Studies on Defluoridation Techniques: A Critical Review

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Abstract: Fluoride is frequently depicted as a ‘double-edged sword’ as an insufficient intake is related to dental caries and too much ingestion leads to dental and skeletal fluorosis which has no treatment. Prevention is only through the supply of fluoride safe water by using de-fluoridation techniques. This review article is aimed at providing specific information on attempts made by various researchers in the field of fluoride removal from polluted waters. Various de-fluoridation techniques are currently in use viz., precipitation, ion-exchange, reverse osmosis, electro dialysis, donnan dialysis, nanofiltration, electro coagulation and adsorption etc. And among these techniques, adsorption process is economical, efficient, ease of operation and produces high-quality water. Studies on fluoride removal from waste waters using various adsorbents such as alumina/aluminium based materials, lime, clays and soils, calcium based minerals, bone, bone char, synthetic compounds and carbon based materials are reviewed. The present paper reviews the techniques available and ongoing attempts for fluoride removal from polluted waters.

Key words: Fluoride pollution, fluorosis, de-fluoridation techniques, adsorption.

Introduction

The major sources of water pollution can be classified as: Municipal, Industrial and Agricultural. Industries discharge a variety of pollutants in their waste water including anionic and cationic contaminates, heavy metals, organic toxins, dissolved inorganic compounds, oils, nutrients, solids and dyes. Humic acids, hydrocarbons and pectins are amongst the natural organic substances occurring in groundwater. The presence of various hazardous contaminants like fluoride, nitrate, pesticides, arsenic, other heavy metals, etc. in under groundwater has been reported from different parts of India¹-⁶. Fluoride is a potential pollutant in India and it is mainly of natural origin found in groundwater and it leaches into the groundwater due to disintegration and dissolution of igneous and metamorphic rocks containing minerals such as amphiboles and micas⁷. Further, the effluents from chemical, metallurgical industries and coalmines also contribute to the fluoride contamination in surface waters.

Fluorine is a ubiquitous and the 13th most abundant naturally occurring element in the Earth’s crust, one of the most reactive and electronegative of all the elements⁸. Fluorides are found in a wide variety of minerals, including fluorspar [CaF₂], sellaite [MgF₂], rock phosphate/fluoroapatite [Ca₅(PO₄)₃F], cryolite [Na₃AlF₆], mica, hornblende and others⁹. Fluorite (CaF₂) is a common bearer of fluoride and is found in granite, granite gneisses and pegmatite¹⁰-¹¹ and as cryolite in igneous rocks. Fluoride also associated with mono valent cations such as NaF and KF which are water soluble.
Fluoride pollution in the environment occurs through two different channels which are natural and anthropogenic sources\textsuperscript{12}. During weathering of alkali, igneous and sedimentary rocks and circulation of water in fluoride bearing rocks and soils, fluorine can be reached out and dissolved in groundwater\textsuperscript{13-18}.

A large number of fluoride contaminated areas are founded throughout the world where ground waters contain exceeded levels of fluoride. In worldwide, India is most severely fluoride affected country. Of the 85 million tons of fluoride deposits on the earth’s crust, 12 million are found in India\textsuperscript{19}. In India in 2002, 17 states have been identified as endemic for fluorosis\textsuperscript{20} and now the problem exist in more than 17 states\textsuperscript{21-23}. In India, concentrations of fluoride in drinking water in different parts of the country vary from 0.5 to 50 mg/lit and the extent of fluoride contamination in groundwater varies from 1.0 to 48 mg/lit\textsuperscript{24}. In India alone, a total of 60-70 million people including children are affected with dental and skeletal fluorosis\textsuperscript{25-27}. Several areas of composite state of Andhra Pradesh have fluoride concentration greater than the permissible limit by WHO.

Fluoride has dual significance, i.e., beneficial effects on teeth at low concentrations in drinking-water\textsuperscript{28-29} but on excessive exposure to fluoride in drinking-water can give rise to a number of adverse effects. The permissible limits of fluoride concentration in drinking water prescribed by various organizations\textsuperscript{24, 30} are shown in Table 1. The requirement of fluoride content varies among countries and depends on the geography and the age of people involved\textsuperscript{31}.

