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FEASIBILITY STUDIES ON DEFLUORIDATION OF WATER USING NATURAL ADSORBENTS

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Abstract - *Fluorides are the major pollutants present in the effluents from various industries and ground water sources. These are highly toxic to living beings and have a hazardous effect on their health. Thus the removal of fluoride using bio sorbents is a major step towards the protection of environment. Adsorption is the most effective and widely used method and is applicable for the removal of fluoride even at low concentrations This paper presents the results of investigations carried out for removal of Fluoride from water by using natural adsorbents i.e. Amla bark powder, Neem bark powder, Ground granulated blast furnace slag, Waste lime, Sugarcane baggase. The fluoride removal efficiency of adsorbents was investigated by batch wise adsorption experiment. The effect of various important parameters on the % removal was studied to find the optimum condition for the maximum removal of fluorides. The parameters like contact time, adsorbent dose, height of column and pH were investigated. The optimum pH, height of column, adsorbent dose and contact time were found to be 5, 15ml, 20g/l and 50 min. respectively for which there was maximum fluoride removal. All the results were validated on the basis of statistical analysis. All the graphs were fitted to various trend lines. Out of which the equation of most fitted trend line was adopted for the validation and deviation were observed.*

Keywords - *Adsorption, biosorbents, statistical analysis, fluoride, GGBFS.*

I. INTRODUCTION

Fluoride is an inorganic, monatomic anion of fluorine with the chemical formula F^- which is of halogen group. Fluoride is the simplest anion of fluorine. Its salts and minerals are important chemical reagents and industrial chemicals, mainly used in the production of hydrogen fluoride for fluorocarbons. In terms of charge and size, the fluoride ion resembles the hydroxide ion. Fluoride ions occur on earth in several minerals, particularly fluorite, but are only present in trace quantities in water. Fluoride contributes a distinctive bitter taste. It contributes no colour to fluoride salts.

Fluoride can act as a base. It can combine with a proton (H^+):

$$F^- + H^+ \rightarrow HF$$

This neutralization reaction forms hydrogen fluoride (HF), the conjugate acid of fluoride.

In aqueous solution, fluoride has a pK_b value of 10.8. It is therefore a weak base, and tends to remain as the fluoride ion rather than generating a substantial amount of hydrogen fluoride. That is, the following equilibrium favours the left-hand side in water:

$$F^- + H_2O \qquad HF + HO^-$$

However, upon prolonged contact with moisture, soluble fluoride salts will decompose to their respective hydroxides or oxides, as the hydrogen fluoride escapes. Fluoride is distinct in this regard among the halides. The identity of the solvent can have a dramatic effect on the equilibrium shifting it to the right-hand side, greatly increasing the rate of decomposition.

1.1 Occurrence

Fluoride is found in all natural waters at some concentration. Seawater typically contains about 1mg/l while rivers and lakes generally exhibit concentrations of less than 0.5 mg /l. In ground waters, however, low or high concentrations of Fluoride can occur, depending on the nature of the rocks and the occurrence of fluoride-bearing minerals. High fluoride Concentrations may therefore be expected in ground waters from calcium-poor aquifers and in areas where fluoride bearing minerals are common.

1.2 Effect of fluoride

1.2.1. Dental fluorosis- If fluoride present in this range of 0.7 to 1.5 mg F/l in drinking water may cause dental fluorosis, in dental fluorosis loss of lustre and shine of the dental enamel. The discoloration starts from white yellow, brown to black enamel matrix is laid down on incremental lines before and after birth. Fluorosis is seen as mild moderate and severs depending on the amount of fluoride ingested during the stages of formation of the teeth.

1.2.2 Skeletal fluorosis- Excessive quantity of fluoride deposited in the skeleton, Fluoride poisoning leads to severe pain associated with rigidity and restricted movements of cervical and lumbar spine, knee and pelvic joints as well as shoulder joints. Symptoms of skeletal fluorosis are Pain in neck, back bone or joints, stiffness in the neck, backbone or joints sever pain and rigidity in the hip region (pelvic girdle) construction of vertebral canal and intervertebral forearm exerts pressure on nerves and blood vessels leading to paralysis and pain. Skeletal fluorosis is an irreversible process as the dental fluorosis.

