

# Removal of Fluoride From Drinking Water Using Tea Waste as Adsorbent

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**Abstract** - Fluoride concentration above 1.5 ppm in drinking water can cause serious health hazards. A low cost and highly efficient adsorbent is prepared by using tea waste and ability to remove fluoride from drinking water is tested. Tea waste is cleaned and chemically treated with sulphuric acid and formaldehyde which is then digested in alum. The fluoride removal studies are done by adsorption method by tea waste. The effect of contact time, pH and adsorbent dosage are considered in batch study. For column study, a column with 50 mm diameter and height 60cm is used. The experiment is conducted for varying bed depth of 3.5 cm, 5.4 cm, 8.9 cm and flow rate of 5ml/min and 10ml/min. Regeneration study is conducted by using sodium hydroxide and desorption efficiency of cycle 1 and cycle 2 is compared. The exhausted adsorbent is regenerated upto two cycles.

**Key Words:** Fluoride, Adsorption, regeneration, column, batch, removal.

## 1. INTRODUCTION

Groundwater is a renewable resource, yet the world's supply of groundwater is steadily decreasing especially in Asia and North America [3]. Groundwater quality deterioration and supply of safe drinking water is a major concern throughout the world. Groundwater with high fluoride concentration (>1.5 mg/L), according to WHO (2011), is affecting more than 260 million people around the world [1].

Groundwater is one of the primary sources of water for domestic and agriculture uses in Kerala. During the last two decades, the groundwater scenario of Kerala has been witnessing several changes [2].

## 2. MATERIALS AND METHODOLOGY

### 2.1 Adsorbent Used

Camellia sinensis is a species of evergreen shrubs from which an aromatic beverage called tea is made. It is originated in borderlands of China and Northern Myanmar. Recently the tea consumption around the world is increasing which results in the production of tea leaf waste from the industrial scale extraction of tea leaves to produce instant tea and bottled tea drinks.

### 2.2 Adsorbent Preparation

In this study waste tea dust from the home is used to carry out the experiment. The waste tea dust is collected and washed properly to remove the milk and sugar. Then boiled twice in order to remove the colour. The tea waste is sun dried to remove moisture. The dried tea waste is sieved to get the particles having size 250µm-500µm. The oversized particles are grounded by mortar and pestle and again sieved.

For chemical treatment take 10 g of tea leaves and add 100 mL of 0.4 N H<sub>2</sub>SO<sub>4</sub> and 20 ml of 30% formaldehyde. This mixture is kept at a constant temperature of 50° C for 3 hours. Then tea leaves washed with distilled water to remove the acid and formaldehyde. Then it is kept in hot air oven to remove moisture. Tea leaves is then digested in 2% alum solution. This is designated as tea leaves chemically treated with sulphuric acid and formaldehyde.

### 2.3 Collection of Sample

The sample was collected from Palakkad district and various parameters such as pH, TDS, Total hardness, Electrical conductivity, Alkalinity and Fluoride are tested.

### 2.4 Preparation of Fluoride Solution

The required sample solution is made up by diluting Fluoride standard solution traceable to SRM (Standard Reference Material) from NIST (National Institute of Standards and Technology) NaF in H<sub>2</sub>O, made in Germany, EMD Millipore Corporation.

### 2.5 SPADNS Spectrophotometric Method

SPADNS spectrophotometric method is a commonly used method for the determination of fluoride in drinking water.

This method involves the reaction of fluoride with a red zirconium - dye solution. The basic principle of spectrophotometric method is that each compound absorbs or transmits light over a certain range of wavelength.

### 2.6 Batch Study

Batch study is conducted in order to study the effects of various parameters like adsorbent size, adsorbent dosage, Initial fluoride concentration, pH and contact time.

And adsorbent dosage 3 g/l, 5 g/l, 7 g/l, 9g/l, 11 g/l, 13g/l, 15g/l, were considered. Initial fluoride concentrations considered were 1 mg/l, 3 mg/l, 5 mg/l and 7 mg/l. Contact time considered were 30, 60, 90, 120, 150 and 180 minutes.

