

Removal of Fluoride from Ground Water by Using Bio-Adsorbent like *lantana camera* (Jamri)

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Abstract: Drinking water contamination by fluoride is recognized as a major public health problem in many parts of the world. In fact, although fluoride is an essential trace element for animals and humans, excessive fluoride intake may cause adverse health effects. Among several treatment technologies applied for fluoride removal, adsorption process has been explored widely and offers satisfactory results, so objective of this study was to investigate or check efficiency of low cost adsorbent The feasibility of low-cost biomass based adsorbent like *lantana camera* used for defluoridation of ground water at different pH and contact time the effect of this adsorbent on fluoride removal was compared with other available adsorbents. It was found to be much better adsorbent with high removal efficiency at higher concentration (5 ppm) of fluoride in ground water. The effect of time for adsorbent and initial concentration of fluoride on the % removal was also studied. Adsorption was found to be pH dependent with maximum removal efficiency at pH 7.0 (in neutral medium).

Keywords: Fluoride, Adsorption, Low cost Adsorbents, Contact time, Environmental Impact, pH.

1. Introduction

Fluoride is a naturally occurring compound derived from fluorine, the 13th most abundant element on Earth. It is found in rocks, soil, and fresh and ocean water. Fluoride occurs naturally in public water systems as a result of runoff from weathering of fluoride-containing rocks and soils and leaching from soil into groundwater. When the level of fluoride in water is beyond its permissible limit, it responsible for various types of fluorosis among human being. Around one million people in India are affected by endemic fluorosis [1, 2]. Maximum permissible limit of fluoride in drinking water has been set as 1.5 mg/ L by many regulatory authorities like WHO, US EPA, CPCB etc Since many other available methods of defluoridation are costlier, there is an urgent need of developing a low cost method. there are many processes available such as adsorption, ion exchange, electro dialysis, coagulation/ precipitation, dialysis, reverse osmosis, nano-filtration, ultra-filtration, etc., for the removal of fluoride from water [3-16]. All these processes have their advantages and limitations such as less efficiency, sensitive operating conditions, production of secondary sludge in application. However, adsorption is a simple and attractive method for the removal of metal from the effluents due to its high efficiency, easy handling and economic feasibility. Further, agro based adsorbents are getting more attention now a days due to their abundant availability and low cost. Some literatures are available on the removal of fluoride from water using various agro based adsorbents like rice husk ash, neem leaf, peepal leaf, khair leaf, tamarind fruit shell, etc. [17-19]. Fluoride is a commonly occurring element in minerals, geochemical deposits, and usual water systems and enters food chains through either drinking water or eating plants and cereals [20]. In most of this literature the concentration of fluoride is in between (1.5-5 mg/l), In India, endemic fluorosis affects more than one million populations and is a major problem in 17 of the 29 states. Similar health problems due to high fluoride content in ground water have also been reported worldwide and it is estimated that around 260 million people are adversely affected in 30 countries of the world [21]. The

effects of fluoride on human health are dependent on the concentration of fluoride in water [22].

Table 1: Biological Effects on Human Health

| Fluoride conc. (mg/lit) | Source | Effects |
|-------------------------|-------------|------------------------------|
| 1 | Water | Prevention of dental caries |
| 2 | Water | Effect dental enamel |
| 3 to 6 | Water | Osteoporosis |
| 8 | Water | 10 % Osteoporosis |
| 20 to 80 | Air & Water | Crippling skeletal fluorosis |

Permissible Limits for Fluoride Concentration in Drinking Water [23]

- Bureau of Indian Standards (BIS)-0.6 to 1.2 mg/lit
- World health Organization (WHO-1984) for drinking water-1 to 1.5 mg/lit
- Indian Council of Medical Research (ICMR-1975)-1 mg/lit
- World Health Organization (WHO) European Standards- 0.7 to 1.7 mg/lit related to temperature.

Dental fluorosis, also called “mottled enamel”, occurs when the fluoride level in drinking water is marginally above 1.0 mg/l. A relationship between fluoride concentration in potable water and mottled enamel was first established in 1931. Typical manifestations of dental fluorosis are loss of shining and development of horizontal yellow streaks on teeth. Since this is caused by high fluoride in or adjacent to developing enamel, dental fluorosis develops in children born and brought up in endemic areas of fluorosis. Once formed, the changes in the enamel are permanent. When the above manifestations are seen in an adult, they clearly indicate that the person has been exposed to high fluoride levels during her or his childhood. Skeletal fluorosis affects both adults and children and is generally manifested after consumption of water with fluoride levels exceeding 3 mg/l. typical symptoms of skeletal fluorosis are pain in the joints and backbone. In severe cases this can result in crippling the patient. Recent studies have shown that excess intake of fluoride can also have certain non-skeletal health impacts such as gastro-intestinal problems, allergies, anemia and

urinary tract problems. Nutritional deficiencies can enhance the undesirable effects of fluoride.

