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Removal Studies of Fluoride Ions by Column Chromatography Using Scolecite

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Abstract: Fluoride is known to play double role in human beings. At one end, its deficiency in ingestion may result in dental diseases and skeletal weakness, whereas on the other side excess of it causes serious health hazards. The sufferers of fluorosis (caused by excessive fluoride) are estimated to be about 80 million all over the world. Surface water, which is main source of drinking water in rural area is more likely to be contaminated with fluoride, due to its natural occurrence in the form of inorganic and organic fluorides and as a consequence of the anthropogenic activities. Researchers are in constant search of suitable materials to be used for removal of fluoride from drinking water. Zeolites are surface active agents and also act as molecular sieves. These features of zeolites can be utilized for water defluoridation. This study is an effort made in this direction, by using the remarkable adsorption tendencies of natural zeolite, scolecite. Adsorption capacities of scolecite for fluoride ions have been studied using column chromatography in a packed column of adsorbent at different time of flow, sample concentrations and flow rates at neutral pH. The results indicate the success of removal of fluoride ions up-to a certain extent and there is a scope for further development of the process.

Keywords: Fluoride, Defluoridation, Zeolite, Drinking water, Scolecite.

I. INTRODUCTION TO PROBLEM

Fluoride ions (F) are considered to be essential to avoid the cavities formation in the teeth and providing strength to human skeletal system.

This ion have natural occurrence in a variety of dietary items and also in drinking water distribution sources in many areas. It is also added in drinking water distribution sources in some regions to overcome the abovementioned health issues. The appropriate range of concentration of fluoride in drinking water is considered to be between 0.7ppm-1.2ppm.

However excessive consumption of fluoride by ingestion of drinking water causes serious health problems in humans worldwide. As per the guidelines of WHO 2011 the permissible limit of concentration of fluoride for human beings is 1.5 ppm. According to UNICEF official position (Water front 1999), on water fluoridation, fluorosis is endemic in around 25 countries of the world¹. It is difficult to know the exact number of affected people, but a rough estimation predicts the number in the tens of millions. In India 15 out of 32 states were identified in 1993 as greatly affected by fluoride.

About 5 million people (approximately 6% of the population) are affected by fluorosis in Mexico. Central and Western regions of China are also found to be affected regions. Worldwide instances of fluorosis due to drinking water are on the rise.

As per the findings by researchers (Busulu et al 1980, Meier et al 1992)^{2,3} a low concentration of calcium and high alkalinity due to bicarbonates is favorable for high concentration of fluoride in ground water. At present, the enhanced consumption of fluoride in industries and dental health products, resulted in the raise in fluoride concentration in the environment and as a consequence in drinking water and other sources.

As major health hazards dental and skeletal fluorosis are reported world-wide, on the other hand, excessive fluoride may be the cause of other health problems like degeneration of muscle fiber, abnormalities of RBC's, neural diseases, gastric problems, low levels of hemoglobin, nausea, disorders of urinary tract, sensation problems in fingers, sterility in males etc. Despite this serious raise in consequences of excessive concentration of fluorides in water, a lack of full awareness is observed.

Great efforts are therefore required to support more research for the eradication of this problem. Excessive fluoride in drinking water may cause teeth molting or dental fluorosis and causes enamel layer discoloration, which may form spots and horizontal streaks on teeth and more severely teeth chipping in children (Choubisa et.al-1996)⁴. Bone fluorosis is reported while concentration of fluoride exceeds 1.5 mg/lit.



Photographs showing Dental fluorosis and skeletal fluorosis



Photograph showing children affected from skeletal fluorosis M.P. (INDIA).

Great efforts are therefore required from all angles for the effective eradication of the health hazards caused due to excessive fluoride. Either use of alternative water sources or removal of excess of fluoride ions from drinking water are required to minimize the fluoride poisoning. The process of removing fluoride from water is called water defluoridation. Flocculation and adsorption are two ways followed in general practice for the treatment of water supplies. Extensive research has been done for the development of effective methods for the removal of fluoride from drinking water. Among several methods used for the defluoridation of drinking water, adsorption processes are generally found effective due of their convenience, ease of operation, simplicity of design, and for economic and environmental reasons. The principles used in these methods are adsorption, ion exchange, precipitation, coagulation, membrane separation, electrolytic defluoridation, electrodialysis etc. (Raichur et al.-2001), (Singh et.al-1999), (Saha et.al-1993, Reardon-2000) (Amer et.al-2001, Dieye et.al-1998) (Mameri et.al-2001), (Hichour et.al-1999, Hichour et.al-2000)⁵⁻¹³.

