

Economical technology for fluoride removal from drinking water using tea ash and fish bone

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ABSTRACT

Fluoride contamination in drinking water due to natural and anthropogenic activities has been recognized as one of the major issue imposing a serious threat to human health. Among several treatment technologies applied for fluoride removal, adsorption process has been explored widely and offers satisfactory results, so objective of this study was to investigate or check efficiency of low cost adsorbent or locally available materials (Tea ash and fish bone) for the removal of excess fluoride from aqueous solution. Excess fluorides can cause skeletal and dental fluorosis. The optimum fluoride level in drinking water for general good health set by WHO is considered to be between 0.5 and 1.2 mg/l. Recently, scientists are mostly focusing their study on various types of inexpensive and effective materials. So, there is an urgent need to explore low cost & locally available defluoridation materials for safe and easy use at both household and small community levels. Residual parts of tea and fish bone as household waste were effectively used for removal of fluoride from aqueous medium. It was also shown that tea ash has enough potential to remove fluoride from aqueous solution and efficiency of removal of fluoride is 64.29%. And fish bone removes fluoride from drinking water and its defluoridation efficiency is 42.86%. Compared with traditional fluoride removal methods, the locally available materials such as tea ash and fish bone could be used as a cost-effective adsorbent for efficient fluoride removal from water.

KEY WORDS: Tea ash, economy, fluoride.

1. INTRODUCTION

Defluoridation refers to methods of water treatment that reduce the concentration of fluoride in the water, normally, in order to make it safe for human consumption. Some water treatments that have the capacity of reducing the fluoride concentration along with most other anions, or anions and cations in the water, are not considered as defluoridation methods. Thus general demineralising methods like distillation, reverse osmosis, electro dialysis and resin de-anionisation, which are able to remove fluoride fully or partly from the water, are not considered as defluoridation methods. On the other hand methods that only remove fluoride without any addition or reduction of other parameters are not yet discovered.

The objective of defluoridation is to prevent or to mitigate endemic fluorosis. Over the years groundwater has generally been considered to be a protected and safe Source: of water, fit for drinking without treatment, as the main focus has been on the bacteriological quality of potable water. The presence of fluoride in exceeding limits and its related problems of drinking water prevailing in many parts of India is well documented. Fluoride in drinking water is known for both beneficial and detrimental effects on health. Fluoride from water or wastewater can be removed by an ion exchange/adsorption process or by coagulation, precipitation process.

The ion exchange/adsorption can be applied to either concentrate or diluted solutions and they are capable of achieving complete removal under proper conditions. The method suitable for a given situation needs to be judiciously selected considering the various aspects. So, there is an urgent need to explore low cost & locally available defluoridation materials for safe and easy use at both household and small community levels. Residual parts of tea and fish bone as household waste were effectively used for removal of fluoride from aqueous medium. Fluoride is beneficial to health if the concentration (CF) of the fluoride ion (F⁻) in drinking water is less than 1.2 mg/L (WHO 1994). A higher concentration causes serious health hazards. The disease caused manifests itself in three forms, namely, dental, skeletal, and non-skeletal fluorosis. Dental fluorosis produces widespread brown stains on teeth and may cause pitting. Skeletal fluorosis causes crippling and severe pain and stiffness of the backbone and joints. The main objectives of the studies are

- To use locally available materials that has properties to remove excess fluoride from drinking water
- To reduce the fluoride of drinking water without disturbing the other physiochemical characteristics
- To compare the efficiency of defluoridation by both tea ash and fish bone using adsorption method