### 2.7 Column Study

Inorder to investigate the practical aspect of application of tea leaves for the removal of fluoride column study is conducted by considering bed depth and flow rate as parameters. The column experiment is carried out in a PVC pipe of 50 mm diameter and 60 cm length and it is attached to a flow control valve for adjusting the flow through column. Bed depth of 3.5 cm, 5.4 cm and 8.9 cm are considered. And flow rate of 5 ml/min and 10 ml/min are considered.

### 2.8 Regeneration of Adsorbent

In the environmental aspect and economic aspect the regeneration of adsorbent is important. Here regeneration study is conducted using Sodium Hydroxide as regenerant. After saturation of adsorbent the regenerant is passed through the adsorbent bed and fluoride removal is checked. The experimental setup used for regeneration was same which was used for the break point analysis. The bed depth of adsorbent used was 5.4 cm at a flow rate of 5 ml/min and initial fluoride concentration of 3 mg/l.

## 3. RESULTS AND DISCUSSION

### 3.1 Testing of Raw Water

The collected sample of water is tested for various parameters. The values had been compared with the drinking water standards IS 10500:2012.

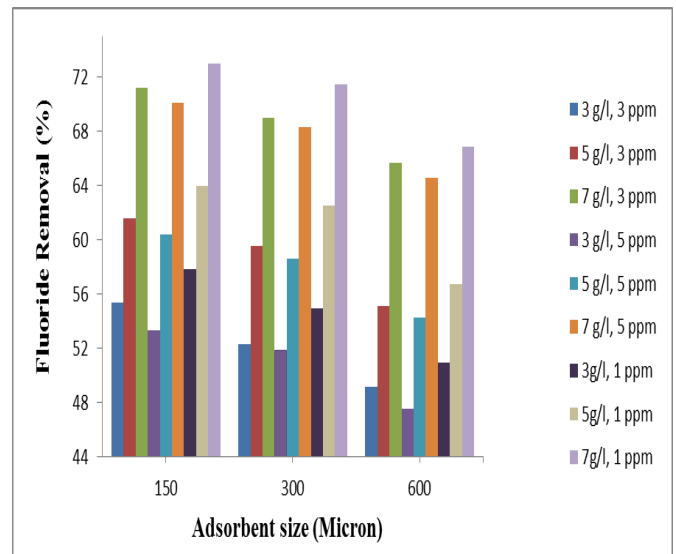
All the parameters except hardness and fluoride is within the desirable limit as per Drinking water standards IS 10500:2012. The value of fluoride in the sample collected from Palakkad district is 2.63 mg/l which is above the desirable limit of 1 mg/l.

### 3.2 BATCH STUDY

In batch study Adsorbent size, Adsorbent dosage, initial fluoride concentration pH and contact time were considered, the detailed results of these parameters are included in following sections.

#### (i) Effect of Adsorbent size

The study of the effect of adsorbent size on fluoride removal efficiency was carried out by using 150 µm, 300 µm, 600 µm sized tea waste.

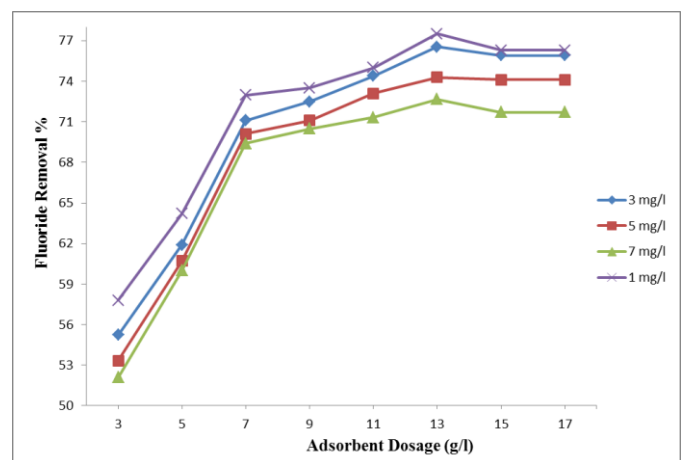


**Chart - 1:** Comparison on the effect of Adsorbent size on the removal efficiency of Fluoride at different initial fluoride concentrations of 1 mg/l, 3 mg/l and 5 mg/l and adsorbent dosages of 3 g/l, 5 g/l and 7 g/l.