<table>
<thead>
<tr>
<th>S. No.</th>
<th>Name of Organization</th>
<th>Permissible limit of fluoride ion (mg/lit)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>World Health Organization (International standard for drinking water)</td>
<td>0.6-1.5</td>
</tr>
<tr>
<td>2</td>
<td>US Public Health Standards</td>
<td>0.8</td>
</tr>
<tr>
<td>3</td>
<td>The committee on public health engineering manual and Code of practice, Government of India, Indian Council of Medical Research (ICMR)</td>
<td>1.0</td>
</tr>
<tr>
<td>5</td>
<td>Bureau of Indian Standards (BIS)</td>
<td>0.6-1.5</td>
</tr>
</tbody>
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Table 1: Permissible limit of fluoride in drinking water prescribed by various Organizations

Excessive fluoride exposure may cause irreversible demineralization of bone and tooth tissues, a condition known as fluorosis. With prolonged exposure to higher fluoride concentrations, dental fluorosis progresses to skeletal fluorosis. The World Health Organization permits maximum fluoride concentration in drinking water up to 1.5 mg/lit\textsuperscript{32} at which fluorosis is low and this is being followed in most of the nations and is also the Australian recommended limit\textsuperscript{33-34}. The effect of fluoride on human health by the prolonged use of drinking water with fluoride content\textsuperscript{35-36} is presented in Table 2.

<table>
<thead>
<tr>
<th>Fluoride ion concentration,(mg/lit)</th>
<th>Health outcome</th>
</tr>
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<tbody>
<tr>
<td>&lt;0.5</td>
<td>Dental caries</td>
</tr>
<tr>
<td>0.5-1.5</td>
<td>Optimum dental health</td>
</tr>
<tr>
<td>1.5-4.0</td>
<td>Dental fluorosis</td>
</tr>
<tr>
<td>4.0-10</td>
<td>Dental and skeletal fluorosis</td>
</tr>
<tr>
<td>&gt;10.0</td>
<td>Crippling fluorosis</td>
</tr>
</tbody>
</table>

Table 2: Health effects on prolonged use of fluoridated drinking water

Fluoride is thus considered beneficial in drinking water at levels of about 0.7 mg/lit but once it exceeds 1.5 mg/lit causes adverse effects including severe dental, skeletal fluorosis and crippling fluorosis\textsuperscript{27, 36-40} and also other adverse affects on human health have been reviewed\textsuperscript{41-42}.

Hence, it is necessary to reduce the fluoride concentrations to permissible levels and for which wonderful investigations are being put all over the world. The present paper reviews the various techniques available for de-fluoridation of water.
Methods of Defluoridation

In India fluoride contamination in drinking water is mainly with rural population, where largely they depend on the groundwater as their drinking water sources. An excessive level of fluoride in drinking-water is a serious problem. The only way to protect from fluorosis is supply of drinking water with recommended levels of fluoride concentration. This can be accomplished by de-fluoridation of fluoride-contaminated water. De-fluoridation is “adjustment of level of fluoride in drinking water to the optimal level”.

It is better to select an appropriate de-fluoridation technique by considering the local conditions, economic status, adoptability of the method, knowledge of community, easy availability of materials and reuse of exhausted materials for treatment purpose etc. The techniques which use locally and cheaply available materials as de-fluoridation agents are preferred. Now-a-days, varieties of methods are available for fluoride removal mainly based on the mechanism such as precipitation[^43-48], ion-exchange[^49-54], reverse osmosis[^55-56], Donnan dialysis[^57-58], electro dialysis[^59-62], nano filtration[^63], membrane based methods[^64-67], electro coagulation[^68] and adsorption on to various adsorbents[^68-73].

Precipitation Methods:

Precipitation methods are based on the addition of chemicals (coagulants and coagulant aids) and the subsequent precipitation of a sparingly soluble fluoride salt as insoluble fluorapatite[^74]. Fluoride removal is accomplished with the separation of solids from liquid. Limestone (calcite) can remove the fluoride from water through a combination of adsorption and precipitation processes[^45, 75-76] and magnesium oxide also acts as the precipitating agent[^77-78]. Aluminium salts (eg. Alum), lime, Poly Aluminium Chloride, Poly Aluminium Hydroxy sulphate and Brushite are some of the frequently used materials in de-fluoridation by precipitation technique[^74, 79]. The best example for this technique is the famous Nalgonda technique of de-fluoridation.