1.2.3. Non –skeletal fluorosis- Fluoride when consumed in excess can cause several other kind of manifestation. The WHO guideline value for fluoride in drinking water is 1.5 mg/l. Above 1.5 mg/l mottling of teeth may occur to an objectionable degree. Concentrations between 3 and 6 mg/l may cause skeletal fluorosis. Continued consumption of water with fluoride levels in excess of 10 mg/l can result in crippling fluorosis. In many arid regions, drinking water is such a scarce commodity that governments have been forced to set the standard at higher levels, in order to have any drinking water at all.

II. LITERATURE REVIEW

Several adsorbent materials have been tried in the past to find out an efficient and economical defluoridating agent. Some of those adsorbents are Amla bark powder, Neem bark powder and Ground granulated blast furnace slag etc. On the basis of past research in the field of fluoride removal, a wide range of bio adsorbents were experimented. An overview of some of the research works were represented in table 1. Neem bark shows greater removal efficiency about 90% at a controlled pH 6. The effect of pH, adsorbent dosage, contact time, height of column etc. .will affect the fluoride removal efficiency.

Table1. Comparative Data of Different Adsorbents

Adsorbent	Initial Conc.(mg/L)	pH	% Removal
GGBFS	3	6	75
Amla bark	3	6	84
Neem bark	3	6	90

III. MATERIALS AND METHODS

3.1 Preparation of adsorbents

Ground granulated blast furnace slag is collected from the steel industries located in MIDC area. Amla bark is collected from the trees of Amla from farm. Neem bark powder is directly purchased from the shop. Both the materials were bringing directly into its original form without any treatment. Further washing of the adsorbents was done to remove colour as well as lignin. Alternate 4-5 cycles of acid and alkali wash was done on adsorbents. Two separate solutions were prepared approximately 1 litre each. One solution contains 10 ml hydrochloric acid in 1 lit water and other contains 10 gm sodium hydroxide (NaoH) in 1 lit. After soaking up the adsorbent alternately in both the solution the adsorbent get filtered by piece of cloth. After complete washing the adsorbents were allowed dry for 24 hrs at room temperature. After drying the adsorbents were crushed into fine powder.

3.2 Preparation of fluoride standards

Prepare a series of standards by diluting with distilled water 5, 10 and 20 mL of standard fluoride solution to 100 mL with distilled water. These standards are equivalent to 0.5, 1.0 and 2.0 mg F/L.

Fig.1. Reference and fluoride standard solutions

3.3 Preparation of standard curve

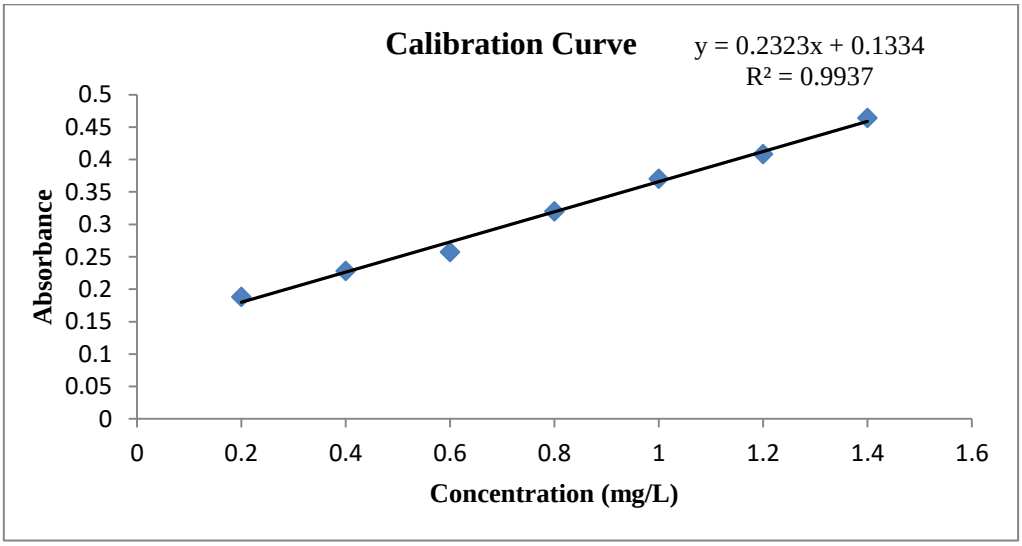

Fig.2. Calibration Curve for fluoride

Prepare a series of fluoride standards in the range of 0 to 1.40 mg F/L by diluting appropriate quantities of standard fluoride solution to 50 ml with distilled water. Pipet 5 mL each of SPADNS solution and Zirconyl-acid reagent or 10 mL mixed acid zirconyl SPADNS reagent, to each standard and mix well. Set photometer to zero absorbance with reference solution and obtain absorbance reading of standards. Plot a curve of milligrams fluoride absorbance relationship. As an alternative to using a reference, set photometer at some convenient point (0.3 or 0.5 absorbance) with prepared 0 mg F/L standard.