A maximum of 73 % removal was obtained for 150 µm sized particles of adsorbent at an adsorbent dosage of 7 g/l and initial fluoride concentration of 1 mg/l and 71.5 % is obtained for 300µm sized adsorbent, but considering the 600 µm sized adsorbent the removal efficiency declines to 66.90 % at 3mg/l initial fluoride concentration.

#### (ii) Effect of Adsorbent Dosage

The study of the effect of adsorbent dosage on the fluoride removal efficiency was carried out with a dosage of 3 g/l, 5 g/l, 7 g/l, 9 g/l, 11 g/l, 13 g/l, 15 g/l. The study is done by keeping adsorbent size as 150 µm and varying initial fluoride concentration as 3 mg/l, 5 mg/l and 7 mg/l.



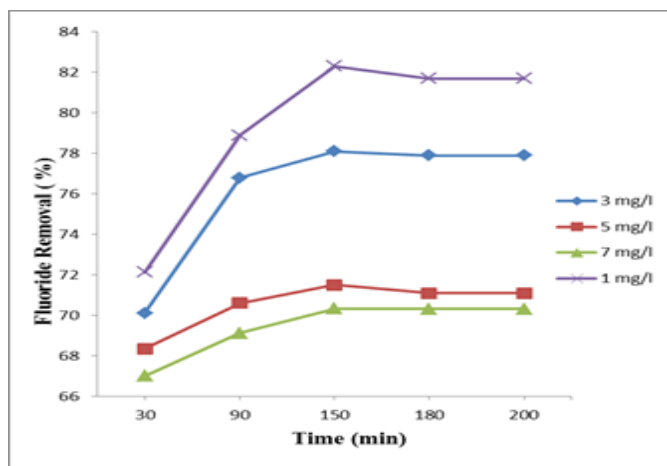
**Chart 2:** - Effect of adsorbent dosage on Fluoride Removal at different initial fluoride concentrations of 1 mg/l, 3 mg/l, 5 mg/l and 7 mg/l.

A maximum efficiency of 76.54 % and 74.3 % was obtained at 13 g/l adsorbent dosage, at initial fluoride concentration 3 mg/l and 5 mg/l. And 72.67 % was obtained at 7 mg/l initial fluoride concentration.

**(iii) Effect of Initial Fluoride Concentration and Contact time**

The study is conducted by varying the concentrations from 1 mg/l, 3 mg/l, 5 mg/l and 7 mg/l, while keeping the adsorbent size and dosage as 150 µm and 13 g/l.

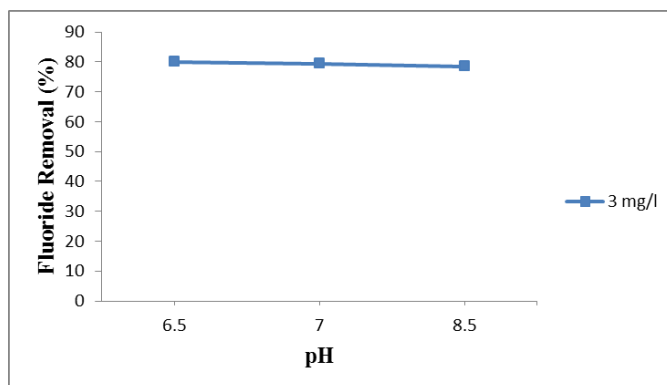
The maximum removal efficiency is obtained at 1 mg/l initial fluoride concentration. We can see that the percentage removal has decreased with the increase in initial concentration of fluoride.



**Chart 3:** - Effect of Initial Fluoride Concentration and contact time on Fluoride Removal.

**(iv) Effect of pH**

The effect of pH on the extent of removal of the fluoride was studied by varying the pH from 6.5, 7 and 8.5 while keeping the adsorbent size and dosage as 150 µm and 13 g/l and initial fluoride concentration 3 mg/l.



