International Journal of ChemTech Research
CODEN (USA): IJCRGG ISSN: 0974-4290
Vol.8, No.8, pp 295-309, 2015

Studies on Defluoridation Techniques: A Critical Review

M. Suneetha[1], B. Syama Sundar[1] and K. Ravindhranath*[2]

[1]Department of Chemistry, Acharya Nagarjuna University, Guntur, India
[2]Dept. of Chemistry, KL University, Vaddeswaram, Guntur Dt., A.P., India

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 India[1-6]. 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 micas[7]. 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 elements[8]. Fluorides are found in a wide variety of minerals, including fluorspar [CaF_2], sellaite [MgF_2], rock phosphate/fluoroapatite [$Ca_5(PO_4)_3F$], cryolite [Na_3AlF_6], mica, hornblende and others[9]. Fluorite (CaF_2) is a common bearer of fluoride and is found in granite, granite gneisses and pegmatite[10-11] 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[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[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[19]. In India in 2002, 17 states have been identified as endemic for fluorosis[20] and now the problem exist in more than 17 states[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[24]. In India alone, a total of 60-70 million people including children are affected with dental and skeletal fluorosis[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 [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[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[31].

Table 1: Permissible limit of fluoride in drinking water prescribed by various Organizations

S. No.	Name of Organization	Permissible limit of fluoride ion (mg/lit)
1	World Health Organization (International standard for drinking water)	0.6-1.5
2	US Public Health Standards	0.8
3	The committee on public health engineering manual and Code of practice,Government of India.	1.0
	Indian Council of Medical Research (ICMR)	1.0
5	Bureau of Indian Standards (BIS)	0.6-1.5

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[32] at which fluorosis is low and this is being followed in most of the nations and is also the Australian recommended limit [33-34]. The effect of fluoride on human health by the prolonged use of drinking water with fluoride content[35-36] is presented in Table 2.

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

Fluoride ion concentration,(mg/lit)	Health outcome
<0.5	Dental caries
0.5–1.5	Optimum dental health
1.5–4.0	Dental fluorosis
4.0–10	Dental and skeletal fluorosis
>10.0	Crippling fluorosis

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 [27, 36-40] and also other adverse affects on human health have been reviewed[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[12, 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, $Al_2 (SO_4)_3 18H_2O$, 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 Al^{3+}- 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/1it to 0.2 mg/1it. 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[87]. Bone char produced by carbonizing bone at temperature of 1100-1600ºC 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[88]. It has been utilized successfully in the United States for municipal water de-fluoridation and sugar refining[89]. 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[90]. 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[91].
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[92-94]. The de-fluoridation of water by a chloride-loaded anion exchanger Amberlite IRA-400 was studied[95]. 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[96].

Activated Alumina:

UNICEF has sponsored research and development on the use of activated alumina for removal of fluoride from water in rural India[74]. Patil and Kulkarni[97]studied the ion exchange resin and activated alumina as defluoridating media. Activated alumina is a granular, highly porous material consisting essentially of aluminium oxide (Al_2O_3). 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[98]. Many reports are available for the de-fluoridation of water on large scale installations[99-100]. In rural as well as urban water treatment, activated alumina is used as an adsorbent[101-102].

A wide array of adsorbents has been used for de-fluoridation which includes activated and impregnated alumina[70, 73, 75, 103-120]. Alumina composites have been examined as de-fluoridation adsorbents. These include alumina cement granule[121-122], manganese oxide coated alumina[123], magnesia-amended activated alumina[124], hydrous manganese oxide-coated alumina[125], and Manganese dioxide-coated activated alumina[126]. Electro activated alumina has also been used for defluoridination of water[127]. The kinetics of removal of fluoride from groundwater has been studied by using activated alumina[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[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[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/1it. Few authors have studied reverse osmosis technique for purification of water[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[136]and electro-dialysis is effective in de-fluoridation of waters having even 3000 mg/lit of total dissolved salts and 3mg/lit fluoride[137]. Electro-dialysis can reduce the TDS and fluoride from 5000 mg/lit to 600 mg/lit and 15 mg/lit to 1.5 mg/lit 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[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[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[60, 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[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[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[145]. Electro coagulation process has been widely studied in water and wastewater treatment to remove fluoride[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 [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[152],amorphous alumina [153],calcite[75], rare earth oxides[69],bleaching earth[154],fly ash[155],limestone[156],clay minerals[28] ,bone char[157],heat-activated bauxite[158],natural soil[159], red mud[12, 160] ,Magnesium incorporated bentonite clay[161],Titanium rich bauxite[111],Brick powder[46] ,Hydrated cement[162],Pumice[163],Acid activated kaolinite clay[164],iron(III)-aluminum (III) mixed oxide[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 [153,168-169]are effective only at extreme pH (pH ≤ 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], Delonixregia tree dry fruit carbon[177], neem leafs carbon (Azadiraktaindica), papal leaves (Ficus religiosa), khair leafs 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], Morringa 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],cynodon 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 it 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.

