

Removal of fluoride Using Brick Powder by Adsorption

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ABSTRACT

Fluoride contamination in drinking water due to natural and anthropogenic activities has been recognized as one of the major problems worldwide imposing a serious threat to human health. Presence of fluoride in low concentration (<1.5 mg/l) is beneficial to the people for the calcification of dental enamel and maintenance of healthy bones. On the other side, its presence in higher concentration become a monster and can cause dental, skeletal fluorosis and also non-skeletal problems. Among several treatment technologies applied for fluoride removal, adsorption process has been explored widely and offers satisfactory results especially with mineral-based and/or surface modified adsorbents. It is evident from the literature survey that various adsorbents have shown good potential for the removal of fluoride. However, still there is a need to find out the practical utility of such developed adsorbents on a commercial scale, leading to the improvement of pollution control. Many investigators conducted experiments and developed various low-cost technologies to remove fluoride from water and wastewater. However, selection of appropriate treatment method depends on local situation. In this experiment fluorine removal from water by adsorption is studied. Optimum conditions of the experiment have been expressed. Environmental aspects of the adsorbent have been analysed.

Key words: adsorption, adsorbent, defluoridation, fluoride, human health, isotherm, low cost material, skeletal fluorosis

INTRODUCTION

Water is one of an essential commodity for the survival of all forms of lives on the earth. The population explosion and rapid industrial growth demands huge quantity of fresh water to meet various requirements. Of the total quantity of water (1.38x10⁹km³) available on the earth, 3% only is the fresh water. Of this 3% of fresh water, 1.74% is in glacier form, 1.7% as ground water and the rest in the other forms. Very small quantity of water (i.e 0.0132% of total water) only is available

in the rivers and lakes and most of these water sources are getting polluted due to waste disposal and human activities. India is one among the 23 nations in the world, where the fluoride contaminated groundwater is creating the health problems. Fluorosis was first detected in early 1930s in four states of India. In 1986, 13 states and in 1992, two more states were added to this list. It has been reported as many as 177 districts in 19 states and Union Territories of India have

already been affected by fluorosis. The most seriously affected states in India are Andhra Pradesh, Punjab, Haryana, Rajasthan, Gujarat, Tamil Nadu and Uttar Pradesh. The highest concentration observed to date in India is 48 mg/l in Rewari District of Haryana. Most of the rural population depends on the ground water sources for drinking purposes which generally contain underground deposits such as salts and minerals. One such is the fluoride which causes an adverse impact on human as well as animal health if fluoride concentration is higher. Consumption of water having excess fluoride over a prolonged period leads to a chronic ailment known as fluorosis. According to

the recent study, 62 million people are affected by various degrees of fluorosis in India alone. Out of these; 6 million are affected with a dental, skeletal and non-skeletal form of fluorosis and associated health problems. Fluorine is often called as two-edged sword. The positive side is about 96% of fluoride in the human body is found in bones and teeth. Fluorine is essential for the normal mineralisation of bones and formation of dental enamel. On the negative side, prolonged ingestion of fluoride through drinking water in excess of the daily requirement is associated with dental and skeletal fluorosis. Standards prescribed by various regulatory bodies for fluoride concentration in drinking water are different according to their climatic conditions. According to the World Health Organization (WHO), the standard prescribed for fluoride ion concentration in drinking water is 1.0 mg/l whereas by Bureau of Indian Standards (BIS) it is 1.0mg/l. Fluoride in smaller dose

(0.8-1.0mg/l) helps to prevent dental caries particularly in the children below 8 years of age. Fluoride in higher concentration causes dental fluorosis (1.5 -2.0mg/l) and skeletal fluorosis (>3.0mg/l).