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

- Chemical additive methods
- Contact precipitation
- Adsorption/ion exchange methods

2. 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 tried for fluoride uptake. Bauxite, magnetite, kaolinite, serpentine, various types of clays and red mud are some of the naturally occurring materials 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-treatments like acid washing, calcinations, etc. None of the mentioned materials generally exhibits high fluoride uptake capacities. Rajasthan is one state where fluoride in high level is prevalent in all the 32 districts and has become a serious health hazard in 18 of them.

According to the survey conducted by the Public Health Engineering Department in the recent past, the drinking water sources in 9741 out of 37889 villages, 25.7 %, and 6819 out of 45311, 15 %, habitations were found to contain fluoride more than 1.5 mg/L In the absence of perennial rivers, surface sources and canal systems, groundwater, which generally contains high fluoride concentrations, Many natural and low cost materials such as red mud [24,25], zirconium impregnated coconut shell carbon [26], cashew nut shell carbon [27], ground nut shell carbon and clays [28] have been used as adsorbents for fluoride removal from drinking water. Recently, amorphous alumina supported on carbon nanotubes [29], aligned carbon nanotubes [30], ion exchange polymeric fiber [31], and an ion exchanger based on a double hydrous oxide of Al and Fe (Fe₂O₃Al₂O₃xH₂O) [32] have been assayed for removing fluoride from drinking water as well as industrial wastewater.

3. Material and Methods

Preparation of Standard fluoride solution (5ppm solution)

A stock solution of 100mg/ liter (100 ppm) of fluoride was prepared by dissolving 221 mg of anhydrous sodium fluoride (NaF) in 1000 ml distilled water. Now 5 ppm solution prepare from stock solution by suitable dilution.

Preparation of adsorbent *lantana camera*

Lantana camera leaves easily available at tree, collect and washed with distilled water several times then dried in sunlight for 3-4 days . Now grind this dry leaves with mixer & sieved with suitable mesh size (30 bss size) taken for analysis

Measurement

To observe fluoride Removal used Fluoride Ion Meter Panomex Model PX/IMC/321. First Calibrate with 100 ppm & 10 ppm solution prepared by stock solution. Now check 5 ppm fluoride solution for system response Washed Plastic beaker with tap water then distilled water Take 100 ml of 5 ppm solution of water in 250ml plastic beaker weigh 1.0 gm of adsorbent hold for 30 minutes after 30 min filter the solution take reading using various parameter like pH, dose of adsorbent and contact time of adsorbent.

4. Results & Discussion

(i) Effect of pH

| pH | Initial concentration (ppm) | Final concentration (ppm) | Removal in ppm | % Removal |
|------|-----------------------------|---------------------------|----------------|-----------|
| 2.0 | 5.0 | 4.2 | 0.8 | 16 |
| 3.0 | 5.0 | 3.9 | 1.1 | 22 |
| 4.0 | 5.0 | 4 | 1.0 | 20 |
| 5.0 | 5.0 | 3.5 | 1.5 | 30 |
| 6.0 | 5.0 | 3.1 | 1.9 | 38 |
| 7.0 | 5.0 | 2.7 | 2.3 | 46 |
| 8.0 | 5.0 | 3.2 | 2.1 | 42 |
| 9.0 | 5.0 | 3.3 | 1.7 | 34 |
| 10.0 | 5.0 | 3.2 | 1.8 | 36 |
| 11.0 | 5.0 | 3.2 | 1.8 | 36 |

Figure 1

The pH can influence the surface charge of the adsorbent, the degree of ionization also the species of adsorbate. In a particular pH range, most metal sorption is enhanced with pH, increasing to a certain value followed by a reduction when further pH increases. The dependence of the metal uptake on pH can be associated with both the surface functional groups on the biomass' cell walls and also the metal chemistry of the solution The effect of pH on removal of fluoride was studies in the range of 2-11 and results are illustrated in the Fig-1. pH plays an important role in adsorption process on bio adsorbents. The removal efficiency of the adsorbents is concluded that, it is depending on the pH of the test sample of fluoride, as shown in Fig.1. The results confirm a strong dependence between the adsorption of fluoride and pH, whereby adsorption appears to increases with increasing pH, within a pH range of 1–7. Maximum adsorption was observed at a pH of 7 and 8 for *lantana camera*

