

## TAMARIND SEED: THE ECONOMICAL BIOADSORBENT FOR DEFLUORIDATION OF WATER

S. S. Mokashi<sup>1</sup>, A. S. Parlikar<sup>2</sup>

<sup>1</sup>Asst. Prof. in Engg. Chemistry , SVPM's College of Engineering, Malegaon(Bk),  
Baramati Dist. Pune (Maharashtra)

<sup>2</sup>Asst. Prof. in civil Engg. SVPM's College of Engineering, Malegaon(Bk), Baramati Dist. Pune  
(Maharashtra)

### Abstract

Adsorption is widely used to remove various pollutants & contaminants from water. The water available for daily consumption may be contaminated by natural sources or by industrial effluents. One such a contaminant is fluoride. Exposure to fluoride in drinking water has a number of adverse effects on human health. One of the naturally available alternatives for defluoridation of water is Tamarind Seed. Tamarind Seed is cheap & easily available bioadsorbent to remove fluoride content from water. It is a kitchen waste & easily available. Spectrophotometric studies can be efficiently conducted to get optimal values of pH of water in acid and alkali treated powder of seeds of Tamarind. Optimal contact time for 212 $\mu$  and 600 $\mu$  were determined. For higher removal percentage efficiency of fluoride, optimal dose of adsorbent was also determined.

**Keywords:** Fluoride, adsorption, Tamarind Seed, contact time, P<sup>H</sup>, particle size, adsorbent dose.

---

### INTRODUCTION

Water is our life. It is most abundant and essential component for human body for different functions. But the availability of pure water is very limited. Groundwater is the prime source of 60 percent of India's households. But in groundwater high Fluoride content has been reported in various parts of the world. According to World Health Organization (WHO 1984), the maximum acceptable fluoride concentration in drinking water is considered to be safe for human consumption is only upto 1.0 mg/lit. In many parts of the world fluoride occurs. It occurs in some region of Afghanistan, Algeria, Australia, Burundi, China, Japan, India, Kenya, Nepal, Pakistan, Thailand, Uganda, Zimbabwe and Zambia. In India, fluoride occurs in some regions of Assam, Andhra Pradesh, Bihar, Gujarat, Haryana, Jammu Kashmir, Karnataka, Kerala, Maharashtra, Madhya Pradesh, Orissa, Punjab, Rajasthan, Tamilnadu, Uttar Pradesh and West Bengal. The effects of fluoride on human health are dependent on the concentration of fluoride in water<sup>(21)</sup>.

**Table 1 – Biological Effects on Human Health**

Fluoride conc. (mg/lit)	Source	Effects
1	Water	Prevention of dental caries
2	Water	Effect dental enamel
3 to 6	Water	Osteoporosis
8	Water	10 % Osteoporosis
20 to 80	Air & Water	Crippling skeletal fluorosis
50	Food & Water	Changes in thyroid
100	Food & Water	Defective development
>125	Food & Water	Changes in Kidney
2500	Acute dose	Death

Following are the permissible limits for fluoride concentration in drinking water<sup>(19)</sup>

1. World health Organization (WHO-1984) -1 to 1.5 mg/lit
2. Bureau of Indian Standards (BIS)-0.6 to 1.2 mg/lit
3. Indian Council of Medical Research (ICMR-1975)-1 mg/lit

Deficiency of fluoride intake leads to dental caries while excess consumption leads to dental and skeletal fluorosis. Fluoride is more toxic than lead. Its higher quantity may cause damage to brain/mind development of children. It produces abnormal behavior in animals and reduces IQ in humans (1). Due to high toxicity of fluoride to mankind, now there is urgent need to make the drinking water safe for human consumption. The conventional methods of fluoride removal include, Ion-Exchange, Reverse Osmosis and Adsorption. But adsorption is cheaper and convenient than Ion-Exchange and Reverse Osmosis for the removal of fluoride. For drinking water treatment, coagulants are used which may be chemical or naturally occurring coagulant. Some studies on natural coagulants have been carried out and various natural coagulants were produced or extracted from microorganisms, animals or plants. One of these alternatives is Tamarind seeds. Practically bioadsorbtivedefluoridation study is limited and so in present study, the Tamarind Seed is used for defluoridation purpose. It is a household material and left as waste after removing Tamarind pulp for preparation of food.