**Chart 4:** - Effect of pH

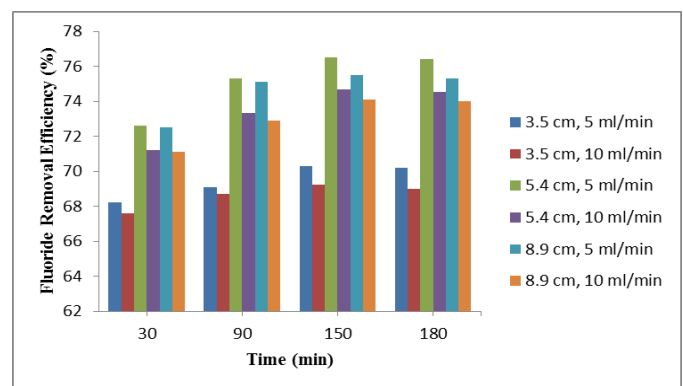
The maximum removal efficiency is obtained at a pH of 6.5 and there is no great variation in removal efficiency within pH 6.5 to 8.5.

**3.3 COLUMN STUDY**

In column study we are mainly considering parameters such as bed depth of adsorbent and flow rate.

**(i) Effect of Bed Depth and Flow Rate**

In this experiment the depth varies from 3.5 cm, 5.4 cm and 8.9 cm and the different flow rate chosen are 5 ml/min and 10 ml/min at time interval of 30, 90, 150 and 180 minutes at pH 6.

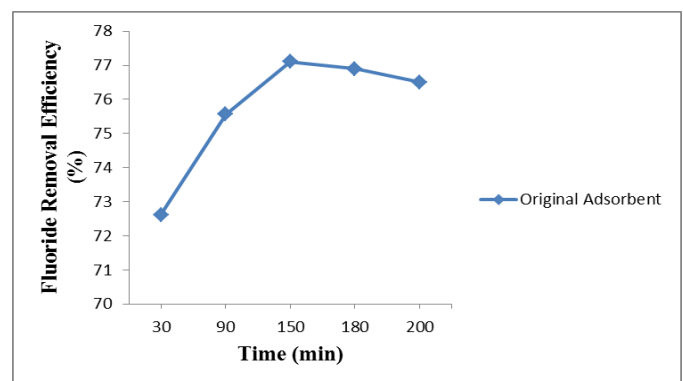


**Chart 5:** - Effect of Bed Depth and Flow rate in fluoride removal.

The maximum removal efficiency of 76.5 % is obtained at a adsorbent bed depth of 5.4 cm and at a flow rate of 5 ml/min after 150 minutes. But after 180 minutes there is a decrease in the fluoride removal efficiency. Removal efficiency decreased from 76.5 % to 76.41 %.

**(ii) Break through Study**

In order to find the breakthrough time of column we conducted the study till the saturation of the column in the first cycle.



**Chart 6:** - Breakthrough Curve of Original Adsorbent

A fluoride removal efficiency of 72.62 % obtained at 30 minutes and gradually increases to 75.56 % at 90 minutes and 77.1 % removal at 150 minutes.

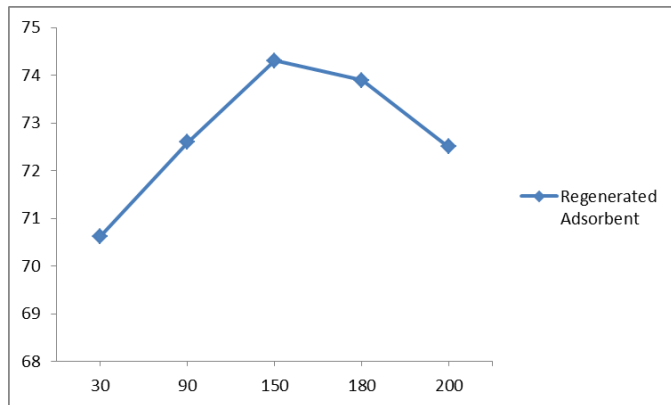


Chart 7: - Breakthrough Curve of Regenerated Adsorbent

### 3.4 REGENERATION STUDY

We need to check the possibility of regeneration before throwing the adsorbent after it reaches the saturation point. Sodium Hydroxide solution was used as regenerant.