References

1. Akoijam, Distribution of fluoride in ground water and pollution of shallow aquifers in parts of Imphal valley, Manipur, India, Chem. Environ. Research, 1997, 6, 301.
2. Mulligan C.N., Yong R.N. and Gibbs B.F., Remediation technologies for metal contaminated soils and groundwater: an evaluation, Eng. Geol., 2001, 60 (1-4), 193-200.
3. Charles F.H., Swartz C.H., Badruzzaman A.B.M., Nicole K.B., Yu W., Ali A., Jay J., Beckie R., Niedan V. and Brabander D., Groundwater arsenic contamination on the Ganges Delta: biogeochemistry, hydrology, human perturbations and human suffering on a large scale, Comptes Rendus Geosci., 2005, 337(1/2), 285-296.
4. Singh A.K., Chemistry of arsenic in ground water of Ganges-Brahmputra river basin, India, Curr. Sci., 2006, 91(5), 1-7.

5. Suthar S., Bishnoi P., Singh S., Mutiyar P.K., Nema A.K. and Patil N.S., Nitrate contamination in groundwater of some rural areas of Rajasthan, India, J Hazard Mater., 2009,171(1-3), 189-99.
6. Agrawal, A., Ravi S. P., and Sharma B., Water Pollution with Special Reference to Pesticide Contamination in India, J. Water Resource and Protection, 2010, 2, 432-448.
7. Hem J. D., Study and Interpretation of the Chemical Characteristics of Natural Water (2^{nd} ed.), U.S. Geological Survey, Washington, DC, USA, 1970, 363.
8. Hem J.D., Study and Interpretation of the Chemical Characteristics of Natural Water (3^{rd} ed.), US Geological Survey, Washington, DC, USA, 1989, 263.
9. Murray J.J., Appropriate Use of Fluorides for Human Health, WHO, Geneva, 1986.
10. Deshmukh A.N., Shah K.C. and Sriram A., Coal Ash: A Source of Fluoride Pollution, A Case Study of Koradi Thermal Power Station, District Nagpur, Maharashtra, Gondwana Geological Magazine, 1995, 9, 21-29.
11. Rao S., Fluoride in groundwater, Varaha River Basin, Visakhapatnam District, Andhra Pradesh, India, Environ. Monitoring & Assessment, 2009, 152, 47-60.
12. Cengeloglu Y., Kir E. and Ersoz M., Removal of Fluoride from aqueous solution by using red mud, Separation and Purification Technology, 2002, 28(1) 81-86.
13. Handa B.K., Geochemistry and genesis of fluoride containing ground waters in India, Groundwater, 1975, 3(3), 275-281.
14. Rao N.S., The occurrence and behaviour of fluoride in the groundwater of the Lower Vamsadhara river basin, India, Hydrological Sciences Journal, 1997, 42, 877-892.
15. Saxena V. K. and Ahmed S., Dissolution of fluoride in groundwater: a water-rock interaction study, Environ. Geol., 2001, 40, 1084-1087.
16. Edmunds W.M. and Smedley P.L., Fluoride in natural waters, Essentials of Medical Geology, Elsevier Academic Press, London, 2005, 301-329.
17. Chae G.T., Yun S.T., Kwon M.J., Kim S.Y. and Mayer B., Batch dissolution of granite and biotite in water: implication for fluorine geochemistry in groundwater, Geochemical Journal, 2006, 40, 95-102.
18. Guo Q., Wang Y., Ma T. and Ma R., Geochemical processes controlling the elevated fluoride concentrations in groundwater of the Taiyuan Basin, Northern China, Journal of Geochemical Exploration, 2007, 93, 1-12.
19. Teotia S.P.S. and Teotia M., Endemic fluorosis in India: A challenging national health problem, Journal of Association of Physicians of India, 1984, 32, 347-352.
20. Choubisa S.L., Endemic fluorosis in Southern Rajasthan, India, Fluoride, 2001, 34, 61-70.
21. Kumaran P., Bhargava G.N. and Bhakuni T.S., Fluorides in groundwater and endemic fluorosis in Raajasthan, Indian J. Environ. Health, 1971, 13, 316-324.
22. Teotia S.P.S., Teotia M., Singh D.P., Rathour R.S., Singh C.V., Tomar N.P.S., Nath M. and Singh N.P., Endemic Fluorosis: change to deeper bore wells as a practical community-acceptable approach to its eradication, Fluoride, 1984, 17, 48-52.
23. Agrawal V., Vaish A.K. and Vaish P., Groundwater quality: Focus on fluoride and fluorosis in Rajasthan, Current Science, 1997, 73(9), 743-746.
24. WRSD, Water Resources Systems Division, National Institute of Hydrology, Jalvigyan Bhawan, Roorkee, India.
25. UNICEF, State of the art report on the extent of fluoride in drinking water and the resulting endemicity in India, Report by Fluorosis Research & Rural Development Foundation for UNICEF, New Delhi, 1999.
26. Susheela A.K., Fluorosis: Indian scienario, a treatise on fluorosis, Fluorosis Research and Rural Development Foundation, New Delhi, India, Fluoride, 2001, 34, 181-183.
27. Ayoob S. and Gupta, A. K., Fluoride in drinking water: A review on the status and tress effects, Critical Reviews in Environmental Science and Technology, 2006, 36(6), 433-487.
28. Srimurali M., Pragathi A. and Karthikeyan J. A., A study on removal of fluoride from drinking water by adsorption onto low cost materials, J. Envir. Pollut., 1998, 99(2), 285-89.
29. Rahmani B. M., Mahvi, A. H., Dobaradaran, S., Hosseini, S. S., Evaluating the effectiveness of a hybrid sorbent resin in removing fluoride from water, Int. J. Environ. Sci. Tech., 2009, 6(4), 629-632.