3.4 Methodology

In present study the concentrations of aqueous solution of fluoride was determined by spectrophometric method by using SPADNS.

Table 2. Data for calibration curve

ml. standard solution	blank	0.5	1	1.5	2.5	5	6.25
ppm F	0	0.1	0.2	0.3	0.5	1	1.25

3.4.1 Batch Adsorption Experiment- In batch adsorption experiment , the 25 ml of sample having 3 mg/L of initial fluoride concentration had been taken in a small beaker of volume 100 ml containing 20 gm/ml was thoroughly mixed on a magnetic stirrer. After the agitation the beaker was allowed to stand for 2 minutes for settling of adsorbent. Further it is filtered by using filter paper. By taking 10 ml of filtered sample and by adding 2 ml of acid- zirconyl SPADNS reagent the absorbance is checked by using spectrophotometer. Similar experiment was carried out for determining the optimum condition and to study the effect of pH, adsorbent dose, contact time and height of column. The materials was optimised for the pH of 4,6,8 etc. The adsorbent dose was varied as 100, 200, 300, 400 and 500gm in 25 mL solution. Contact time was taken at equal intervals of 15 minutes i.e absorbance was checked after 30, 45, 60, 75, 90 and 120 minutes. Height of column varies between 10 to 30 ml at regular intervals of 5 mL in a burette having a volume of 50 mL. From this batch study the optimized parameters are obtained at which the removal percentage of fluoride was found to be maximum.

IV. Results and Discussion

The present study provides the comparison of fluoride removal efficiency of bio-adsorbents such as Amla bark powder, Neem bark powder and GGBFS from a solution contaminated with 3 mg/l fluoride under variable adsorbent dosages. The fluoride concentration and removal efficiencies for different bio adsorbents at different adsorbent dosage are shown in fig.

4.1 Effect of pH

The pH of the aqueous solution is an important controlling parameter in the adsorption process. The effect of the pH on the removal of fluoride was studied by varying the pH from ph 4 to 9. The results are presented, where it can be seen that maximum removal of GGBFS about 74% is found at a dose of 20 g/L at pH of 6 and thereafter the percent removal became more or less constant. Maximum removal of Neem bark powder about 90% is found at a dose of 20 g/L at pH of 6.Maximum removal of Amla bark powder about 84% is found at a dose of 20 g/L at pH of 6.

4.2 Effect of Adsorbent Dosage

To study the effect of biosorbent dose, the experiments are performed at various doses of biosorbent lying between 4-20 g/L of fluoride solution with 3 mg/L initial concentration of Fluoride. A pH of 6 was adjusted for GGBFS, Amla bark as well as for Neem bark powder. Observations were made at adsorbent dose of 4, 8,12,16,20 g/L and these experiments were conducted at room temperature for all the adsorbents. The percentage removal of fluoride increases with increase of adsorbent dosage for all the adsorbents, but after sometimes it gradually approaches a state of equilibrium. The percentage removal of fluoride was found to be maximum for all the adsorbents at a maximum dose of 20g/L.

4.3 Effect of Contact time

To study the effect of contact time, the experiments were performed at various contact time lying between 30 to 120 min. Observations were made at an regular interval of 15 min and last was of 30 min i.e. 30,45,60,75,90 and 120 min. Amla bark removes the maximum fluoride in initial 30 minutes further there are variations in the % removal. Neem bark also removes the maximum fluoride in initial 30 min. Up to 60 min's the removal goes on decreasing and again starts increasing up to 120 min's. GGBFS removes the maximum fluoride in 45 min and thereafter goes on decreasing.

4.4 Effect of Height of column.

To study the effect of height of column various heights were optimised and % removal of fluoride was obtained. Materials were feed into the burrates having an volume of 50ml. Observations were made at various column heights of 10,15,20,25,and 30 min. Amla bark removes maximum fluoride at a height of 10mL. Neem bark removes maximum fluoride removal at a height of 15mL. Further rate of removal goes on decreasing. GGBFS removes Maximum fluoride at a height of 20mL. Further rate of removal goes on decreasing. GGBFS removes Maximum fluoride at a height of 20mL.