After saturation of adsorbent column in cycle 1 is washed with NaOH solution. Initially a fluoride removal of 66.4 % occurs at 10 minutes, then gradually increases to 67.1 % in 30 minutes, 68.6 % in 50 minutes, 69.73 % in 70 minutes, 71.33 % in 90 minutes, 73.1 % in 110 minutes, 74.9 % in 130 minutes, then reduced to 74.6 % in 150 minutes and 72.8 % in 170 minutes.

In cycle 2 initially fluoride removal efficiency was 60.33 % in 10 minutes, then gradually increases to 62.51 % in 30 minutes, 63.7 % in 50 minutes, 64.11 % in 70 minutes, 65.4 % in 90 minutes, 67.5 % in 110 minutes, 68.13 % in 130 minutes, then reduced to 67.12 % in 150 minutes, 65.3 % in 170 minutes.

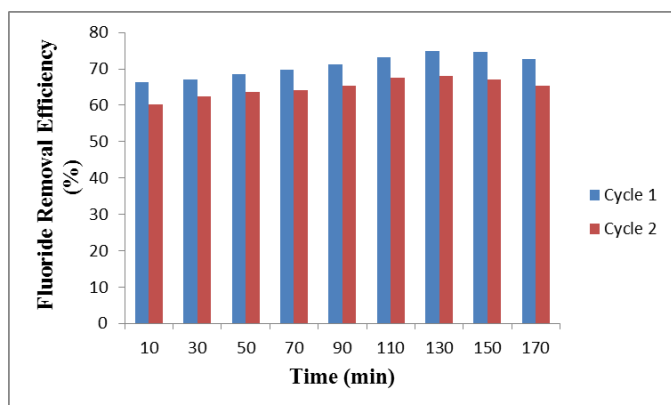


Chart -8: Comparison of regeneration of Original adsorbent and regenerated adsorbent

This study shows that the regeneration of adsorbent is possible and a maximum fluoride removal of 74.9 % and 68.13 % is obtained in cycle 1 and cycle 2.

### 4. CONCLUSIONS

The adsorbent size used in this study are 150 μm, 300 μm and 600 μm. The Adsorbent dosage are 3 g/l, 5 g/l, 7 g/l, 9 g/l, 11 g/l, 13 g/l and 15 g/l. The initial fluoride concentration of 1mg/l, 3 mg/l, 5 mg/l, 7 mg/l. The time varying from 30 minutes to 180 minutes as contact time. The effect of pH on fluoride removal efficiency was conducted by varying pH from 6 to 8. Max removal efficiency was obtained for 150 μm adsorbent size and 13 g/l adsorbent dosage with 3 mg/l initial fluoride concentration at 150 minutes at pH 6.

In the column study the parameters mainly considered were flow rate and Bed depth. Flow rate chosen for the column study were 5 ml/min and 10 ml/min. The bed depth chosen were 3.5 cm, 5.4 cm and 8.9 cm. As the first cycle completed a maximum fluoride removal efficiency of 77.1 % was obtained at a bed depth of 5.4 cm and flow rate of 5 ml/min in breakthrough analysis. And as the second cycle has completed a maximum removal efficiency of 74.31 % was also obtained at a bed depth of 5.4 cm and flow rate of 5 ml/min.

In regeneration study of cycle 1 the fluoride removal of 66.4 % was obtained at 10 minutes the gradually increased to 74.9 % at 130 minutes.

In cycle 2 the fluoride removal of 60.33 % was obtained at 10 minutes then gradually increased to 68.13 % at 130 minutes. After 130 minutes the removal efficiency reduced to 67.12 % at 150 minutes and then declined to 65.3 % at 170 minutes.

From this sets of experiment, we can conclude that tea waste can be used as an adsorbent for removing fluoride from water, as it shows great removal efficiency and regeneration characteristics.

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