In 1961, National Environmental Engineering Research Institute (NEERI), Nagpur has evolved an economical and simple method for removal of fluoride which is referred to as Nalgonda Technique[^43]. Nalgonda Technique involves addition of Aluminium salts, lime and bleaching powder followed by rapid mixing, flocculation, sedimentation, filtration and disinfection. The process is aluminium sulfate based coagulation-flocculation sedimentation. When Aluminium sulfate, $\text{Al}_2(\text{SO}_4)_{3} \cdot 18\text{H}_2\text{O}$, is added to the water, Aluminium hydroxide micro-flocs are produced rapidly. As the aluminium sulfate solution is acidic, simultaneous addition of lime is often needed to ensure neutral pH in the treated water and complete precipitation of aluminium. Surplus lime is used to facilitate more complete settling. Lime facilitates forming larger and dense flocks for rapid settling. During this flocculation process many kinds of micro-particles and negatively charged ions including fluoride are partially removed by electrostatic attachment to the flocs. The best fluoride removal is accomplished at pH range of 5.5-7.5[^80]. Bleaching powder is added to the raw water at the rate of 3 mg/lit for disinfection. Bulusu et al.,[^81] stated that Nalgonda Technique is preferable at all levels because of the low price, ease of handling and suitable for either community or household use[^82]. The technique is highly versatile and has been claimed as the most effective technique for fluoride removal[^43, 83]. The Nalgonda technique is modified by using Poly Aluminium Chloride because its efficiency is high when compared with Alum and less cost; less flocculation time is sufficient for Poly Aluminium Hydroxy Sulphate than the alum.

Limitations

1. The Nalgonda technique is more time consuming and requires more diligence than other de-fluoridation techniques.
2. It is difficult to regulate the pH with the addition of lime, the correct dose of chemicals to be added.
3. Removes only (18-33%) of fluoride in the form of precipitates and converts (67-82%) of fluoride into soluble toxic $\text{Al}^{3+}-\text{F}^{-}$ complex ions[^84-86].
4. A large dose of aluminium sulphate, up to 700-1,200 mg/lit, may be needed. The large dose results in a large sludge disposal problem in the case of water works treatment. And sulfate ion concentration crosses the maximum permissible limit of 400 mg/lit, which causes ill-effects in human beings.
5. The residual aluminum in excess of 0.2 mg/lit in treated water causes dangerous dementia disease as well as pathophysiological, neuro-behavioral, structural and biochemical changes.
6. It also affects musculoskeletal, respiratory, cardiovascular, endocrine and reproductive systems[^85].
Ion-Exchange Methods:

The different ion exchange materials studied include bone, bone char and activated alumina, serpentine, anion and cation exchange resins.

**Bone:**

The degreased, acid and alkali treated and grounded bones were first used in the form of home filters and showed a good capacity of fluoride removal from the water and showed that it could reduce fluoride concentration very effectively from 3.5 mg/lt to 0.2 mg/lt. The removal mechanism suggested was an exchange of the carbonate radicals with fluoride.

**Limitations**

1. The use of these materials was discontinued due to some objections of taste of treated water.
2. It was not used widely, because of the high cost of the bone and costly regeneration at domestic level.

**Bone char:**

Ms. Nutthamon Fangsrekam described the process of de-fluoridation by bone char as the ion-exchange. Bone char produced by carbonizing bone at temperature of 1100-1600°C had superior qualities than those of unprocessed bone. Bone char is cellular in structure and consists of calcium phosphate and carbonates. Bone charcoal is the oldest known water de-fluoridation agent, being successfully used since the 1940s. Using of a bone charcoal column for de-fluoridation of water is very simple and rapidly regenerated without significant loss of binding capacity for fluoride. Bone charcoal de-fluoridation technique has been replaced by the use of ion-exchange resins and activated alumina.

**Limitations**

1. For bone charcoal there is no social acceptance because of some religious and cultural objections.
2. The bone char harbors bacteria and hence unhygienic.
3. It is a technique sensitive procedure, since the efficiency of bone char as an adsorbent for fluoride is a function of the charring procedure which should be done cautiously and when the material is exhausted, the fluoride uptake is ceased.