In Nalgonda technique<sup>(17)</sup> a sequential addition of sodium aluminates or lime, bleaching powder and filter alum to the fluoride water is followed by flocculation, sedimentation and filtration. The disinfection is ensured by sodium aluminates settlement of precipitate and bleaching powder. The poly aluminum hydroxylsulphate (PAHS) was used for defluoridation studies<sup>(10)</sup> and it was concluded that, floc formation and settling are quick and volume of resulting sludge is less. The use of Aloe Vera (Indian aloe), a medicinal plant was done<sup>(14)</sup> concluding maximum defluoridation at neutral pH. A study of defluoridation methods was done<sup>(21)</sup> by using indigenous chemicals and minerals. Moringa Oliefera<sup>(2)</sup> is one of the effective natural adsorbent for defluoridation of water. For optimum alum dose, alkalinity of water, pH and colloidal concentration, effective defluoridation can be ensured. The fluoride removal technique by using poly aluminum chloride (PAC) was studied<sup>(14)</sup> and compared with Nalgonda technique. It was observed that PAC can be an effective coagulant for fluoride removal with higher removal efficiency of about 65% -75% with less detention time. Also it was observed that fluoride removal was dependent on initial fluoride ion concentration and dose of coagulant. Defluoridation techniques by using lignite rice husk and rice husk powder as adsorbent was

studied<sup>(19)</sup> by varying pH, concentration of fluoride, weight of adsorbent and contact time. A better result was obtained after 2 hrs at pH 6 by using lignite. Tea waste<sup>(22)</sup> is also one of the economical bioadsorbent. Fishbone charcoal prepared from fishbone was used for defluoridation studies. The fluoride removal was found to be a function of pH, contact time, initial fluoride ion concentration and adsorbent (fishbone charcoal) dose. The boiler bottom ash was successfully used<sup>(4)</sup> as an adsorbent material for separating fluoride from water for the pH between 6 to 7.

The Tamarind tree grows in tropical and subtropical regions around the world. Tamarind seeds are non-toxic. As the Tamarind seed is biological and has been reported as edible, it has an added advantage over the chemical treatment of water. It mainly consists of polysaccharides. It is familiar to all kinds of people in villages as well as in cities and so can be conveniently used for defluoridation of water.

## Methodology

Adsorption is defined as the change in concentration at the interfacial layer between the two phases of a system due to surface forces. Adsorption is mass transfer operation. The adsorbate is substance that is being removed from the liquid phase and transferred to the solid phase. The adsorbent is the solid, liquid, or gaseous phase onto which the adsorbate accumulates. Factors affecting adsorption methods are i) surface area ii) nature of the adsorbate iii) pH iv) temperature v) presence of mixed solutes and vi) nature of adsorbent.

For preparing synthetic fluoride water sample anhydrous sodium fluoride (NaF) and distilled water were used. For preparing adsorbent powder, Tamarind seeds are used which was collected from the kitchen as a waste material. It was soaked in water for one to two hours. The pulp obtained is removed. The seeds are washed with tap water then double distilled water dried in an oven at 110°C for an hour and then grinded in mill. About 40 gm of powder sample was added to 400 ml of 1N HNO<sub>3</sub> for acid treatment and 0.5N NaOH for alkali treatment. Finally, it was dried again in an oven at 50°C for 6 hrs. The nitric acid (1N HNO<sub>3</sub>) was used for acid washing of adsorbent. The sodium hydroxide (0.5 N NaOH) was used for alkali washing of adsorbent. For fluoride detection studies with spectrophotometer, various solutions were prepared. Reference solution was prepared by using conc. hydrochloric acid (HCl), SPADNS reagent zirconyl chloride, octahydrate reagent were used. Synthetic fluoride bearing water sample having initial fluoride ion concentration of 10 mg/lit used. The sample was filtered by using Whatmann's filter paper no.41 for further uses. In this filtrate, SPADNS and zirconyl acid solution of 5ml each was used. The sample was checked for fluoride detection in spectrophotometer at wavelength 570nm.