30. Gopalkrishnan, S., Narsimha, S. and Seenipandian, C., Incidence of fluorosis in ten village near Tuticorian-suggestions for prevention and control, Indian journal of Environmental Protection, 1991, 11(2), 118-123.
31. WHO, World Health Organization, Guidelines for drinking water quality, Drinking water quality control in small community supplies, Geneva, 1985, 8,121.
32. WHO, World Health Organization, Guidelines for drinking water quality, Health criteria and other supporting information, Geneva, 1984.
33. Smet J., Fluoride in drinking water, Endemic Fluorosis in Developing Countries-Causes, Effects and Possible Solution: Report of a Symposium Held in Delft, The Netherlands, Netherlands Organization for Applied Scientific Research, 1990.
34. NHMRC, National Health and Medical Research Council. Australian Drinking Water Guidelines, 2004.
35. Dissanayake C.B., The fluoride problem in the groundwater of Sri Lanka-environmental management and health, Int. J. Environ. Stud., 199119, 195-203.
36. Edmunds W.M. and Smedley P.L., Groundwater geochemistry and health: an overview, Environmental Geochemistry and Health, Geol. Soci. Speci. Publication, 1996, 113, 91-105.
37. Savinelli E.A. and Black A.P., De-fluoridation of water with activated alumina, J. American Water Works Association, 1958, 50 (1), 33-44.
38. Bishop P. L. and George S., Fluoride removal from drinking water by Fluidized activated alumina, J. American Water Works Association, 1978, 70, 10, 55.
39. Susheela A.K., Fluorosis Types: Skeletal Fluorosis. Treatise on Fluorosis, (3rd ed.), Fluorosis Research & Rural Development Foundation of India, Delhi, 2007, 30-8.
40. Srimurali M., Karthikeyan G., De-fluoridation of water and household application-A Study, Proceeding of the Twelfth International Water Technology Conference, Alexandria, Egypt, 2008, 153-163.
41. Bailey K., Chilton J., Dahi E., Lennon M., Jackson P. and Fawell J., Fluoride in Drinking Water, World Health Organization, IWA Publishing, London, 2006.
42. Ganvir V. and Das K., Removal of Fluoride from drinking water using Aluminum Hydroxide coated rice husk ash, J. of Hazardous Materials, 2011, 185(2-3), 1287-1294.
43. Nawlakhe W. G., Kulkami D. N., Pathak B. N. and Bulusu K. R., De-fluoridation of water by Nalgonda technique, Indian Journal of Environmental Health, 1975,17, 26-65.
44. Saha S., Treatment of aqueous effluent for fluoride removal, Water Res., 1993, 27, 1347-50.
45. Reardon E.J. and Wang Y., A limestone reactor for fluoride removal from waste waters, Environmental Science & Technology, 2000, 34, 3247-3253.
46. Yadav A.K., Kaushik C. P., Haritash A. K., Kansal A. and Neetu R., De-fluoridation of drinking water using brick powder as an adsorbent, J. of Haz. Materials, 2006,128, 289-293.
47. Aldaco R., Garea A. and Irabien A., Calcium fluoride recovery from fluoride waste water in a fluidized bed reactor, Water Research, 2007, 41, 810-818.
48. Akbar E., Maurice S. O., Aoyi O. and Shigeo A., Removal of fluoride ions from aqueous solution at low pH using schwertmannit, J. of Haz. Mat., 2008, 152, 571-579.
49. Apambire W. B., Boyle D.R. and Michel F.A., Geochemistry, genesis and health implications of fluoriferous groundwaters in the upper regions of Ghana, Environ. Geol., 1997, 33(1), 13-24.
50. Singh G., Kumar B., Sen P.K. and Maunder J., Removal of fluoride from spent pot liner leachate using ion exchange, Water Environment Research, 1999, 71, 36-42.
51. Castel C., Schweizer M., Simonnot M. O., and Sardin M., Selective removal of fluoride ions by a two-way ion-exchange cyclic processes, Chem. Eng. Science, 2000, 55, 3341-3352.
52. Feng Shen X., Chen P. G. and Guohua C., Electrochemical removal of fluoride ions from industrial wastewater, Chemical Engineering Science, 2003, 58, 987-993.
53. Chubar N.I., Samanidou V.F., Kouts V.S., Gallios G.G., Kanibolotsky V.A., Strelko, V.V. and Zhuravlev I.Z., Adsorption of fluoride, chloride, bromide and bromate ions on a novel ion exchange, Journal of Colloid and Interface Science, 2005, 291, 67-74.
54. Meenakshi S. and Viswanathan N., Identification of selective ion-exchange resin for fluoride sorption, Journal of Colloid and Interface Science, 2007, 308, 438-450.
55. Simons R., Trace element removal from ash dam waters by nanofiltration and diffusion dialysis, Desalination, 1993, 89, 325-341.