**Ion-exchange resins:**

For de-fluoridation of water various types of anion and cation exchange resins have been used. Some of the anion exchangers are Tulsion A-27, Deaceodite FF-IP, Amberlite IRA 400, Lewatit MIH-59, Polyanion (NCL), Amberlite XE-75 and cation exchangers are Carbion,Wasoresin-14, Polystyrene resin, Sulphonated saw dust carbon. The de-fluoridation of water by a chloride-loaded anion exchanger Amberlite IRA-400 was studied. The fluoride exchange capacity of resins depends upon the ratio of fluoride to total anions in water.

**Limitations**

1. A large volume of regenerate is required for the regeneration of cation and anion exchange resins.
2. The resins are complex, contamination prone and expensive and the waste produced is very large.

**Activated Alumina:**

UNICEF has sponsored research and development on the use of activated alumina for removal of fluoride from water in rural India. Patil and Kulkarni studied the ion exchange resin and activated alumina as defluoridating media. Activated alumina is a granular, highly porous material consisting essentially of aluminium oxide (Al₂O₃). When the water passes through a packed column of activated alumina, pollutants and other components in the water are adsorbed onto the surface of the grains. Activated alumina is used to treat water with fluoride concentrations from 4-20 mg/lit in an adsorption process. Many reports are available for the de-fluoridation of water on large scale installations. In rural as well as urban water treatment, activated alumina is used as an adsorbent.
A wide array of adsorbents has been used for de-fluoridation which includes activated and impregnated alumina\textsuperscript{76, 73, 75, 103-120}. Alumina composites have been examined as de-fluoridation adsorbents. These include alumina cement granule\textsuperscript{121-122}, manganese oxide coated alumina\textsuperscript{123}, magnesia-amended activated alumina\textsuperscript{124}, hydrous manganese oxide-coated alumina\textsuperscript{125}, and Manganese dioxide-coated activated alumina\textsuperscript{126}. Electro activated alumina has also been used for defluoridination of water\textsuperscript{127}. The kinetics of removal of fluoride from groundwater has been studied by using activated alumina\textsuperscript{128}.

**Limitations**

1. De-fluoridation with activated alumina is possible only at specific pH range, needing pre-and post-pH adjustment of water.
2. Some toxic aluminium-ions may enter in water\textsuperscript{76}.
3. The technique is expensive because frequent activation of alumina is needed.
4. Disposal problems arise because of high fluoride concentrated solution is generated during regeneration.
5. The efficiency of the activated alumina diminishes with increasing number of usage-generation cycle.

**Membrane Filtration Process:**

Reverse osmosis (RO) and electro dialysis are two membrane filtration processes which can be used for the removal of fluoride. De-fluoridation studies on membrane based have been reported in Finland\textsuperscript{129}. Similarly, use of Reverse Osmosis membranes for fluoride removal from contaminated water sources has also been reported. RO membrane rejects ions based on size and electrical charge. Membranes provide an effective barrier to the suspended solids, all inorganic pollutants, organic micro pollutants, pesticides and microorganisms, etc. and are highly effective for fluoride removal up to 85-95%.

In reverse osmosis, the pressure is exerted on one side of the semi-permeable membrane which forces the water across the membrane leaving the pollutants behind and extremely high pure water is produced. Using a very small 7 cm reverse osmosis cell containing a commercially available cellulose acetate membrane on aqueous solutions containing only fluoride, the concentration of fluoride can be lowered from 58.5 to 1.0 gm/lt. Few authors have studied reverse osmosis technique for purification of water\textsuperscript{130-135}.

In electro dialysis, the membranes allow the ions to pass but not the water. In electro-dialysis pollutants (ionic components) can be removed from aqueous solution through ion exchange membranes under the influence of an electric field. Electro-dialysis can bring the fluoride level in water to within WHO guidelines\textsuperscript{136} and electro-dialysis is effective in de-fluoridation of waters having even 3000 mg/lt of total dissolved salts and 3mg/lt fluorid\textsuperscript{137}. Electro-dialysis can reduce the TDS and fluoride from 5000 mg/lt to 600 mg/lt and 15 mg/lt to 1.5 mg/lt respectively.