SOURCES OF FLUORIDE

The amount of fluoride stored across the world in the ground is estimated to 85 million tons and nearly 12 million tons are located in India. The principal sources of fluorine were drinking water and food such as sea fish, cheese and tea. Fluoride is present in all waters particularly in ground water with higher concentrations. There has been an escalation in daily fluoride intake via the total human food and beverage chain. Carbonated soft drinks have considerable amounts of fluorides. Beers brewed in locations with high fluoride water levels may contribute significantly to the daily fluoride intake and sweetened iced teas contain significant amounts of fluoride. A single serving of chicken sticks alone would provide about half of a child's up per limit of safety for fluoride. Children's ingestion of fluoride from juices and juice-flavoured drinks can be substantial and a factor in the development of fluorosis. Fluoride is also present in most of everyday needs, viz. toothpastes, drugs, cosmetics, chewing gums, mouthwashes, and so on. The most common fluorine-bearing minerals, which constitute natural source for fluoride in drinking water are fluorite (CaF_2), apatite ($\text{Ca}_{10}(\text{PO}_4)_6\text{F}_2$), rock phosphate and topaz ($\text{Al}_2(\text{F},\text{OH})_2\text{SiO}_4$). Artificially high soil F levels can occur through contamination by application of phosphate fertilizers or sewage sludges, or from pesticides. Rocks in

southern India are rich with fluoride which forms the major reason for fluoride contamination in groundwater, and the granites in the district of Nalgonda, Andhra Pradesh contain much higher fluoride than

the world average fluoride concentration of 810 mg/kg. Fluoride contamination in ground water is a world-wide issue . The information regarding occurrence of fluoride in ground water in India is given in Table 1

Table 1: Summarized Information on the Occurrence of Excessive Fluoride in Ground Water in India

state	Number of habitation with excess fluoride	state	Number of habitation with excess fluoride
Andhra Pradesh	7548	Kerala	287
Madhya	201	Rajasthan	13560
Gujarat	2378	Meghalaya	33
Orissa	1138	Tamilnadu	527
Karnataka	860	Haryana	334
Punjab	700	Uttar Pradesh	1072
Himachal Pradesh	488	West Bengal	21

Conventional methods for fluoride removal

Treatment of water and wastewater containing fluoride ions requires a suitable and effective method. Membrane filtration, precipitation, nanofiltration, ion-exchange, electro coagulation, flotation, reverse osmosis and adsorption have been used for fluoride removal. Most of these methods have high operational and maintenance cost, low fluoride removal capacities, lack of selectivity for fluoride, undesirable effects on water quality, generation of large volumes of sludge and complicated procedures involved in the treatment. Among these methods, adsorption is the most effective and widely used method because it is universal, has a low maintenance cost, and is applicable for the removal of fluoride even at low concentrations. In recent years, considerable attention has been focused on the study of fluoride removal using natural, synthetic and biomass

materials such as activated alumina, fly ash, alum sludge, chitosan beads, red mud, zeolite, calcite, hydrated cement, attapulgit, and acid-treated spent bleaching earth.

Reverse osmosis is an excellent choice for the reduction of fluoride. Using a cellulose acetate/cellulose triacetate (CA/CTA) membrane, rejection rates of 80-90 percent are achievable when the pH is in the 4-8.5 range. Thin Film Composite membranes (TFC) will yield a higher rejection rate (up to 95 percent) in the 3-11 pH range. Both the CA/CTA and TFC membranes should be operated at a minimum membrane pressure differential of at least 30 psi.

Defluoridation methods can be broadly divided into three categories according to the main removal mechanism:

Chemical additive methods

Contact precipitation

Adsorption/ion exchange methods

Chemical additive methods

These methods involve the addition of soluble chemicals to the water. Fluoride is removed either by precipitation, coprecipitation, or adsorption onto the formed precipitate. Chemicals include lime used alone or with magnesium or aluminum salts along with coagulant aids. Treatment with lime and magnesium makes the water unsuitable for drinking because of the high pH after treatment. The use of alum and a small amount of lime has been extensively studied for defluoridation of drinking water. The most popular method for removal of fluoride from water is the Nalgonda technique, involves adding lime (5% of alum), bleaching powder (optional) and alum ($\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$) in sequence to the water, followed by coagulation, sedimentation and filtration. A much larger dose of alum is required for fluoride removal (150 mg/mg F-), compared with the doses used in routine water treatment.