The fluoride removal studies by adsorption were conducted in 250 ml conical flask using 100 ml of synthetic water sample containing different pH and initial concentrations of fluoride ion. In these conical flasks adsorbent with varied dosage was added. Then the contact period was given for different particle sizes. After giving a required contact time, the contents of the flasks were filtered using Whatmann's filter paper number 41. The filtrate was used for fluoride ion estimation using SPADNS method.

The above procedure was repeated for different pH, contact times, adsorbent doses, particle sizes and different initial fluoride ion concentrations. The pH was varied from 1 to 10. The contact time was varied from 0.5 hr to 3.5 hrs for various adsorbent sizes. The adsorbent dosages used were 0.5 gm/lit to 4 gm/lit in multiple of 0.5 gm/lit. The initial fluoride ion concentration was varied from 2 mg/lit to 11 mg/lit for the Tamarind seed powder. The parameters were varied to find the maximum fluoride removal efficiency. Synthetic fluoride bearing water sample having initial fluoride ion concentration of 10 mg/lit was used.

## Results and Discussion

### Effect of pH

The experiments were carried out for acid treated and alkali treated Tamarind seeds powder for determining optimum pH. The procedure was similar for acid and alkali treated powder. The pH was varied from 1 to 10 for acid treated Tamarind seed powder and 2 to 10 for alkali treated Tamarind seed powder. The adsorbent having 600  $\mu$  size, acid washed as well as alkali washed, was used to determine optimal pH at which the adsorption was maximum. For these experiments initial fluoride ion concentration was 10 mg/lit, with adsorbent dose of 2.5 gm/lit and contact time of 1 hr. In case of acid washed adsorbent the maximum removal efficiency was 37 % at pH 1. Whereas in case of alkali washed adsorbent the maximum removal efficiency was 51 % at pH 10. The extreme pH values will give rise to higher costs for post treatment. Therefore it is not advisable to adopt extreme pH values. It is generally recommended to maintain near neutral pH for the solution. At pH 8, the percentage removal was 13% and 50% for acid washed and alkali washed adsorbents respectively. Therefore, it was decided to use alkali washed adsorbent and to maintain pH 8.

**Table 2- Determination of optimum pH**

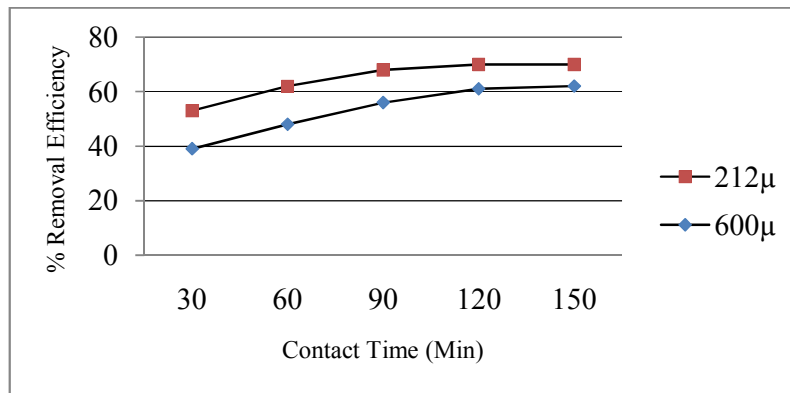
Sr. No.	pH	Acid washed powder (600 $\mu$ )		Alkali Washed Powder (600 $\mu$ )	
		Absorbance	% Removal Efficiency	Absorbance	% Removal Efficiency
1	1	0.680	37	-	-
2	2	0.765	31	0.760	31
3	4	0.815	24	0.730	37
4	6	0.850	20	0.665	41
5	8	0.905	13	0.595	50
6	10	0.920	12	0.580	51

### Effect Contact Time

The adsorbent dose of 2.5 gm/lit was taken and kept constant throughout the experimental work. The contact time was varied from 0.5 to 2.5 hrs for alkali treated Tamarind seed powder of 600 $\mu$  and 212 $\mu$  respectively. The experimental study was carried out to determine optimal contact time using adsorbents with different particle sizes. The pH was 8 and dose was 2.5 gm/lit for the study. It was observed that the contact time reduces with decrease in particle size. For the given particle size, after a particular contact time, the removal efficiency remains almost constant. Contact time of 2.0hrs and 2.5hrs were optimal for adsorbents having particle size of 212  $\mu$  and 600  $\mu$  respectively.