Adsorbent materials tried earlier in the search for an efficient and economical defluoridating agent are activated alumina, activated carbon, activated alumina coated silica gel, calcite, activated saw dust, activated coconut shell carbon and activated fly ash ground nut shell, coffee husk, magnesite, serpentine, tricalcium phosphate, bone charcoal, activated soil sorbent, carbon, defluoron-1, defluoron-2 etc. are different adsorbents materials reported in the literature (Muthukumaran et.al-1995, Min et.al 1999, Wang et.al-2001, Nava et.al-2003, Padmavathy-2003)¹⁴⁻¹⁸. The most common among these adsorbents are activated alumina and activated carbon.

Techniques using activated alumina for defluoridation are being propagated in many villages by the voluntary organizations funded by UNICEF or other agencies. The adsorption efficiency for removing fluoride of activated alumina is dependent on hardness and surface loading (the ratio of total fluoride concentration to activated alumina dosage). The process is pH specific and it is necessary to maintain the pH of the solution in the range 5.0-6.0 because at pH > 7.0 silicates and hydroxides comes out as stronger competitor of fluoride ions for exchange sites on activated alumina and at pH < 5.0, activated alumina gets dissolved in acidic environment

leading to loss of adsorbing media (Thergaonkar et.al-1987, Bishop et.al-1978)¹⁹⁻²⁰. It is the highly selective process and has very low adsorption capability, pretreatment and acidification is required. The effectiveness of this material for removal of fluoride becomes less after each regeneration.

Powdered activated carbon has been reported for fluoride removal to achieve good results. The process is pH dependent and yields better results at pH 3.0 or less. For this reason pH adjustment is required and the process becomes expensive (McKee and Johnston 1934)²¹.

The literature survey has indicated that by the use of each of the above- mentioned techniques removal of fluoride can be done only under specified conditions and the suitability of a particular process will differ from place to place. This study and the remarkable tendency of zeolites of adsorption and ion exchange inspired us to undertake the present investigation to find out some utilities of zeolites for chromatographic work and how these properties can be used for removal of fluoride from drinking water. It is worth mentioning here that scolecite (a zeolite) has been used as a sorbent for the first time in the history of defluoridation.

II. EXPERIMENTAL

A. Materials

The scolecite sample used for chromatographic studies was obtained from Ahemadnagar near Pune, Maharashtra India. Scolecite is a mineral of zeolite family, white in color. It is a framework silicate with tetrahedron framework. It can be represented as $\text{CaAl}_2\text{Si}_3\text{O}_{10} \cdot 3\text{H}_2\text{O}$ (hydrated calcium silicate), little amount of sodium and potassium in traces substitutes calcium .



Photograph showing natural scolecite cluster.

B. Principle

This technique is based on differential adsorption of the components of a mixture (sample) in mobile phase, on suitable adsorbent packed in a column (stationary phase). The component with greater affinity to the stationary phase adsorbs early and hence moves slow and gets eluted out later ,while components with lower adsorption moves fast and gets eluted out before other components. The retention factor of the individual components is determined by calculating the ratio of the distances travelled by the individual components to that by mobile phase.

C. Column

In column chromatography, most commonly a vertically placed cylindrical glass column fitted with a porous disc at the lower end, to hold the adsorbent packed in the column and allow only mobile phase with dissolved sample to pass through it, is used. The bottom end is shaped in the form of a tube as outlet with a controlling knob.

D. Mobile Phase

Suitable solvent or mixture of solvents is used as a medium to carry with them the sample mixture by properly dissolving them.

E. Stationary Phase

It is generally a solid or in some cases a liquid supported by sold. In column chromatography usually an adsorbent in uniform and fine particle size is used as column packing material.

III. METHOD

Fluoride removal was studied using packed columns of crushed and sieved scolecite. Adsorbent material(Scolecite) was suitably moistened with solvent distilled water and packed in the column over a sintered disc at the bottom. The (NaF) sample of different concentrations, ranging from 2- 10 ppm, prepared in distilled water is poured from the top of packed column through funnel with flow control. A suction pump was connected to provide a suction with a constant rate and the elute is collected in a beaker. The solution of NaCl was used for regeneration of scolecite. The natural zeolite powder was put for reactivation in hot air oven for about one hour at 110°C after distilled water washing and air stripping.

A. Determination Of Fluoride Concentration

- 1) *Apparatus:* Quantitative analysis of fluoride was performed with HI 98402 Fluoride Ion and temperature meter (HANNA Instruments USA) , HI 4010 Fluoride ion selective electrode and HI 5313 Ag/ AgCl reference electrode (HANNA Instruments USA).
- 2) *Material:* Scolecite is used as adsorbent for present investigation because of its unique characteristics mentioned earlier as well as it is easily available in bulk from natural sources decreasing its cost as compare to other available adsorbents.
- 3) *Procedure:* A stock solution of fluoride ions with 100 ppm concentration was prepared. In a 1000ml volumetric flask 221mg of anhydrous NaF was dissolved in distilled water and volume made up to the mark.