56. Shen W., Li Z. and Liu Y., Surface Chemical Functional Groups Modification of Porous Carbon, Recent Pat. Chem. Eng., 2008, 1(1), 27-40.
57. Garmes H., Persin F. and Sandeaux J., De-fluoridation of groundwater by a hybrid process combining adsorption and Donnan dialysis, Desalination, 2002,145, 287-291.
58. Tor A., Removal of fluoride from water using anion-exchange membrane under Donnan dialysis condition, J. Hazard. Mater., 2007, 141**,** 814-818.
59. Adhikary S.K., Tipnis U.K., Harkare W.P. and Govindan K.P., De-fluoridation during desalination of brackish water by electrodialysis, Desalination, 1989, 71, 301-312.
60. Hichour M., Persin F., Sandeaux J. and Gavach C., Water de-fluoridation by Donann Dialysis and electro dialysis, Separation & Purification Technology, 2000, 18, 1-11.
61. Amer Z., Bariou B., Mameri N., Taky M., Nicolas S. and Elimidaoui A., Fluoride removal from brakish water by electro dialysis,Desalination, 2001, 133, 215-223.
62. Lahnida S., Tahaikta M., Elarouia K., Idrissia I., Hafsi M., Laaziz I., Amora Z., Tiyala F. and Elmidaoui A., Economic evaluation of fluoride removal by electrodialysis, Desalination, 2008, 230, 213-219.
63. Liu J., Xu Z., Li X., Zhang Y., Zhou Y., Wang Z. and Wang X., An Improved process to prepare high separation performance PA/PVDF hollow fiber composite nano filtration membranes. Separation & Purification Technol., 2007, 58, 53-60.
64. Dieye A., Larchet C., Auclair B. and Mar-Diop C., Elimination des fluorures parla dialyse ionicque croisee, European Polymer Journal, 1998, 34, 67-75.
65. Lhassani A., Rumeau M., Benjelloun D. and Pontie M., Selective demineralization of water by nanofiltration application to the de-fluoridation of brackish water, Water Research, 2001, 35, 3260-3264.
66. Mameri N., Yeddou A.R., Lounici H., Belhocine D., Grib H. and Bariou B., De-fluoridation of septentrional Sahara water of North Africa by electrocoagulation process using bipolar aluminium electrodes, Water Res., 1998, 32, 1604-1612.
67. Mjengera H. and Mkongo G., Appropriate de-fluoridation technology for use in fluorotic areas in Tanzania, Physics and Chemistry of the Earth, 2003, 28, 1097-1104.
68. Hu C.Y., Lo S.L. and Kuan W.H., Effect of co-exiting anions on fluoride removal in electrocoagulation process using aluminium electrodes, Water Res ., 2003,37, 4513-4523.
69. Raichur A.M. and Basu M.J., Adsorption of fluoride onto mixed rare earth Oxides, Sep. Purif. Technol., 2001, 24, 121-127.
70. Mohapatra D., Mishra D., Mishra S.P., Chaudhury G.R. and Das R.P., Use of oxide minerals to abate fluoride from water, J. Colloid Interface Sci., 2004, 275, 355-359.
71. Onyango M.S., Matsuda H. and Alain T., Chapter 1: Fluoride removal from water using adsorption technique, Adv. Fluorine Sci., 2006, 2, 1.
72. Shihabudheen M.M., Atul K.S. and Ligy P., Manganese-oxide-coated alumina: a promising sorbent for defluoridation of water, Water Res., 2006, 40, 3497-3506.
73. Tripathy S.S., Bersillon J.L. and Gopal K., Removal of fluoride from drinking water by adsorption on alum-impregnated activated alumina, Sep. Puri. Tech., 2006, 50(3), 310-17.
74. Mariappan P. and Vasudevan T., Domestic de-fluoridation techniques and sector approach for fluorosis mitigation, 2011, 1-11.
75. Yang M., Hashimoto M.T., Hoshi N. and Myoga H., Fluoride removal in a fixed bed packed with granular calcite,Water Res., 1999, 33, 3395-3402.
76. Turner B.D., Binning P. and Stipp S.L.S., Fluoride removal by calcite: evidence for fluoride precipitation and surface adsorption, Envir. Sci. Technol., 2005, 39, 9561-9568.
77. Eckenfemder W. W. Jr., Industrial water pollution control, Mc Graw Hill International Publications, 3rd edition, Environmental Engg. Series, 2000.
78. Rao S. M. and Mamatha P., Water quality in sustainable water management, Current science, 2004, 87(7), 942-947.
79. Larsen M.J. and Pearce E.I.F., Partial de-fluoridation of drinking water using fluorapatite precipitation, Caries Res., 1992, 26, 22-28.
80. Potgeiter J.H., An experimental assessment of the efficiency of different de-fluoridation methods, Chem. SA, 1990, 317-318.