2. MATERIALS AND METHODS

2.1. Study area: In Tamil Nadu, the high concentration of fluoride in groundwater is found to be in Dharmapuri and Salem district closely followed by Coimbatore, Madurai, Trichy, Dindukal and Chidambaram district. The study area, lies between the longitude of 77°50'53" to 77°52'43" and North latitude of 11° 53'43" to 12°19'49" covering an area of 1096.22 km². In these, Plain area covers an area of 604.48 Km² (Fig.1). The study area falls in Pennagarm taluk, Dharmapuri district of Northern Tamil Nadu. The major Source: for recharge of water in this

area is rainfall, during monsoon season. The average annual rainfall is 980 mm (2001 to 2011). As the study area is underlain by the Archaean crystalline rock, groundwater may occur in the fractured rocks. Geologically, the study area is mainly underlain by charnockite, hornblende biotite gneiss and Epidote-hornblende gneiss. Charnockite is the dominant group of rocks covering major parts of the study area, followed by the hornblende biotite gneiss. Hornblende biotite gneiss and Epidote-hornblende gneiss area is high thickness of the formation (Top soil and weathered zone) related to the water quality for high concentration.

2.2. Water sample analysis: The water samples are collected from pennagaram, Dharmapuri district for defluoridation process. The samples are involved in the series of water parameters tests and analysis is prepared based on their tests. In this analysis, the fluoride test is considered as one of the essential test and this test is repeated more time. The tests to be carried with tea ash and fish bone done in separately. For getting better efficiency, the defluoridation methods are divided into two methods such as defluoridation with tea ash and fish bone. In this first method, the residue tea ash from house is considered as medium which downs the excess fluoride from drinking water when sample passes through it. In second techniques, the fish bone from house waste is taken as medium that reduces the fluoride content from drinking water effectively.

2.3. Defluoridation with Fly Ash: The tea residue collected from house was used for the experiments. Soluble and colored components were removed from tea by washing with boiling water. This was repeated until the water was virtually colorless. After thoroughly washing, the adsorbent was sun dried and it is burned in muffle furnace at 500°C for 30 minutes and dried tea ash was sieved and stored in sealed polythene bags. The activated tea ash powder (AcTAP) material was subjected to various physicochemical parameters and used for sorption. The fraction 50 μm was used for all the experiments except for the effect of particle size tests. Adsorption experiments were carried out for the determination of pH, adsorbent dose variation, equilibrium time and kinetics, selection of an isotherm, effect of temperature and evaluation of thermodynamic parameters.

The influence of pH (2.0-10.0), adsorbent concentration (0.2-2.4 g/l) and contact time (40, 60, 120, 180, 240 and 300 min), initial fluoride concentration (1.5,5.0,7.0,10.0,15.0,35.0 and 50.0 mg /l) and temperature (303, 313, 323 and 333K) were evaluated during the present study in a 250 mL Erlenmeyer flasks and 100 ml of fluoride solution of known concentration was added for constant shaking (at 300 rpm) during 1hr in a temperature controlled magnetic stirrer at $303 \pm 1\text{K}$ and then the solids were separated through filtration. The solutions were collected for analysis and fluoride concentration in the solution was determined by using ion selective meter. Each experiment was conducted three times and average values are reported. Control experiments, performed without addition of adsorbent, confirmed that the sorption of fluoride on the walls of Erlenmeyer flasks was negligible.

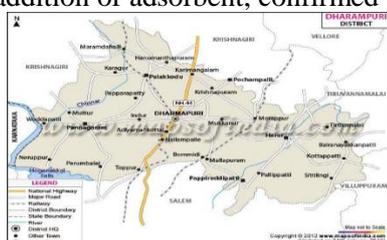


Figure.1.Map of Dharmapuri



Figure.2.Map of Dharmapuri surroundings



Figure.3.Map of Pennagaram



Figure.4.Defluoridation setup



Figure.5.Defluoridation with tea ash



Figure.6.Fish bone defluoridation test

3. RESULTS AND DISCUSSION

The water samples were collected from Pennagaram taluka, Dharmapuri district and involved in the physiochemical tests. Table 2 showing the physiochemical characteristics of drinking water before defluoridation.