(ii) Dose of Adsorbent (*lantana camera*)

| S. No. | wt of adsorbent (gm) | Amount of water 5.0 ppm (ml) | initial concentration before treatment (ppm) | after 5.0 hrs (ppm) | ppm removal | % Fluoride Removal |
|--------|----------------------|-------------------------------|--|---------------------|-------------|--------------------|
| 1 | 0.2506 | 100 ml | 5.00 | 2.7 | 2.30 | 46 |
| 2 | 0.5016 | 100 ml | 5.00 | 2.5 | 2.50 | 50 |
| 3 | 1.016 | 100 ml | 5.00 | 2.2 | 2.80 | 56 |
| 4 | 2.0138 | 100 ml | 5.00 | 1.9 | 3.10 | 62 |

Figure 2

Initial concentration of metal ions can alter the metal removal efficiency of fluoride through a combination of

factors such as the availability of specific surface functional groups and the ability of surface functional groups to bind metal ions. Initial concentration of solution can provide an important driving force to overcome the mass transfer resistance of metal between the aqueous and solid phases. Removal efficiency of fluoride is strongly dependent on concentration of adsorbent dose in test sample. Removal of fluoride increases as increasing dose of adsorbent in the sample as shown in Fig-2. At the starting, removal of fluoride increases as increasing the dose until a some extent after that very slightly change in the removal of fluoride it means, the curve lapse as flat indicating the higher fluoride adsorption occurs at their maximum dose and the removal remains constant. Adsorbents have a higher availability of surface and pore volume because of this adsorption increases after that adsorption of fluoride is constant at higher dose because of saturation of pore volume and surface

(iii) Effect of Contact Time

(a)

| dose gm/100ml | Contact time (min) | initial conc | final fluoride (mg/l) | Reduction of fluoride | % removal efficiency |
|---------------|--------------------|--------------|-----------------------|-----------------------|----------------------|
| 0.2501 | 30 | 5.00 | 4.4 | 0.60 | 12 |
| 0.2501 | 60 | 5.00 | 3.5 | 1.50 | 30 |
| 0.2501 | 120 | 5.00 | 3.2 | 1.80 | 36 |
| 0.2501 | 180 | 5.00 | 3 | 2.00 | 40 |
| 0.2501 | 240 | 5.00 | 2.8 | 2.20 | 44 |
| 0.2501 | 300 | 5.00 | 2.7 | 2.30 | 46 |

(b)

| dose gm/100ml | Contact time (min) | initial conc | final fluoride (mg/l) | Reduction of fluoride | % removal efficiency |
|---------------|--------------------|--------------|-----------------------|-----------------------|----------------------|
| 0.5016 | 30 | 5.00 | 4.2 | 0.80 | 16 |
| 0.5016 | 60 | 5.00 | 3.2 | 1.80 | 36 |
| 0.5016 | 120 | 5.00 | 3 | 2.00 | 40 |
| 0.5016 | 180 | 5.00 | 2.7 | 2.30 | 46 |
| 0.5016 | 240 | 5.00 | 2.5 | 2.50 | 50 |
| 0.5012 | 300 | 5.00 | 2.5 | 2.50 | 50 |

(c)

| dose gm/100ml | Contact time (min) | initial conc | final fluoride (mg/l) | Reduction of fluoride | % removal efficiency |
|---------------|--------------------|--------------|-----------------------|-----------------------|----------------------|
| 1.016 | 30 | 5.00 | 4 | 1.00 | 20 |
| 1.016 | 60 | 5.00 | 2.8 | 2.20 | 44 |
| 1.016 | 120 | 5.00 | 2.5 | 2.50 | 50 |
| 1.016 | 180 | 5.00 | 2.2 | 2.80 | 56 |
| 1.016 | 240 | 5.00 | 2.2 | 2.80 | 56 |
| 1.016 | 300 | 5.00 | 2.1 | 2.90 | 58 |