81. Bulusu K.R., Sundaresan B.B., Pathak B.N., Nawlakhe W.G., Kulkarni D.N. and Thergaonkar V.P., Fluorides in water, de-fluoridation methods and their limitations, J. Institute Engineers (India), Environmental Engineering Division, 1979, 60, 125-133.
82. Dahi E., Mtalo F., Njau B. and Bregnhj H., De-fluoridation using Nalgonda Technique in Tanzania, 22nd WEDC Conference, New Delhi, 1996.
83. NEERI, National Environmental Engineering Research Institute, De-fluoridation Technology mission on drinking water in villages and related water management, NEERI Manual, Nagpur, India, 1961.
84. Apparao B.V. and Kartikeyan G., Permissible limits of fluoride on in drinking water in India in rural environment, Ind. J. Environ. Protec., 1986, 6(3), 172-175.
85. Nayak P., Review aluminium: impacts and disease, Envtl. Res. Sec., 2002, 89, 101-115.
86. Meenakshi S. and Maheshwari R.C., Fluoride in drinking water and its removal, Microfiltration and ultrafiltration membranes in drinking water, AWA publication, J. Hazard. Mater., 2006, B137, 456-463.
87. De-fluoridation by bone char, Available from: URL: http://www.grad.cmu.ac.htm.
88. Fawell J., Bailey K., Chilton J., Dahi E., Fewtrell L. and Magara Y., World Health Organization, Fluoride in Drinking-water, IWA Publishing, London, UK, 2006.
89. Bregnhoj, E. D. A. H., Significance of Oxygen in Processing of Bone Char for De-fluoridation of Water, The 1st international workshop on Fluorosis Prevention and De-fluoridation of Water, Ngurdoto, Tanzania, The Int. Society for Fluoride Research, 1995.
90. Christoffersen J., Christoffersen M.R. and Larsen R., Regeneration of surface coating of bone char used for de-fluoridation of water, Water resources journal, 1991, 25, 223-229.
91. Larsen M.J. and Pearce E.I.F., De-fluoridation of Drinking Water by Boiling with Brushite and Calcite, Caries Research, 2002, 36, 341-346.
92. Thompson J. and Mc Garvey F.X., Ion-Exchange Treatment of Water Supplies, Journal American Water Works Association, 1953, 45, 145-152.
93. Suzuki S., Matsumoto T., Tsukada K. and Aizawa K., Antimetastatic Effect of N-acetyl Chitohexaose on Mouse Bearing Lewis Lung Carcinoma, Elsevier, London, 1989, 707-713.
94. Lopez A.V., Reyes J.L.B, Song S. and Urbina R.H., Temperature effect on the zeta potential and fluoride adsorption at the α-Al_2O_3/aqueous solution interface, J. of Colloid and Interface Science, 2006, 298(1), 1-5.
95. Chikuma M. and Nishimura M., Selective sorption of fluoride ion by anion exchange resin modified with alizarin fluorine blue-praseodymium (III) complex, Reac. Polym., 1990, 13,131-138.
96. Kumar S. and Gopal K., A Review on Fluorosis and its Preventive Strategies, Indian J. Environmental Protection, 2000, 20(6), 430-6.
97. Patil A.R. and Kulkarni B.M., Study of ion exchange regime and activated alumina as de-fluoridation media, Jour of Institution of Engineers, 1989, 3, 14-16.
98. Chikte A., Fluoride and Fluorosis: The Status of Research in South Africa, 2nd International Workshop on Fluorosis Prevention and De-fluoridation of Water, Nazreth, Ethiopia, The International Society For Fluoride Research, 1997.
99. Rubble F. W. and Dale R.D., The Removal of excess fluoride from drinking water by Activated Alumina, J. American Water Works Association, 1979, 71, 45.
100. Clifford, D.A., Ion exchange and inorganic adsorption in water quality and treatment, Water Quality and Treatment, 4th edition, ed. Pontius, FW, McGraw-Hill Publication, 1990, 522.
101. Bhatnagar A., Fluoride removal from water by adsorption-A Review, Chem. Eng. J., 2011, 171, 811-840.
102. Vivek Vardhan, C.M. and Karthikeyan, J., Removal of fluoride from water using low-cost materials, Fifteenth International Water Technology Conference, Alexandria, Egypt, 2011.
103. Wu Y.C. and Nitya A., Water De-fluoridation by Activated Alumina, J Am. Soc. of Civ. Eng., 1979, 105(2), 357-367.
104. Gopal R. and Ghosh, P.K., Fluoride in Drinking Water-Its Effects and Removal, Defence Laboratory Jodhpur, 1985, 35(1), 71-88.
105. Schoeman J. and Macleod H., The Effect of Particle Size and Interfering Ions on Fluoride Removal by Activated Alumina, Water SA, 1987, 13(4).
106. Wasay S.A., Tokunaga S. and Park S.W., Removal of hazardous anions from aqueous solutions by La (III)- and Y (III)-impregnated alumina, Sep. Sci. Tech., 1996, 31,1501-1514.