**Table 3- Optimum contact time**

Sr. No.	Contact Time(min)	600 $\mu$		212 $\mu$	
		Absorbance	% Removal Efficiency	Absorbance	% Removal Efficiency
1	30	0.677	39	0.565	53
2	60	0.625	48	0.480	62
3	90	0.524	56	0.520	68
4	120	0.475	61	0.415	70
5	150	0.485	62	0.415	70



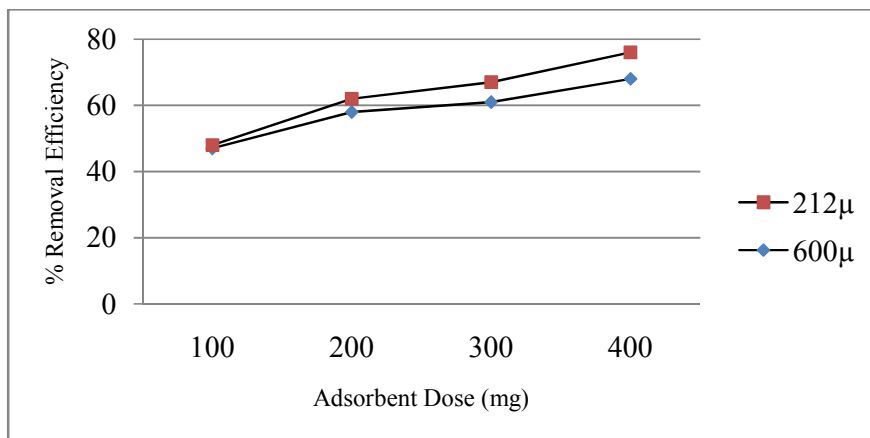
**Fig.1 -Optimum contact time**

**Effect Adsorbent Dose**

It was seen that the 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/lit, pH was 8, and optimum contact time was 2.5hrs and 2 hrs for 600 µ and 212 µ respectively. The amount of adsorbent dose was varied from 0.5 gm/lit to 4 gm/lit in aqueous solutions. Results show that for 212µ alkali treated Tamarind seed powder bioadsorbent, the maximum removal efficiency of fluoride was 76 % at 400mg/lit. Similarly, for 600µ alkali treated bioadsorbent, the maximum removal efficiency of fluoride was 68 % at 400 mg/lit.

**Table 4- Optimal adsorbent dose**

Sr. No.	Adsorbent Dose (mg)	600 µ		212 µ	
		Absorbance	% Removal Efficiency	Absorbance	% Removal Efficiency
1	100	0.617	46	0.615	48
2	200	0.525	58	0.485	62
3	300	0.490	61	0.525	67
4	400	0.525	68	0.365	76



**Fig.2- Optimal adsorbent dose****CONCLUSIONS**

From the adsorption studies using Tamarind seed powder, it is concluded that

1. Use of the Tamarind seed powder as cheap bioadsorbent for removal of fluoride is feasible.
2. The alkali treated Tamarind seed powder was found better than acid treated Tamarind seed powder for fluoride ion removal.
3. The removal by adsorption was found to be optimum at adsorbent dose of 400 mg/lit.
4. The effective contact time was 2.0hrs and 2.5hrs for 212  $\mu$  and 600  $\mu$  respectively.