(d)

| dose gm/100ml | Contact time (min) | initial conc | final fluoride (mg/l) | Reduction of fluoride | % removal efficiency |
|---------------|--------------------|--------------|-----------------------|-----------------------|----------------------|
| 2.01 | 30 | 5.00 | 3.8 | 1.20 | 24 |
| 2.01 | 60 | 5.00 | 2.6 | 2.40 | 48 |
| 2.01 | 120 | 5.00 | 2.4 | 2.60 | 52 |
| 2.01 | 180 | 5.00 | 2 | 3.00 | 60 |
| 2.01 | 240 | 5.00 | 2 | 3.00 | 60 |
| 2.01 | 300 | 5.00 | 1.9 | 3.10 | 62 |

Figure 3

It is observed that the exclusion of fluoride ions increases with increase in contact time to some level at optimum pH and dose. Further increase in contact time does not increase the Fig-3 explains the optimum percentage removal of fluoride by three considered bio adsorbents at different contact times. The adsorption of metal ion by adsorbent also depends on the interactions of functional groups between the solution and the surface of adsorbent. Adsorptions can be assumed to be complete when equilibrium is achieved between the solute of solution and the adsorbent. However, specific time is needed to maintain the equilibrium interactions to ensure that the adsorption process is complete. However, it progressively approached an almost steady value, denoting accomplishment of equilibrium, these similar trends also observed in Effects of stirring rate and temperature on fluoride removal by fishbone charcoal [33]

(iv) Effect of Temperature

| Dose (g/100ml) | Temperature (in C) | Initial fluoride (mg/l) | Final fluoride (mg/l) | Reduction in fluoride (mg/l) | % removal efficiency |
|----------------|--------------------|-------------------------|-----------------------|------------------------------|----------------------|
| 1.0 | 22 | 5.0 | 2.6 | 2.4 | 48 |
| 1.0 | 30 | 5.0 | 2.4 | 2.6 | 52 |
| 1.0 | 40 | 5.0 | 1.9 | 3.1 | 62 |
| 1.0 | 50 | 5.0 | 1.6 | 3.4 | 68 |

Figure 4

This is an important observation to note when attempting to apply defluoridation methods on site in hot climates, for sorption capacities attained under room temperature conditions may be higher than in the field as a result of increased temperatures [34]. This is an important observation to note when attempting to apply defluoridation methods on site in hot climates, for sorption capacities attained under room temperature conditions may be higher than in the field as a result of increased temperatures [35]. Temperature plays a double role in the fluoride sorption process. Temperature can impact the physical binding processes of fluoride to a sorbent. However, temperature also can have a direct impact on the physical properties of a sorbent, if thermally treated prior to exposure, so that sorption capacities can be significantly altered. As temperature increased, sorption was shown to be less favored most likely due to increased deprotonation or hydroxylation of the surface causing more negatively charges sorbent surfaces.

(v) Physical Characterization

The *lantana camera* leaf powder before (untreated) and after (treated) batch adsorption was Characterized using SEM (Scanning electron microscopy). A representative experiment was carried out under the given conditions: initial fluoride concentration = 5 mg/L, temperature = 30°C, shaking speed = 150 rpm and contact time = 24h respectively, for the preparation of SEM samples.



Figure 5.1

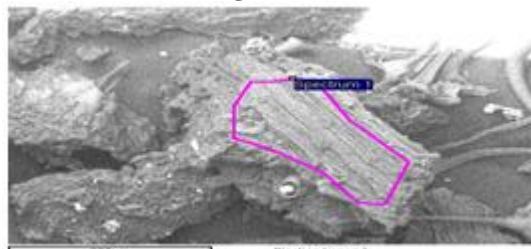


Figure 5.2

The SEM images of the *lantana camera* leaf powder before treatment of fluoride ions shows cluster of very fine particles with a surface coating while the SEM images of the *lantana camera* leaf powder after treatment of fluoride ions shows small irregularly shaped fine particles mostly fluoride ions adhering to the surfaces of the adsorbent. Fig. 5.1 & 5.2

5. Conclusions

The paper briefly highlighted the importance of adsorption process and its benefits. Also the overview of various papers publishes in various journals on removal of fluoride ions from water or wastewater by adsorption using various low cost adsorbents instead of expensive commercial adsorbents. The efficiency of different adsorbents in the removal of fluoride depends on dose of adsorbate, characteristics of adsorbent, pH, temperature, contact time, etc. Observation of various low cost adsorbents presented here shows a great potential for the fluoride removal. The use of commercially available adsorbents can be replaced by the inexpensive and effective low cost adsorbents. There is need for more studies to better understand the process of low-cost adsorption and to demonstrate the technology effectively.

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