Nalgonda technique is carried out with easily available chemicals and the method is economically attractive. The limitations of the method are varying alum doses depending on fluoride levels in water, daily addition of chemicals and stirring for 10-15 min, which many users may find difficult.

Contact Precipitation

Contact precipitation is a recently reported technique in which fluoride is removed from water through the addition of calcium and phosphate compounds. The presence of a saturated bone charcoal medium acts as a catalyst for the precipitation of fluoride either as CaF_2 , and/or fluorapatite. It gives high efficiency.

Adsorption/ion-exchange method

In the adsorption method, raw water is passed through a bed containing defluoridating material. The material retains fluoride either by physical, chemical or ion exchange mechanisms. The adsorbent gets saturated after a period of operation and requires regeneration.

A wide range of materials has been carried out for fluoride uptake such as Bauxite, magnetite, kaolinite, serpentine, various types of clays and red mud are some of the naturally occurring materials were studied. The general mechanism of fluoride uptake by these materials is the exchange of metal lattice hydroxyl or other anionic groups with fluoride. Fluoride uptake capacity can be increased by certain pre-treatment's like acid washing, calcinations, etc.

EXPERIMENTAL METHODOLOGY

Materials: Corning glassware of 'Pyrex' quality and analytical reagent grade chemicals were used. The glassware was soaked overnight in a 5.0 mg/L of Fluoride solution to minimize the possibility of Fluoride getting absorbed. The glassware was washed off with nitric acid and distilled water before use. First, a stock solution of 100 mg F/L was prepared by dissolving appropriate amount of sodium fluoride (NaF) in distilled water and desired concentrations of working solutions were then prepared from stock solution. Sulphuric acid (0.1N) and sodium hydroxide (0.1 N) were used for adjusting the pH values either to acidic or alkaline conditions.



Figure 1. brick powder

Aundantly available low cost materials like brick powder were utilized and shown in Figure 1.. Brick powder was sieved through IS sieves of 150 μm and 300 μm size and the material passing through 150 μm and retained on 300 μm which has a geometric mean size (Gm) of 212 μm was used in all experiments.

Non-flow batch sorption studies were conducted to study the effect of controlling

parameters like contact time, sorbent dosage, solution pH etc. Continuous down flow column studies were also conducted to study the practical applicability of brick powder for removal of Fluorides from water. All the experiments were conducted at room temperature ($29\pm 20\text{C}$). Fluoride concentration was estimated by SPADNS method (APHA) using a SYSTRONICS-105 spectrophotometer.



Figure 2. water samples for analysis

RESULTS AND DISCUSSION

Removal of Fluoride by brick powder

The effect of contact time on removal on Fluoride using brick powder is presented graphically as Fluoride removal in ppm at different contact times in Figure .3 It may be observed from the Figure that as contact time increases, ppm removal also increases

initially and reduces gradually with time and attains almost an equilibrium condition in nearly 72 hours and remains more or less constant thereafter. A maximum of 85 percent removal could be accomplished by brick powder.

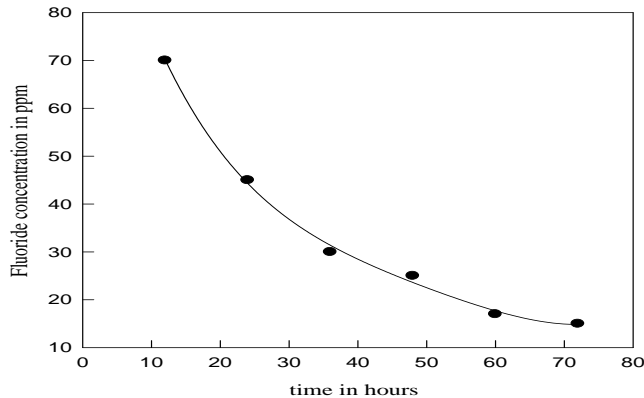


Figure 3. fluoride removal at different contact time

Effect of Solution pH

Removal of Fluoride by brick powder decreased continuously as pH was increased from 2.0 to 12.0.as depicted in Figure 4. Decrease in removal of Fluoride in pH range of 2.0 to 10.0 was low i.e., 12.5% whereas removal of Fluoride deceased significantly