4.5 Statistical Analysis

For the validation of laboratory results, attempts are made to opt statistical modelling from fig.1 to fig. 6 it is observed that polynomial second order curve are best fit to our laboratory works shows the coefficient of correlation (R^2 value) in the tune of 0.88 to 0.96.

4.5.1 Optimization of pH

Table 3. Name of Adsorbent: Amla Bark

Sr. No.	pH	% Removal	% Removal Y=5.441x2 + 66.921x - 116.25
1	4	66	64
2	6	84	89
3	8	79	70
4	9	41	45

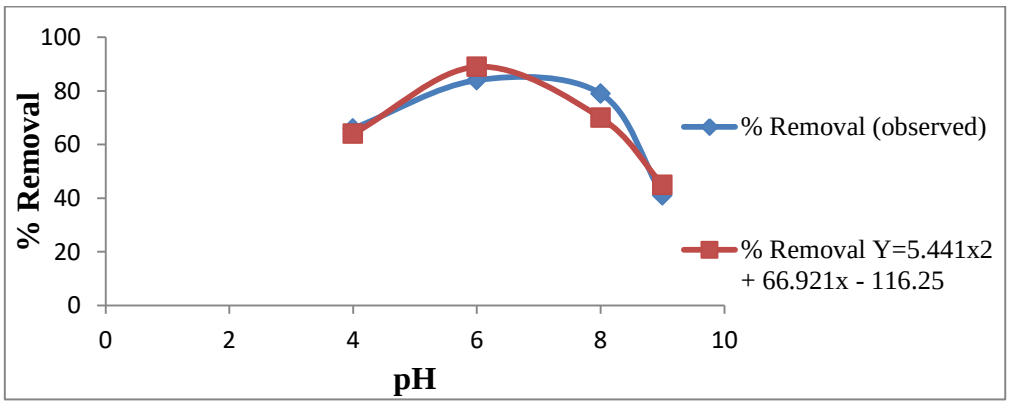

Fig.3 Effect of pH on adsorption of fluoride on Amla bark

Table 4.Name of Adsorbent: Neem Bark for optimised pH 6

Sr. No.	pH	% Removal	% Removal Y = -5.4837x2 + 67.17x - 114.32
1	4	67	67
2	6	90	91
3	8	74	72
4	9	45	46

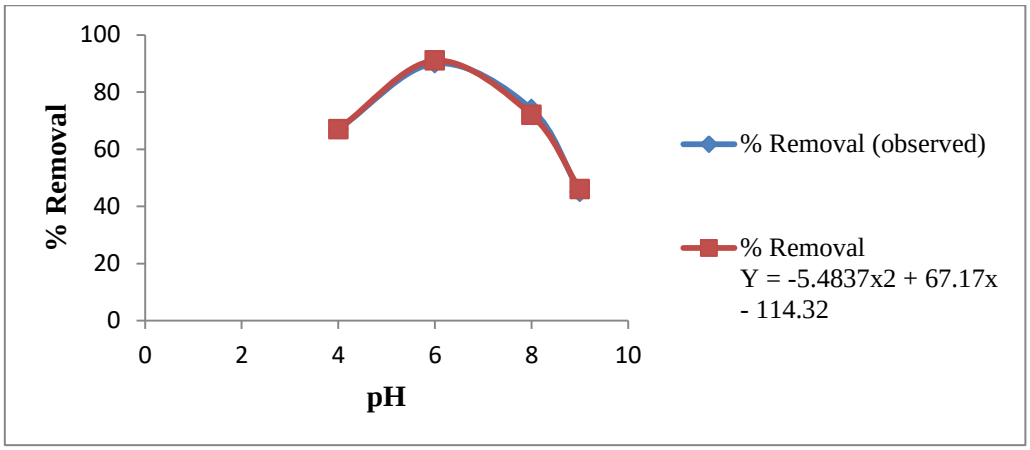

Fig.4 Effect of pH on adsorption of fluoride on Neem bark

Table 5. Name of Adsorbent: GGBFS for optimised pH 6

Sr. No.	pH	% Removal	% Removal Y = -4.0641x2 + 47.373x - 65.113
1	4	59	60
2	6	74	73
3	8	52	54
4	9	33	32