107. Karthikeyan G. Meenakshi S. and Apparel B.V., De-fluoridation Properties of Activated Alumina, 2nd International Workshop on Fluorosis Prevention and De-fluoridation of Water Edited by Eli Dahi & Joan Maj Nielsen, Nazreth, Ethiopia, 1997, 78-82.
108. Lounici H., Addour L., Belhocine D., Grib H., Nicolas S., Bariou B. and Mameri N., Study of a new technique for fluoride removal from water, Desalin., 1997,114,241-251.
109. Vaish A. K. and Vaish P., A Case Study of Fluorosis Mitigation in Duangarpur District, Rajasthan, 3rd International Workshop on Fluorosis Prevention and De-fluoridation of Water, WHO, Chiang Mai, 2000, 97-104.
110. Bahena J.L., Cabrera A.R., Valdivieso A.L. and Urbina R. H., Fluoride Adsorption onto α-Al_2O_3 and its Effect on the Zeta Potential at the Alumina-Aqueous Electrolyte Interface, Sep. Sci. and Technol., 2002, 3(8), 1973-1987.
111. De-fluoridation of drinking water using activated titanium rich bauxite, J. Colloid Interface Sci., 2005, 292(1), 1-10.
112. Ghorai S. and Pant K.K. (2005). Equilibrium, kinetics and breakthrough studies for adsorption of fluoride on activated alumina,Sep. Purif. Technol., 42(3), 265-271.
113. Chauhan V.S., Dwivedi P.K. and Iyengar L., Investigations on activated alumina based domestic de-fluoridation units, J. Hazard.Mater., 2007, 139(1), 103-107.
114. Nagendra Rao C.R. and Karthikeyan J., Adsorption of Fluoride by Gamma Alumina, Proceeding of Twelfth International Water Technology Conference, IWTC12, Alexandria, Egypt, 2008, 141-152.
115. Ramos R. L., Castillo N. A. M., Azuara A. J., Barron J. M., Rodriguez L. E. L., Rosales J. M. and Pina, A.A., Fluoride Removal From Water Solution by Adsorption on Activated Alumina Prepared from Pseudo Boehmite, J. Envir. Eng. and Manage, 2008, 18(5), 301-309.
116. Stewart, T. (2009). Removal of Fluoride from Drinking Water, Analysis of Alumina Base of Sorption, 3, 22-24.
117. George, S. Pandit, P. and Gupta, A.B. Residual Aluminium in Water Defluoridated Using Activated Alumina-Modeling and Simulation Studies, Water Res., 2010, 44(10), 3055-3064.
118. MacDonald P. L. H., Singer, G. and Jaffe, B. P.R., An Integrated Approach to Address Endemic Fluorosis in Jharkhand, J. Water Resource Prot., 2011, 3, 457-472.
119. Shrivastava A. K. and Sharma M., An Innovative Technique for Removal of Fluoride from Drinking Water, Sci. Rev. Chem. Commun., 2012, 2(2) 133-140.
120. Umlong M., Das B., Devi R. R., Borah K. Saikia L. B., Raul P., Banerjee S. and Singh L., De-fluoridation from aqueous solution using stone dust and activated alumina at a fixed ratio, Appl. Water Sci., 2012, 2(1), 29-36.
121. Ayoob S., Gupta A.K., Bhakat P.B. and Bhat V.T., Investigations on the kinetics and mechanisms of sorptive removal of fluoride from water using alumina cement granules, Chem. Eng. J., 2008, 140(1-3), 6-14.
122. Ayoob S. and Gupta, A.K., Performance evaluation of alumina cements granules in removing fluoride from natural and synthetic water, Chem. Eng. J, 2009, 150(2-3), 485-491.
123. Maliyekkal S.M., Sharma A.K. and Philip L., Manganese-oxide-coated alumina: A promising sorbent for de-fluoridation of water, Water Res., 2006, 40, 3497-3506.
124. Maliyekkal S.M., Shukla S., Philip L. and Nambi I.M., Enhanced fluoride removal from drinking water by magnesia-amended activated alumina granules, Chem. Eng. J., 2008, 140(1-3), 183-192.
125. Teng, Shao-Xiang, Wang, Shu-Guang, Gong, Wen-Xin, Liu, Xian-Wei, Gao and Bao-Yu., Removal of fluoride by hydrous manganese oxide-coated alumina: performance and mechanism, J. Hazard. Mater., 2009, 168, 1004-1011.
126. Tripathy S.S. and Raichur A.M., Abatement of fluoride from water using manganese dioxide-coated activated alumina, J. Hazard. Mater., 2008, 153, 1043-1051.
127. Lounici H., Belhocine D., Grib H., Drouiche M., Pauss A. and Mameri N., Fluoride removal with electro-activated alumina, Desalination, 2004, 161, 287-293.
128. Agrawal R., Margandan K., Mishra M.K., Singh K., Acharya R. and Sharma S., Fluoride removal using activated Alumina-A case study of Bhooma Chota Water, Southern Brazialian journal of Chemistry, 2013, 21(21), 61-67.
129. Riitta K. and Pertti, K., Combination of membrane technology and limestone filtration to control drinking water quality, Desalination, 2000, 131, 271-283.
130. Fox K.R., Removal of inorganic contaminants from drinking water by reverse osmosis, Municipal environmental research laboratory, U.S. Envir. Protec. Agency, Cincinnati, 1981.