**Limitations**

1. The membranes are sensitive to temperature, pH and arises maintenance problem because of plugging, fouling by particulate matter, concentrated with large quantity of wastes.
2. Both the processes are expensive and very complicated\textsuperscript{86, 96}.
3. Removes all the ions present in water including some essential minerals for proper growth and hence re-mineralization is required after treatment.
4. High energy consumption and large amount of water gets wasted as brine.

**Donnan dialysis:**

Donnan dialysis is also a separation process based on membrane filtration that utilizes counter diffusion of two or more ions through an ion-exchange membrane to achieve a separation. The process is named in honor of F. G. Donnan\textsuperscript{138} who described the equilibrium that resulted when a semi-permeable membrane separated two solutions of electrolytes, NaA on one side and KA on the other. Donnan dialysis (DD) is highly efficient in treating fluoride contaminated water and used for the treatment of low-concentration waters. In earlier studies, it was tested for the fluoride removal from diluted NaF solutions and synthetic waters, which were the models of waters contaminated by fluoride from countries of Africa\textsuperscript{68, 139-140}. A hybrid process that combines the adsorption on conventional solid adsorbents such as aluminum and zirconium oxides with a specific DD
procedure is applied to treat groundwater with an excessive fluoride concentration of 4 mg/lit resulting from phosphate mining in Morocco\textsuperscript{57}.

Limitations

1. Reduced efficiency in high-saline waters.
2. Expensive technique.

Nano filtration:

Nano filtration is a process which takes in the upper end (in separation size terms) of reverse osmosis, and the lower end of ultra filtration. Permeability of Nano filtration membranes is higher than those of RO. Nano filtration membranes have high retention of charged particles. It requires less pressure and capital than RO and it is widely applicable specially for drinking and waste water treatment and is used in de-fluoridation studies\textsuperscript{141-144}.

Limitations

1. The process is expensive and complicated.

Electro Coagulation:

The electro coagulation is the process of utilizing electricity and sacrificed anodes to form active coagulant which is used to remove pollutant by precipitation and flotation. The conductive metal plates are commonly known as sacrificial electrodes. Electro coagulation process requires less space and does not require chemical storage, dilution and pH adjustment.

It is proven to be effective in water treatment such as drinking water supply for small or medium sized community\textsuperscript{145}. Electro coagulation process has been widely studied in water and wastewater treatment to remove fluoride\textsuperscript{146-148}. The fluoride in the electro coagulation process can be distributed into three parts, i.e. remained in water, adsorbed by flocs generated and formed in situ, and removed by the gelatinous layer attached on the electrodes.

Limitations

1. The process is expensive and complicated.

Adsorption Process:

The adsorption process of fluoride on to the surface of adsorbent may be either physical or chemical depending on the nature of forces involved. Theoretically, the adsorption of fluoride ion takes by three essential steps\textsuperscript{149-151}.

1. Diffusion or transport of fluoride ions to the external surface of the adsorbent from bulk solution across the boundary layer surrounding the adsorbent particle.
2. Adsorption of fluoride ions on to solid adsorbent surfaces.
3. The adsorbed fluoride ions probably exchange with the structural elements inside adsorbent particles depending on the chemistry of solids, or the adsorbed fluoride ions are transferred to the internal surfaces for porous materials (intra particle diffusion).

However, in presence of interfering ions like chloride, nitrate, sulphate, phosphate and carbonate in water results in ionic competition and impairs the efficiency of the fluoride removal system. To select an adsorbent for de-fluoridation process, we must consider the adsorption capacity of an adsorbent in dilute solutions, dosage, pH, temperature, and time required for fluoride removal, loading capacity in presence of interfering ions and finally the overall cost for de-fluoridation. A large number of effective, low-cost adsorbents such as activated alumina\textsuperscript{152}, amorphous alumina\textsuperscript{153}, calcite\textsuperscript{75}, rare earth oxides\textsuperscript{69}, bleaching earth\textsuperscript{154}, fly ash\textsuperscript{155}, limestone\textsuperscript{156}, clay minerals\textsuperscript{28}, bone char\textsuperscript{157}, heat-activated bauxite\textsuperscript{158}, natural soil\textsuperscript{159}, red mud\textsuperscript{160}, magnesium incorporated bentonite clay\textsuperscript{161}, Titanium rich bauxite\textsuperscript{111}, Brick powder\textsuperscript{46}, hydrated cement\textsuperscript{162}, Pumice\textsuperscript{163}, Acid activated kaolinite clay\textsuperscript{164}, iron(III)-aluminum (III) mixed oxide\textsuperscript{165} and some other low-
cost adsorbents $^{28,151,166}$ have been tested for fluoride removal. It is interesting to note that some adsorbents are capable to remove fluoride from water at a low concentration of 2 mg/lit$^{167}$ and some of them $^{155,168-169}$ are effective only at extreme pH (pH $\leq 3$) and, hence, may not be suitable in natural conditions.

