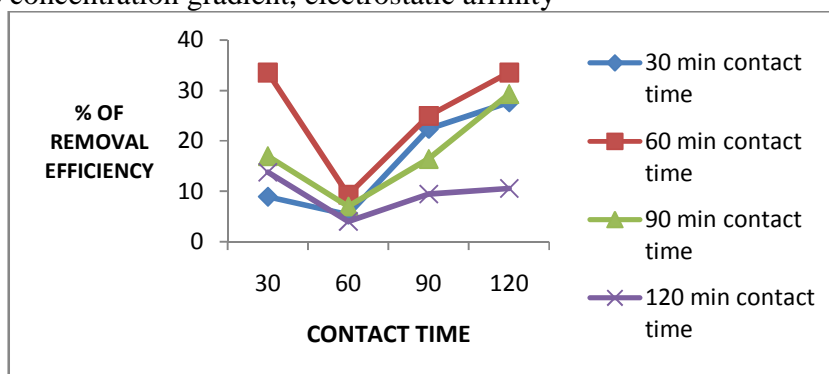


Figure:2

C. OPTIMAL ADSORBENT DOSE

It was seen that removal of fluoride increases with an increase in the amount of adsorbent. For all the experiments, initial fluoride ion concentration was fixed at 10 mg/ litre. PH was 7 and optimum contact time was 2.5 hrs. The amount of adsorbent dose was varied from 1.5gm to 3gm in aqueous solutions. To study the effect of an increase in adsorbent dosage on removal of fluoride experiments were conducted employing adsorbent dose ranging from 1.5gm to 3gm of adsorbents. When the Ramacham powder is used it was found that maximum removal (24.29%) was observed at 3gm of adsorbent dose. When the Chukku powder is used it was found that maximum removal (28.51%) was observed at 1.5 gm of adsorbent dose. When the Pathimugam powder is used it was found that maximum removal (9.48%) was observed at 2 gm of adsorbent dose. when the Grambu powder is used it was found that maximum removal (36.67%) was observed at 2gm of adsorbent dose. The results show that fluoride uptake was high for 1.5 to 3gm of adsorbents. This might be due to the fact that, if the dose concentration of adsorbent is more, more adsorbent surface and more pore volume would be available for the adsorption interaction and this result in high fluoride removal.

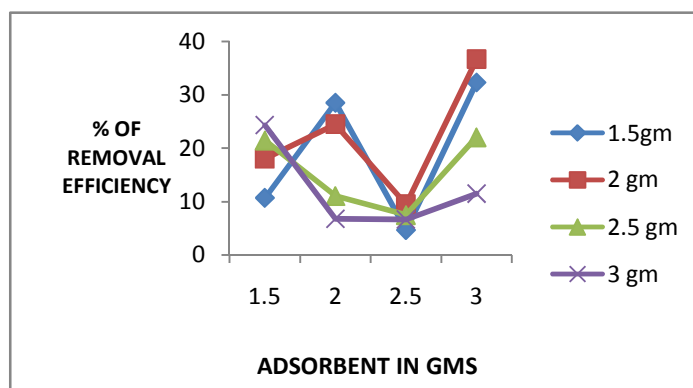
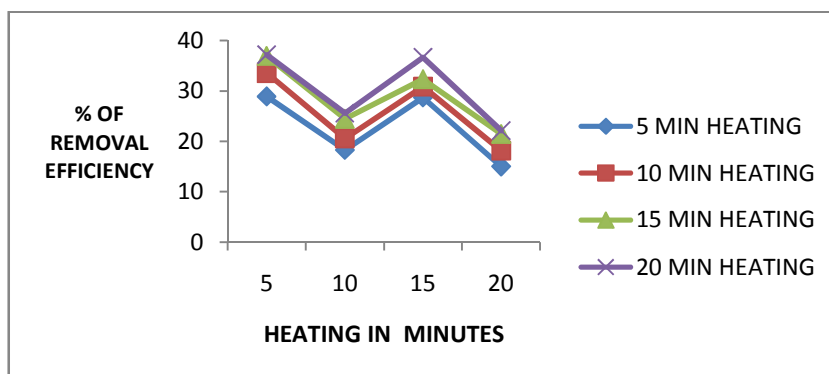


Figure: 3**D.OPTIMAL HEATING MINUTES**

It was seen that the removal of fluoride decreases with increase of temperature. For all the experiments; the initial fluoride concentration was fixed at 10mg/litre. pH was 7 and biosorbent dose of 1.5 gm. Experiment is carried with different heating time of 5 mts, 10 mts, 15mts and 20 mts and with 30^o temperatures. When the Ramacham powder is used it was found that maximum removal (37.16%) was observed at a heating temperature of 30 degree. When the Chukku powder is used it was found that maximum removal (25.57%) was observed at a heating temperature of 30 degree. When the Pathimugam powder is used it was found that maximum removal (22.11%) was observed at a heating temperature of 30 degree. when the Grambu powder is used it was found that maximum removal (36.67%) was observed at a heating temperature of 30 degree centigrade. The percentage of biosorption decreased with increase of temperature. This might be due to the weakening of sorptive forces between active sites of the biosorbent and adsorbate species and also between the adjacent molecules of adsorbed phase. The findings favour this concept. The percentage of sorption was high at 30 degree temperature. This was similar to the findings of viola rose (2014). Hence, this study can reveal that many such adsorbents can be used for defluoridation of drinking water. This can be implemented at domestic or community levels.

**Figure:4****VIII.CONCLUSIONS:**

The purpose of this work was to develop a method for defluoridation of water at home itself using medicinal herbs. The adsorbents used are easily available. This method is so simple. The adsorbents are environment friendly and cause no harm to the user and environment.

1. After 60 minutes there is decrease in adsorption in all four biosorbents.
2. Chukku shows maximum fluoride removal at pH 6.
3. Fluoride uptake was high when the adsorbent dose varies from 1.5 to 3gm of adsorbents.
4. The percentage of biosorption decreased with increase in heating time.

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