Table.1. Physico chemical characteristics of drinking water before defluoridation, after tea ash defluoridation and after fish bone defluoridation

Parameters	Values		
	Before defluoridation	After tea ash defluoridation	After fish bone defluoridation
pH	6.66	6.66	6.66
Conductivity S m /cm	1400	1400	1400
Fluoride mg/L as (F-1)	1.4	0.5	0.8
Total hardness mg/L as Ca and Mg	396	396	396
Chloride mg/L as (Cl-1)	378	378	378
Sulphate mg/L as (SO ₄ -2)	108	108	108
Total dissolved solids mg/l	882	882	882
Silica mg /L	40	40	40
Methyl orange alkalinity mg/ l as caco ₃	162	162	162
Iron mg /L as Fe+2	0.09	0.09	0.09
Turbidity as (NTU)	1.8	1.8	1.8

3.1. Defluoridation with tea ash: The physicochemical characteristics of tea ash are given. The surface characteristics make it suitable for sorptive defluoridation. The sorption is characterized under determination of zero point charge (pH^{ZPC}) of the adsorbent. Adsorption of cation is favored at $\text{pH} > \text{pH}^{\text{ZPC}}$, while the adsorption of anion is favored at $\text{pH} < \text{pH}^{\text{ZPC}}$.

The effect of pH on the fluoride removal from aqueous solution by using AcTAP is shown in Figure 6.2. This result shows that removal of fluoride ion is most appreciable in the pH range 2.0-7.0 with a peak value at pH 6.0. This may be explained by considering pH^{ZPC} as shown in Table 6.1 for the AcTAP because removal of fluoride by adsorption process is highly dependent on solution pH as it can alter the surface charge of the adsorbent. Hence, in the present work, the highest removal capacity was performed at pH 6.0, since $\text{pH}^{\text{ZPC}} = 6.5$, the surface will be positively charged. Decrease in removal capacity was observed below and above pH 6.0 as shown in Table 6.2, which may be attributed to unfavorable surface charges and competition for adsorption sites due to excess anions at alkaline conditions.

Effect of pH on removal of fluoride onto AcTAP (experimental conditions: Initial fluoride concentration: 5.0 mg/l, adsorbent dose: 0.8 g/l with particle size 100 μm , agitation speed: 300 rpm, Contact time: 60 minutes, Temperature: 303 K. The effect of adsorbent dose on fluoride adsorption was studied at fixed initial concentration (5 mg/l) as shown in Figure 6.4. The extent of percentage of adsorption increased with increase in the adsorbent dose while loading capacity (amount of fluoride adsorbed per g of the adsorbent) gradually decreased for decreasing the number of interaction site between the adsorbent and adsorbate. This is probably due to increase of adsorption sites. From the trend it appears that a maximum level (51.3 to 97.6 %) of fluoride adsorption occurs with increase in adsorbent dose from 0.2 to 2.4 g/l and then the adsorption is not changed with the adsorbent dose due to establishment of equilibrium condition. Effect of Adsorbent dose on the adsorption of fluoride by AcTAP (experimental conditions: Initial fluoride concentration: 5.0 mg/l, pH: 6.0, particle size: 50 μm , agitation speed: 300 rpm, Contact time: 60 minutes, Temperature: 303 K)

The adsorption rate of fluoride at different initial concentrations (5.0, 10.0 and 50.0 mg/l) was investigated by varying the time ranging from 40 to 300 min as shown in Figure 6.5. Figure 6.6 also shows that with a fixed amount of AcTAP, the adsorption of fluoride increased with time and then attained equilibrium after 180 min at different initial concentrations indicating the independent nature of time to reach equilibrium conditions. Effect of contact time on removal of fluoride onto AcTAP at various concentrations (experimental conditions: pH: 6.0, adsorbent dose: 2.4 g/l and particle size 50 μm , agitation speed: 300 rpm, Temperature: 303 K). Effect of contact time on removal of fluoride onto AcTAP (experimental conditions: Initial fluoride concentration: 5.0 mg/l, pH: 6.0, adsorbent dose: 2.4 g/l and particle size 50 μm , agitation speed: 300 rpm, Temperature: 303 K)