**References**

1. Awadia A.K., Haugejorden O., Bjorvatn K., Birkeland J.M. (1999), "Vegetarianism and Dental Fluorosis among Children in a high Fluoride Area of Northern Tanzania", *International Journal of Paediatric Dentistry*, 9(1), pp3-11.
2. A.S. Parlikar, S.S. Mokashi (2013) "Defluoridation of water by Moringa Oleifera -a natural adsorbent". *International Journal of Engineering Science and Innovative Technology*. 2, (5), pp245-252
3. Bhargava D.S., Killedar D.J. (2006), "Fluoride adsorption on fishbone charcoal", *Journal of IAEM*, 33(3), pp179-183.
4. Ganguly S., Mukharjee S.N., Misra A.K. (2006), "Removal of Fluoride from aqueous phase using boiler bottom ash as an adsorbent", *Journal of Indian Water Works Association*, pp 47-56.
5. Gopal V., Elango K.P. (2007), "Adsorption of Fluoride from Aqueous Solution by Active Carbon", *Journal of Indian Water Works Association*, pp183-191.
6. Gowda Rudre, Nataraj, A.G., Rao Manamohan, N (2011). "Removal of Nickel (II) from Electroplating Industrial Effluents using Coconut Leaves as low cost Adsorbent." *Journal of Indian Association for Environmental Management*, vol. 38 (2) pp 69-74.
7. Jamode A. V., Sapkal V. S., Jamode V. S. (2004), "Uptake of fluoride ions using leaf powder of *Ficus religiosa*", *Journal of Indian Water Works Association*, pp53-61.
8. Jamode A.V., Sapkal V.S. and Jamode V.S. (2004), "Defluoridation of water using inexpensive adsorbents", *Journal of Indian Institute of Science*, 84, pp163-171.
9. Kaseva M.E. (2006), "Optimization of regenerated bone char for fluoride removal in drinking water: A case study in Tanzania", *Journal of Water and Health*, 4(1), pp139-147.
10. Mariappan P., Yegnaraman V., Vasudevan T. (2002), "Defluoridation of water using PAHS", *Journal of Indian Water Works Association*, pp217-223.
11. Mariappan P., Yegnaraman V., Vasudevan T. (2000), "Occurrence and removal possibilities of fluoride in groundwater of India", *Enviromedia*, 19(2), pp165-177.
12. Mariappan P., Yegnaraman V., Vasudevan T. (2003), "Defluoridation of water using Alumina-Gibbsite", *Indian Journal of Environmental Protection*, 23(9), pp 975-985.
13. Mises Shivakant R., Shivarajappa (2009) "Adsorption Studies of Chromium (VI) on activated carbon derived from Phoenix Dactylifera seeds", *Journal of IPHE, India*, vol. 2009-10 (1) pp 5-13.
14. Murugan M., Subramanian E. (2002), "Application of Aloe Vera (Indian Aloe) – A Plant Material for Defluoridation", *Indian Journal of Environmental Protection*, 22(9), pp1034-1039.
15. Murugan M., Subramanian E. (2006), "Studies on Defluoridation of water by Tamarind seed and unconventional biosorbent", *Journal of Water and Health*, 4(4), pp 453-461.
16. Muthu Ganesh I., Vinodhini V., Padmapriya G., Sathiyarayanan K., Sabumon P.C. (2003), "An Improved Method for Defluoridation", *Indian Journal of Environmental and Health*, 45(1), pp65-72.

17. Nawlakhe, W.G., Kulkarni D. N., Pathak B.N.(1975),“Defluoridation of water by Nalgonda Techniques”,*Indian Journal of Environmental Health*, 17(1), pp26-64.
18. Pathak M. D.,Badave R. (1999), “Occurrence of fluoride in groundwater of Maharashtra”,*Journal of IAEM*, 26,pp168-171
19. Prabavathi N.T., Ramachandramoorthy R., Edison R., Kavitha B., SivrajC.,Srinivasan (2003),“Drinking water of Salem District-Estimation of fluoride and its Defluoridation Using Lignite Rice Husk and Rice Husk Powder”,*Indian journal of Environmental Protection*, 23(3), pp 304-308.
20. Prakasham R. S., Reddy P.C., Amita, Manisha, Ramakrishna S. V(1999), “Defluoridation of Drinking Water Using water hyacinth”, *Indian journal of Environmental protection*, 19(2),pp119-124.
21. Sanjaykumar, Krishna Gopal, Seth P.K.(2002),“Indigenous Minerals for Defluoridation of Water”, *Indian Journal of Environmental Protection*, 22(4), pp361-372.
22. S.S.Mokashi, A.S.Parlikar ( 2014 ) Feasibility of Tea Waste as an adsorbent for defluoridation of water. *International Journal of Engineering Science and Innovative Technology*. 3 (5),pp232-238
23. Varadajan N., Purandara B. K.(2008), “Fluoride Contamination in Ground water of Malaprabha Sub Basin”,*Jouranal of Environ Science and Engg.*, 50(2), pp121-126.
24. Weginwar N., Takarkhede A. G.,Kamble R. K.(2008),“Ground Water Fluoride in Chandrapur city in Maharashtra”, *Indian Journal of Environmental Protection*, 28(7), pp 648-653.