131. Huxstep M.R., Inorganic contaminants removal from driking water by reverse osmosis, Office of research and development, U.S. Envir. Protec. Agency, Cincinnati, 1981.
132. Schneiter R.W. and Middlebrooks E.J., Arsenic and fluoride removal from groundwater by reverse osmosis, Environ. Int., 1983, 9, 289-291.
133. Fu P., Ruiz H., Lozier J., Thompson K. and Spangenberg C., A pilot study on groundwater natural organics removal by low-pressure membranes, Desalination, 1995, 102, 47-56.
134. Arora M., Maheshwari R.C., Jain S.K. and Gupta A., Use of membrane technology for potable water production, Desalination, 2004, 170, 105-112.
135. Ndiaye P.I., Moulin P., Dominguez L., Millet J.C. and Charbit F., Removal of fluoride from electronic industrial effluent by RO membrane separation, Desalination, 2005, 173, 25-32.
136. Annouar S., Mountadar M., Soufiane A., Elmidaoui A., Sahli M.A., Menkouchi A., De-fluoridation of underground water by adsorption on the chitosan and by electrodialysis. Desalination, 2004, 165, 437-438.
137. Sahli M.A., Annouar A., Tahaikt S., Mountadar M., Soufiane A. and Elmidaoui A., Fluoride removal for underground brackish water by adsorption on the natural chitosan and by electrodialysis, Desalination, 2007, 212, 37-45.
138. Donnan F. G., The theory of membrane equilibria, Chem. Reviews, 1924, 1(1), 73-90.
139. Hichour M., Persin F., Sandeaux J. and Molenat J., Fluoride removal from diluted solutions by Donnan dialysis with anion exchange membranes, Desalination, 1999a, 122, 53-62.
140. Hichour M., Persin F., SandeauxJ., Molenat J. and Gavach C., Water de-fluoridation by Donnan dialysis and electrodialysis, Rev. Sci. Eau., 1999b, 12, 671-686.
141. Harma V., Nanofiltration for water quality improvement in Finnish surface waterworks. Licentiate thesis (Finnish), Helsinki University of Technology, Finland, 1999.
142. Tahaikt M., El-Habbani R., Ait Haddou A., Achary I., Amor Z., Taky M., Alami A., Boughriba A., Hafsi M. and Elmidaoui, A., Fluoride removal from groundwater by nanofiltration, Desalination, 2007, 212(1), 46-53.
143. Imen B., Amine M., Bechir H., Influence of operating conditions on the retention of fluoride from water by nanofiltration, Desalination and Water Treat. 2011, 29(1-3), 39-46.
144. Hoinkis J., Freitag S.V., Caporgno M.P.,, Patzold C., Removal of nitrate and fluoride by nanofiltration-a comparative study, Desalination and Water Treat., 2011, 30(1-3), 278-288.
145. Chen G. H., Electrochemical technologies in wastewater treatment, Sep. Purif. Technol., 2004, 38, 11-41.
146. Li S.C., Electro-chemiclal method to remove fluoride from drinking water, Water Supply, 1985, 3, 177-186.
147. Mameri N., Yeddou A.R., Lounici H., Belhocine D., Grib H. and Bariou B., De-fluoridation of septentrional Sahara water of North Africa by electrocoagulation process using bipolar aluminium electrodes, Water Res., 1998, 32, 1604-1612.
148. Emamjomeh M.M. and Sivakumar M., An empirical model for de-fluoridation by batch monopolar electrocoagulation/flotation process, J. Hazard. Mater., 2006, 131, 118-125.
149. McCabe W.L., Smith, J.C. and Harriott P., Unit Operations of Chemical Engineering (4th Edn) McGraw-Hill, New York, 1985, 547-549.
150. McKay G., Guendi, M. and EI-Nasser, M., Adsorption model for the removal of acids dyes from effluent by bagasse pith using a simplified isotherm, Adsorpt. Sci. Technol., 1997, 15(10), 737-752.
151. Fan X., Parker D.J. and Smith M.D., Adsorption kinetics of fluoride on low cost materials, Water Res., 2003, 37, 4929-4937.
152. Ghorai S. and Pant K.K., Investigations on the column performance of fluoride adsorption by activated alumina in a fixed-bed, Chem Eng J., 2004, 98,165-173.
153. Li Y.H., Wang S., Cao A., ZhaoD., Zhang X., Xu C., Luan Z., Ruan D., Liang J., Wu D. and Wei B., Adsorption of fluoride from water by amorphous alumina supported on carbon nanotubes, Chem. Phys. Lett., 2001, 350(5-6), 412-416.
154. Mahramanlioglu M., Kizilcikli I. and Bicer I.O., Adsorption of fluoride from aqueous solution by acid treated spent bleaching earth, J. of Fluorine Chemistry, 2002, 115, 41-47.
155. Chaturvedi A.K., Yadava K.P., Pathak K.C. and Singh V.N., De-fluoridation of water by adsorption on fly ash, Water Air and Soil Pollut., 1990, 49, 51-60.
156. Nair S., Jallan G. and Pandey G.S, Studies on fluoride removal by limestone, Fluoride, 1994, 23, 35-36.