3.2. Defluoridation with fish bone: The breakthrough curves for the initial fluoride concentration (C_0) of 5.00 mg/l corresponding to the various influent flow rates ($Q = 15, 25, 50$ and 75 ml/min) are depicted in Figure 6.6. The retention time or hydraulic residence time corresponding to the flow rates (Q) of 15, 25, 50 and 75 ml/min are 4.91, 2.95, 1.47 and 0.98 min, respectively. The volume of treated effluent (V) at the different flow rates at the chosen effluent fluoride concentration at breakpoint, C_e of 1.0 mg/l is read as 9.45, 8.25, 3.27 and 2.87 l corresponding to the flow rates of 15, 25, 50 and 75 ml/min, respectively. The variation of treated effluent volume (V) with respect to the flow rate per unit weight of adsorbent (Q_w) is shown in Figure 6.7. Retention time (or

hydraulic residence time) of solution in the column may be inadequate. The optimum time of retention required for completing the sorption reaction between the solute and the media, which is likely to result in longer operation period until the breakthrough at $C_e = 1.0$ mg/l takes place, may require lower flow rates. Mass balance calculations were done to determine the amount of fluoride removed (f) at different flow rates as shown in Figure 7.6 till the breakthrough concentration point ($C_e = 1.0$ mg/l). The effluent concentration up to the breakthrough point varied from 0 to 1.0 mg/l as seen in Figure. During the breakthrough time, an average breakthrough effluent fluoride concentration (C_{e-av}) of 0.5 mg/l was assumed for the time interval in which C varied from 0 to 1.0 mg/l instead of using the procedure of integration of breakthrough curve or considering small time intervals. The amount of fluoride removed for flow rates of 15, 25, 50, and 75 ml/min is estimated to be 42.5 mg, 37.1 mg, 14.7 mg, and 12.8 mg, respectively. These correspond to removal capacities of approximately 1, 0.8, 0.4 and 0.3 mg/g respectively. The higher the flow rate the lesser the fluoride removal capacity, due to shorter available contact time (or hydraulic residence time).

Breakthrough curves at the different initial fluoride concentration (C_0) are shown in Figure 6.7. These curves are obtained corresponding to a constant flow rate of 15 ml/min. From Figure 3 the values of the useful volume (V) of the treated effluents corresponding to $C_e = 1.00$ mg/l were estimated as 18.8, 8.1, 5.4 and 3.24 l corresponding to C_0 values of 2.5, 6.0, 10.0 and 20.0 mg/l, respectively. The variation of volume of the treated effluent versus the initial fluoride concentration (C_0) is shown in Figure 6.8. At the chosen breakpoint concentration of $C_e = 1.0$ mg/l, the useful volume (V) of the treated effluent decreases with an increase in C_0 values. This is due to the fact that for a given flow rate and quantity of the adsorbent, the adsorption or exchange sites of the adsorbent are exhausted earlier when a higher initial fluoride concentration influent is encountered.

Therefore, the operation period until the breakthrough point is less. Mass balance calculations were carried out, as before, to determine the amount of fluoride removed at different initial fluoride concentrations Figure 6.8. The amount of removed fluoride (F) at C_0 of 2.5, 6.0, 10.0 and 20.0 mg/l was 28.2 mg, 44.6 mg, 51.30 mg, and 63.18 mg, respectively. These correspond to useful removal capacities of about 0.8, 1, 1.1 and 1.4 mg/g respectively. The breakthrough times are shorter for higher initial fluoride concentrations, but the amount of fluoride removed, and the useful removal capacity of the medium, increases with C_0 . This is probably due to higher concentration gradients at higher C_0 values.