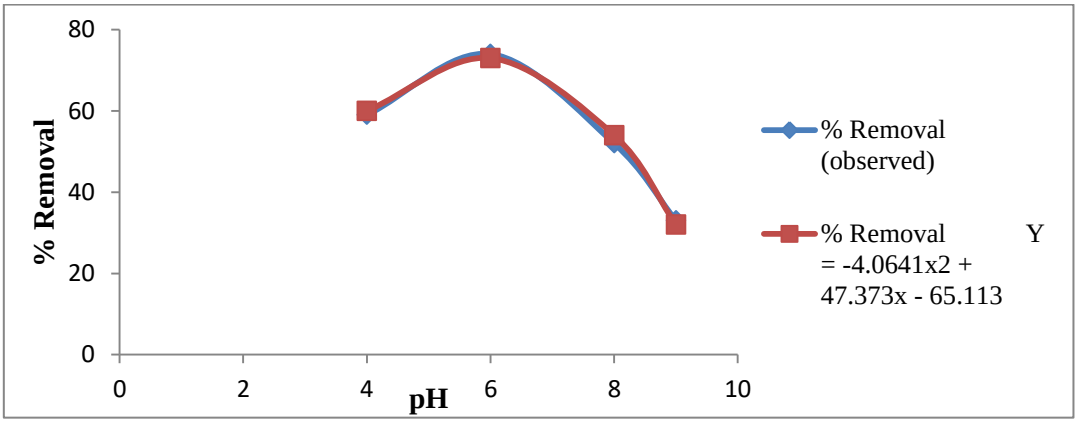

Fig.5 Effect of pH on adsorption of fluoride on GGBFS

4.5.2. Optimization of Contact Time

Table 6. Name of Adsorbent: Awla Bark for optimised pH 6

Sr. No.	Contact Time (min)	% Removal	% Removal y = 0.0057x2 - 0.8143x + 97.5
1	30	84	78
2	45	62	72
3	60	72	69
4	75	67	68
5	90	76	70
6	120	80	82

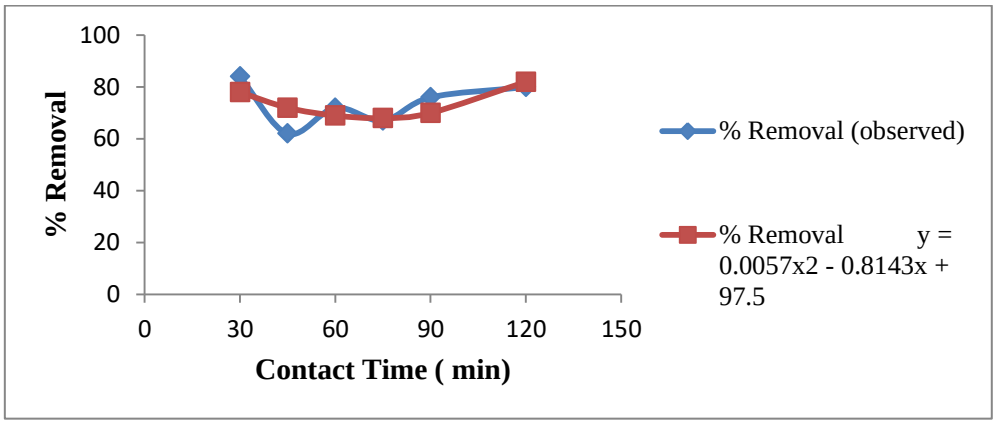

Fig.6 Effect of contact time on adsorption of fluoride on Awla bark

Table 7.Name of Adsorbent: Neem Bark for optimised pH 6

Sr. No.	Contact Time (min)	% Removal	% Removal y = -0.0001x3 + 0.0415x2 - 4.4466x + 192.61
1	30	90	93
2	45	80	67
3	60	40	54
4	75	52	50
5	90	57	56
6	120	80	83