The adsorbent was crushed and after washing by distilled water dried at 105°C for 12 h in hot air oven. Then it was passed through sieve in order to get the desired fine particle size of adsorbent .

Impregnation and activation process was not done prior to the use of the adsorbent scolecite. For measuring concentration of fluoride ions a total ionic strength adjustment buffer solution (TISAB) was used.

IV. RESULTS AND DISCUSSION

All solid materials have certain adsorption capacities. Naturally occurring zeolite scolecite have Ca(II) cation which can catch fluoride anions from water. Another cause of capture of fluoride might be the presence of exchangeable anion OH⁻ because of similar charge and size of these anions. The fluoride removal tendency of scolecite was investigated using column chromatography at different flow rates and time of run of sample solutions of concentrations ranging from 2-10 ppm.

Table: 2.1 Fluoride concentration in ppm in effluent
Adsorbent material: 30 gms., Temperature: 25°C, , Flow rate 2.0 ml/min.

Sr. No.	Time of run in Minutes	Fluoride concentration in ppm in effluent For		
		Sample concentration: 2 ppm	Sample concentration: 5ppm	Sample concentration:10 ppm
1.	2.0	1.6	4.4	9.1
2	5.0	1.2	3.7	7.8
3	10.0	1.0	3.1	6.4
4	15.0	0.53	2.4	5.9
5	20.0	0.41	1.8	3.8
6	25.0	0.23	1.5	3.2
7	30.0	0.22	1.5	2.8
8	35.0	0.22	1.4	2.6

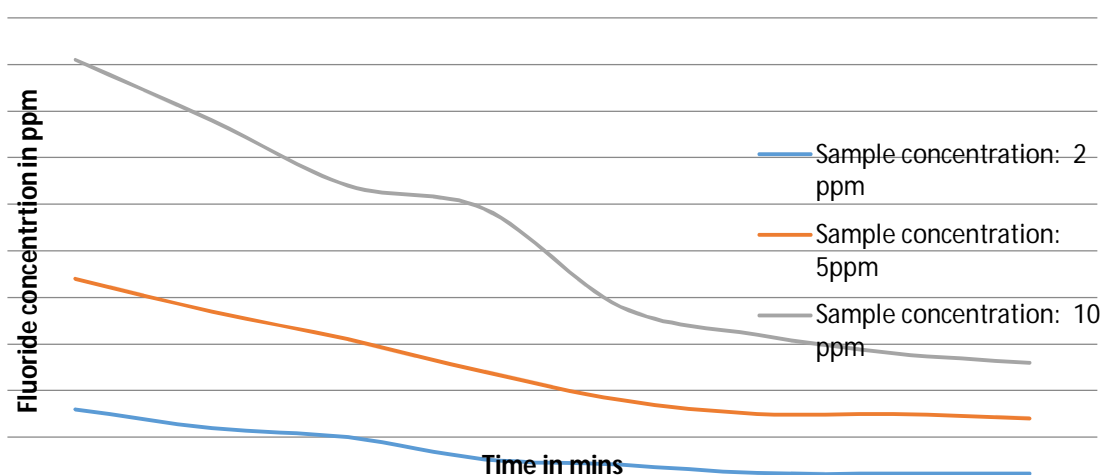


Figure: Graph showing removal of Fluoride as a function of Time of run.

Table: Fluoride concentration in ppm in effluent

Adsorbent material: 30 grms., Temperature: 25°C, Flow rate 1.0 ml/min.

Sr. No.	Time of run in Minutes	Fluoride concentration in ppm. For		
		Sample concentration: 2 ppm	Sample concentration: 5ppm	Sample concentration: 10 ppm
1.	2.0	1.4	4.2	8.8
2	5.0	0.9	3.5	7.2
3	10.0	0.62	2.9	5.9
4	15.0	0.41	1.8	4.4
5	20.0	0.28	1.8	3.5
6	25.0	0.20	1.5	3.2
7	30.0	0.18	1.3	2.8
8	35.0	0.18	1.2	2.8