A variety of activated carbon adsorbents are prepared from domestic, agricultural wastes and plant materials and high fluoride removal capacities of these activated carbons have been reported$^{170}$. Some of the investigations reported in this aspect are: fishbone charcoal$^{171}$, activated coconut shell carbon$^{172}$, coconut coir pit carbon$^{173}$, burnt clay$^{174}$, activated carbon from agriculture-waste$^{175}$, water hyacinth$^{176}$, Delonix regia tree dry fruit carbon$^{177}$, neem leaves carbon (Azadiractaindica), papal leaves (Ficus religiosa), khair leaves carbon (Acacia catechu willd)$^{166}$, KMnO$_4$-modified activated carbon derived from steam pyrolysis of rice straw$^{178}$, mixtures of activated carbon prepared from Neem (Azadirachtaindica) and Kikar (Acacia arabica) leaves$^{179}$, Morrinia Indica-based activated carbon$^{180}$, Zirconium impregnated coconut shell carbon$^{181}$, Kaza’s carbons$^{182}$, dolichos lablab carbon$^{183}$, Activated carbons prepared from the peels of Citrusdocumana, Citrus medica and Citrus aurantifolia fruits$^{184}$, Zirconium impregnated cashew nut shell carbon$^{185}$, Zirconium impregnated ground nut shell carbon$^{186}$, cyanodon dactylon based thermally activated carbon$^{187}$, Phyllanthus emblica activated carbon$^{188}$, Pine wood and Pine bark chars$^{189}$, Acacia Farnesiana Carbon$^{190}$, Lagenaria siceraria shell carbon$^{191}$, Typha Angustata activated carbon$^{192}$, Wheat Straw, Sawdust and activated bagasse carbon of Sugarcane$^{193}$ etc.

From all the above discussions, it has been observed that so many techniques are available for removal of excess fluoride from the drinking water but every technique has both advantages and disadvantages. So, a single method cannot be accepted properly for all areas. Hence, according to the conditions like area, concentration, availability of resources etc. any one method can be selected for removal of excess fluoride from the drinking water. But out of all the above said techniques, for de-fluoridation of water, adsorption techniques have more advantages because of their greater accessibility; do not require complicated hardware, inexpensive and capable of removing fluoride from water up to maximum extent$^{194-196}$.

**Conclusion**

Fluoride in drinking water causes beneficial or harmful effects depending on its concentration and the total amount ingested. The World Health Organization has specified the maximum permissible limit of fluoride concentration of drinking water as 1.5 mg/lit. Excessive fluoride levels cause fluorosis and other related diseases in human beings. Hence, a review on fluoride removal from fluoride contaminated water has been presented. Various de-fluoridation techniques such as precipitation, ion-exchange, reverse osmosis, electro dialysis, donnan dialysis, nanofiltration, electro coagulation and adsorption techniques have been reviewed and every method has their merits and pitfalls. Out of all the methods, the adsorption technique is inexpensive, efficient, ease of operation and produces high-quality water. Adsorption which is a conventional technique deals with various adsorbents such as: alumina/aluminium based materials, clays and soils, calcium based minerals, bone, bone char, synthetic compounds and carbon based materials. Thus we can conclude that the studies in the field of fluoride remediation have been reviewed effectively based on adsorption and the sorbents derived from natural sources is increasingly probed and due to the encouraging results, much research is endeavored to be devoted as these methods are based on eco-friendly and easily available adsorbents.

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