Table.2. Effect of pH on adsorption of fluoride by AcTAP

pH of the solution, mg/g	q_e
2.0	5.45
4.0	5.52
6.0	5.59
7.0	4.69
8.0	3.27
10.0	3.22

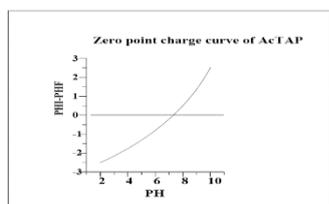


Figure.7.Zero point charge curve

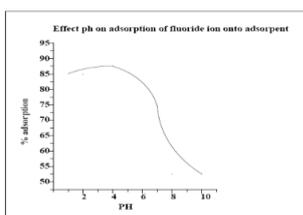


Figure.8.Effect of pH

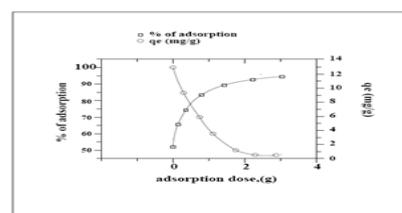


Figure.9.Effect of adsorbent dose

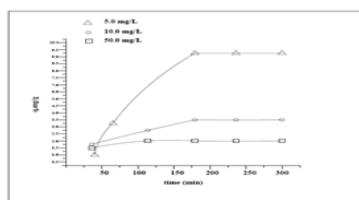


Figure.10.Effect of contact time at various concentration

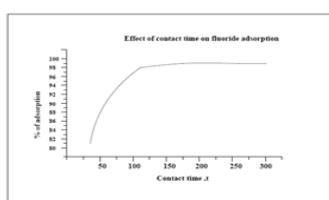


Figure.11.Effect of contact time

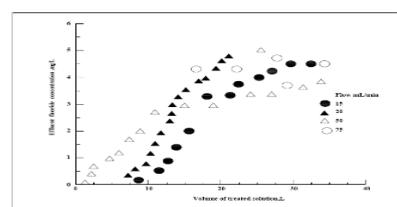


Figure.12.Break through curve corresponding to different flows with column bed depth and adsorbent weight

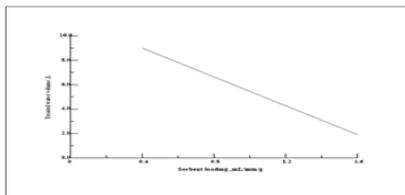


Figure.13. Variation of useful treated effluent volume (V) with influent flow rate per unit weight of adsorbent (Q_w)

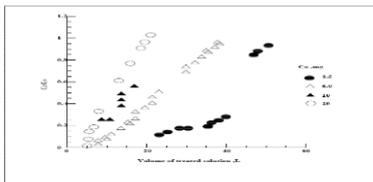


Figure.14. Breakthrough curves (C vs L) corresponding to different initial fluoride concentrations (C_0) with column bed depth (D) and Adsorbent weight (W)

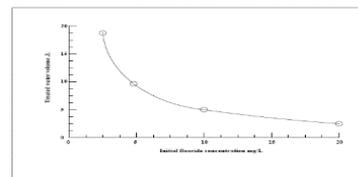


Figure.15. Variation of useful treated effluent volume (V) with initial fluoride concentrations (C_0)

4. CONCLUSION

A new medium, waste tea ash has been developed for fluoride removal in this report. Numerous advantages of the waste tea ash make the waste an excellent adsorbent for removal of fluoride from aqueous solution under acidic pH and further increases of pH showed less adsorption. It was also shown that tea ash has enough potential to remove fluoride from aqueous solution and efficiency of removal of fluoride is 64.29%. And fish bone removes fluoride from drinking water and its defluoridation efficiency is 42.86%. Fish bones can be considered to be a comparatively cheaper and effective material for the preparation of fishbone charcoal for defluoridation of drinking water as equally feasible for isolated communities of small sizes as well as at domestic level in the form of home filters. Compared with traditional fluoride removal methods, the locally available materials such as tea ash and fish bone could be used as a cost-effective adsorbent for efficient fluoride removal from water. But, in fish bone defluoridation, the smell is there and it cannot be removed easily. So, in tea ash defluoridation, the color is not changed after the treatment. Since it doesn't make any health problems. As compared to the fish bone defluoridation, the tea ash defluoridation is considered as one efficient technique which can be usually done at domestic levels.

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