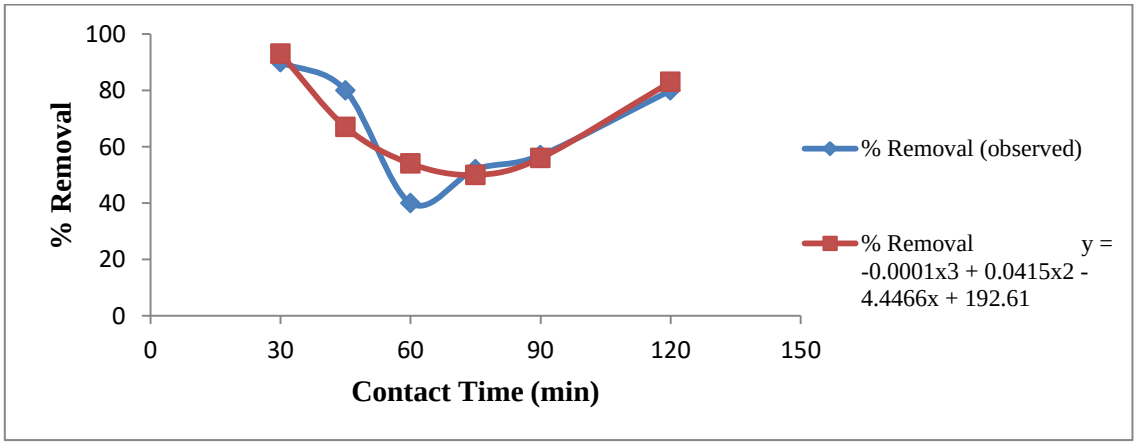

Fig.7 Effect of contact time on adsorption of fluoride on Neem bark

Table 8. Name of Adsorbent: GGBFS for optimised pH 6

Sr. No.	Contact Time (min)	% Removal	% Removal y = 0.0057x2 - 0.8143x + 97.5
1	30	44	50
2	45	60	48
3	60	43	45
4	75	43	42
5	90	33	38
6	120	32	29

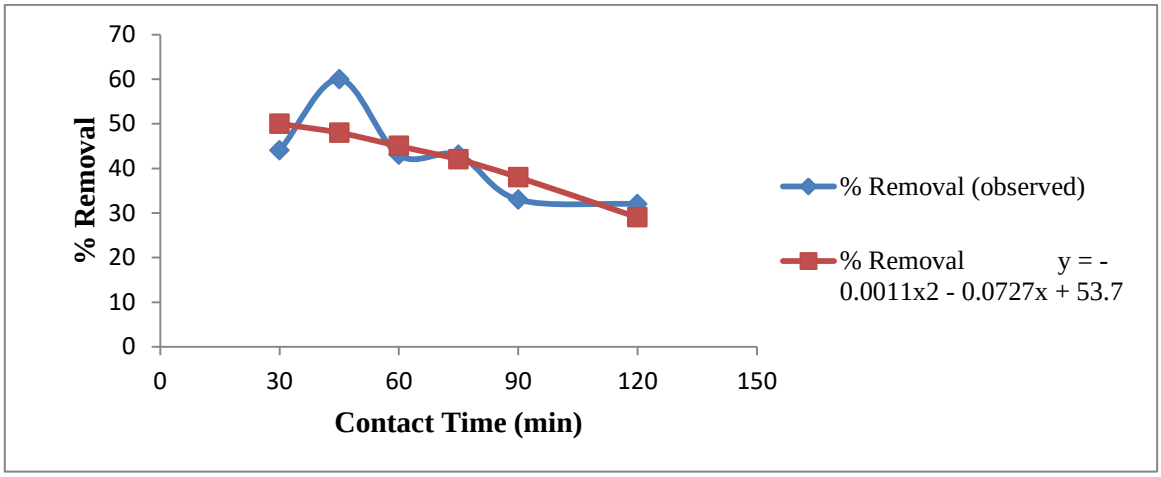

Fig.8. Effect of contact time on adsorption of fluoride on GGBFS

V. CONCLUSION

Based on the results of this study, it can be concluded that sugarcane bagasse have good performance to adsorb fluoride from drinking water. Neem bark gives the maximum removal of 90% of fluoride from water at a maximum dose of 20g/L. .Awla bark gives the removal of 84% of fluoride from water at a maximum dose of 20g/L. GGBFS gives the removal of 74% of fluoride from water at a maximum dose of 20g/L. It was also found that the removal efficiency of adsorbents is affected by the pH, contact time, height of column and adsorbent dose. Also it is concluded that it is necessarily to aware about ground water contamination and use cheaply method specially adsorbent process use to remove fluoride in which several cheap adsorb and use like red mud, pine apple peel powder, orange peel powder, chalk powder, ragi seed powder, and sugar cane bagasse.

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