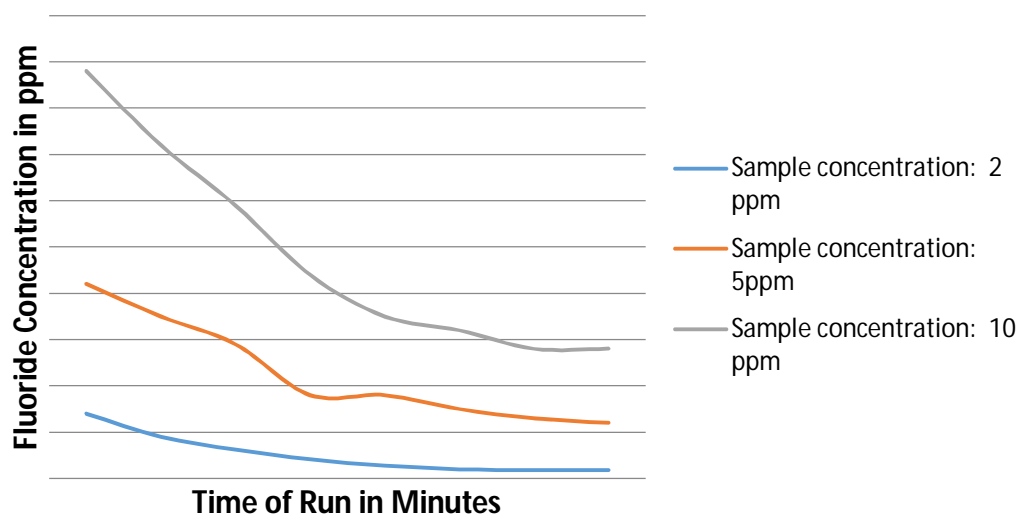


Figure: Graph showing removal of Fluoride as a function of Time of run.

Table: Fluoride concentration in ppm in effluent

3. Adsorbent material: 30 grms., Temperature: 25°C, Flow rate 0.50 ml/min.

Sr. No.	Time of run in Minutes	Fluoride concentration in ppm. For		
		Sample concentration: 2 ppm	Sample concentration: 5ppm	Sample concentration: 10 ppm
1.	2.0	1.2	4.1	8.6
2	5.0	0.61	3.2	6.6
3	10.0	0.56	2.1	5.7
4	15.0	0.39	1.6	4.3
5	20.0	0.26	1.4	3.2
6	25.0	0.20	1.0	3.1
7	30.0	0.18	1.0	2.6
8	35.0	0.18	0.9	2.4

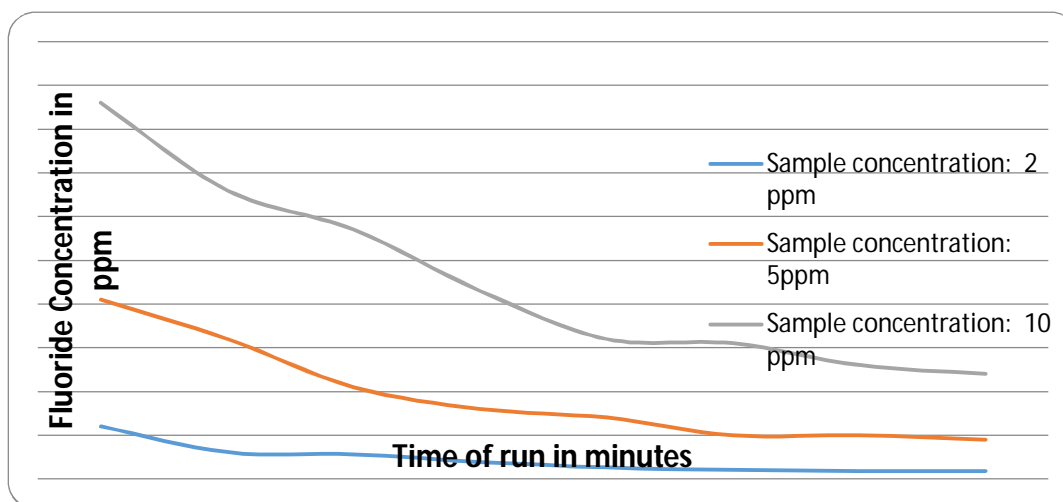


Figure: Graph showing removal of Fluoride as a function of Time of run.

It appears from some significant observations that 30 gms of solecite could effectively remove fluoride ranging from 40% to 91% at time intervals ranging from 2-30 minutes at a flow rate of 0.5ml/min. from a sample of 2 ppm fluoride concentration. At flow rate of 1ml/min the fluoride removal ranged from 30%-91% from a sample of 2 ppm fluoride concentration. After 30-35 minutes the removal of fluoride is reduced at different flow rates. It is also evident from the observations that an increase in flow rate increases the fluoride concentration in effluent water. Yet there is a scope of further investigation that up to what flow rate the removal is effective for short time of runs.

V. CONCLUSION

The above results indicate that the naturally occurring solecite is having fluoride retaining properties and can be applied for the defluoridation of drinking water. This method is innovative and materials of abundant natural occurrence are used for improving the water quality. This method also does not require any specified condition and can be applied under normal conditions. If method is further developed successfully with solecite it could be applied with other zeolites that are having natural occurrence. The zeolite adsorbent can be used as a column material and if required synthetic methods can be developed such as membranes (zeolite membranes) etc.

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