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Fluoride Removal by Adsorption Using Micro Porous Molecular Sieves ($4A^0$)

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Abstract: Fluoride contamination in drinking water due to natural and anthropogenic activities causes major problems though worldwide. Among several treatment technologies applied for fluoride removal, adsorption process has been explored widely and offer satisfactory results. Adsorption process by $4A^0$ molecular sieves done under various conditions (pH, initial fluoride concentration, contact time, adsorbent dose etc.) for fluoride removal. The adsorbents for de-fluoridation can be used micro porous materials ($<20A^0$) are effective, flexible, low-cost adsorbent to improve the pollution control on a commercial scale.

Key words: Fluoride, anthropogenic, molecular sieve ($4A^0$), adsorption, flexible.

I. INTRODUCTION

A fluoride is an ionic form of fluorine. The distribution of an ionic form of fluoride is uneven in the environment. Fluorides can presence in sedimentary formations in minerals, rocks, soil, clay, and anthropogenic activities. Small supplies of fluorine ion (1ppm) are useful in preventing tooth decay.[1] The industries which discharge waste water containing high fluoride concentrations include glass and ceramic production, semiconductor manufacturing, electroplating, coal fired power stations, beryllium extraction plants, brick and iron works and aluminum smelters. The effluents of these industries have higher fluoride concentrations than natural waters, ranging from ten to thousands of mg/L. It is estimated that more than 200million people worldwide rely on drinking water with fluoride concentrations that exceed the WHO guideline of 1.5mg/L.[2]

It's higher concentrations $>1.5\text{mg/L}$ causes irreversible demineralization of bones and tooth tissues i.e., skeletal and dental fluorosis, damage to the brain, harmful effects on kidney and liver. The fluoride removal from drinking water has been successful by different techniques are coagulation and precipitation, adsorption, ion exchange, membrane separation, electro dialysis, electro coagulation. Adsorption process for de-fluoridation has favored for the most part in developing countries as it is techno-economical to the variable method.[3] The various absorbents including activated carbon, activated alumina, metal oxides-hydroxides, bone chars, forex, magnesia and agricultural and industrial wastes were attempting to keep up the standard limit of fluoride concentration.[4] Usage of plant materials and agricultural wastes as absorbents are most adopted in developing countries like India that they are low-cost materials, available in abundance, biodegradable and natural existence.[3]

Here molecular sieves with $4A^0$ used as an adsorbent for de-fluoridation. Molecular sieve is a material with pores of uniform size (very small) hence adsorbing the small molecules not large ones. The diameter of molecular sieve is measured in angstroms (A^0) or nanometers. According to IUPAC notation, micro porous materials have pore diameters of $<2\text{nm}$ ($20A^0$) and macro porous materials have $>50\text{nm}$ ($500A^0$); the mesoporous materials lies in the middle with pore diameters between 2-50nm ($22-500A^0$).

II. MATERIALS AND METHOD

A standard fluoride stock solution is prepared by dissolving 2.21 g of anhydrous sodium fluoride in 1000 ml of double distilled water. Fluoride standard solution was prepared by diluting 100 ml stock solution to 1000 ml distilled water. This 1 ml solution has 0.1 mg of fluoride. The fluoride solution of required concentration is prepared by diluting this standard solution. The volume of sample taken for all the experiments is 1000 ml. All the adsorbents used in this study were collected and their particle size was of $4A^0$ molecular sieves (sodium alumino-silicate) The synthetic solution of fluoride prepared from analytical reagent sodium fluoride was stored in glass bottles. The pH of the solution was adjusted to the required level, using 0.1N H_2SO_4 and 0.1N NaOH solutions.

A. Batch studies

The batch experiments were carried out in 500 ml reagent bottle and by adding a pre-weighed amount of the adsorbent. The solution was then kept in an electric wrist shaker apparatus for stirring at a constant rpm. Varying contact time was given so that adsorption process gets followed properly. The solution was kept still so that adsorbent can get settled easily and then the adsorbent was separated with a filter paper. The concentration of fluoride remaining in the filtrate was analyzed by Ion analyzer. All the analysis was performed at room temperature.

III. RESULTS AND DISCUSSION

A. Effect of Contact time

Time is an essential parameter for attaining equilibrium between adsorbent and adsorbate. Adsorption performed in 180minutes in that achieved equilibrium at 150min with 51% of removal.

Table: 1. Adsorption of fluoride under various time periods.

Time (min)	Removal efficiency (%)
0	2
10	7
20	11
30	15
60	23
90	33
120	44
150	51

Fig.1. Effect of contact time on fluoride removal (initial concentration: 10 mg L⁻¹; pH: 3.0; adsorbent dose: 0.5 g/100 ml).

B. Effect of pH

The pH of the solution is an important parameter in the adsorption process. pH of the solution decreased from 10.0 to 3.0 increasing removal efficiency of fluoride up to 52%.

Table: 2. Removal efficiencies under various pH

Concentration	pH=3.0	pH=7.0	pH=10.0
2	11	14	10
4	34	30	27
6	41	42	37
8	51	39	47
10	30	27	28

The influence of initial solution pH on the fluoride removal efficiency of 4A⁰ molecular sieves (SAS) adsorbents was illustrated in

Fig. 2.

Fig. 2 Effect of pH on fluoride removal (initial concentration: 10 mg L⁻¹; Contact time: 150 minutes; adsorbent dose: 0.5 g/100 ml).

C. Effect of adsorbent dose

The adsorbent dosage influence the removal efficiency of fluoride. Increase in an adsorbent dose that increases the fluoride removal efficiency up to its limit.

Table: 3. Removal efficiencies under various adsorbent dosages.

Concentration	1.0grams	0.25grams
2	30	2
4	39	19
6	50	40
8	57	47
10	40	20

Increase the adsorbent dose from 0.25 to 1.0grams, increase its fluoride removal efficiency up to 57% as shown below:

Fig. 3. Effect of adsorbent dose on fluoride removal (contact time: 150min; pH of solution: 3.0; initial fluoride concentration: 10mg/l).

IV. CONCLUSION

The experimental investigations clearly suggest that abundantly available and low-cost materials like micro porous molecular materials (4A⁰) are effective in removing Fluoride from water to acceptable levels. Equilibrium isothermal sorption experiments

suggested that adsorbent dosages of 1.0g/l of micro porous molecular sieves (4A⁰) accomplished a removal of 57% of Fluoride. The time to reach equilibrium was observed to be 21/2 hours. pH has any significant impact in the range of 3.0, whereas pH of more than 3.0 resulted in a steep decrease in Fluoride removal. These micro porous molecular sieves are good in adsorption of smaller molecules hence used for sensitive molecules like Ions.

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