from pH 10.0 to 12.0. Marginal variation in Fluoride removal by brick powder over pH range of 2 to 10 indicates its usefulness for removal of Fluoride in the pH range normally encountered in ground waters laden with Fluoride.

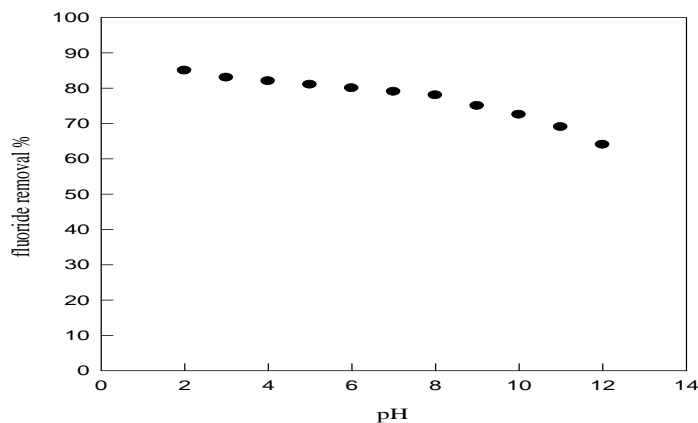


Figure 4. percentage fluoride removal with pH

Effect of Sorbent Dose

Results of equilibrium sorption experiments conducted with a test Fluoride solution of concentrations 10 mg /l are presented in Figure 5. The amount of Fluoride adsorbed increased with increase in dose and 84%

removal was accomplished at a dosage of 9g/l. At higher doses than 9g/l there is no increase in Fluoride removal perhaps due to non absorbability of Fluoride ions.

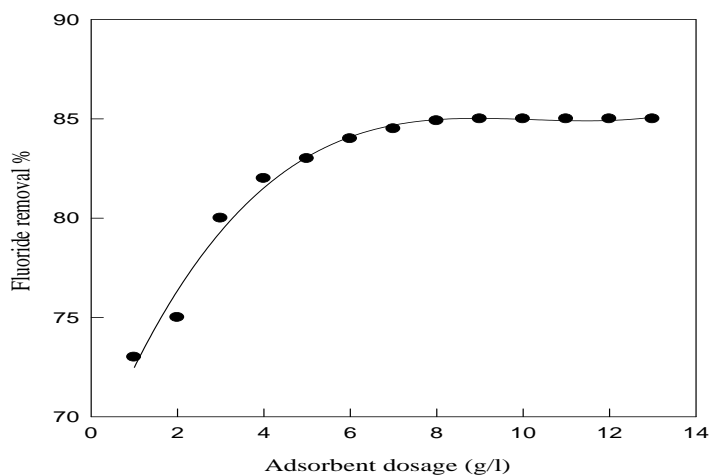


Figure 5. Effect of sorbent dose on Fluoride removal

CONCLUSION

The experimental investigations clearly suggest that abundantly available and low-cost materials like brick powder, is effective in removing Fluoride from water to acceptable levels. Equilibrium isothermal sorption experiments suggested that sorbent dosages of 9g/l of brick powder accomplished a removal of 85% of Fluoride. The time to reach equilibrium was observed to be 72 hours. pH does not have any significant impact in the range of 2-10, where as pH of more than 10 resulted in a steep decrease in Fluoride removal.

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Never think there is anything impossible for the soul. It is the greatest heresy to think so. If there is sin, this is the only sin; to say that you are weak, or others are weak.

~ Swami Vivekananda