157. Ramos R.L., Utrilla J.R., Castillo N.A.M. and Polo, M. S., Kinetic modeling of fluoride adsorption from aqueous solution onto bone char, Chem. Eng. J., 2010, 158, 458-467.
158. Sujana M.G. and Anand S., Fluoride removal studies from contaminated ground water by using bauxite, Desalination, 2011, 267, 222-227.
159. Chidambaram S., Manikandan S., Ramanathan A.L., Prasanna M.V., Thivya C., Karmegam U., Thilagavathi R. and Rajkumar K., A study on the de-fluoridation in water by using natural soil, Appl. Water Sci., 2013, 3(4), 741-751.
160. Tor A., Removal of fluoride from aqueous solution by using montmoorillonite, Desalination, 2006, 201, 267-276.
161. Magnesium incorporated bentonite clay for de-fluoridation of drinking water, J. Hazard Mater., 2010, 180(1-3), 122-30.
162. Hydrated cement: a promising adsorbent for the removal of fluoride from aqueous solution, J. Haz. Mat., 2008, 154(1-3), 88-95.
163. Malakootian M., Fatehizadeh A., Yousefi N. and Moosazadeh M., Fluoride removal from aqueous solution by pumice: case study on Kuhbonan water, African Journal of Environmental Science and Technology, 2011, 5(4), 299-306.
164. Gogoi P.K. and Ruby B., Fluoride removal from water by adsorption on acid activated kaolinite clay, Indian Journal of Chemical Technology, 2008, 15, 500-503.
165. Biswas K., Saha S.K. and Ghosh U.C., Adsorption of fluoride from aqueous solution by a synthetic iron (III)-aluminum (III) mixed oxide, Ind. Eng. Chem. Res., 2007, 46, 5346-5356.
166. Jamode A., Sapkal V. and Jamode V., De-fluoridation of water using inexpensive adsorbents, J. Indian Inst. Sci., 2004, 84(5), 163-171.
167. Majima T. and Takatsuki H., Fluoride removal from smoke-washing wastewater by using CaF_2 separating method, Water Purif Liq. Wastes Treat., 1987, 28, 433.
168. McKee R.H. and Jhonston W.S., Removal of fluorides from drinking water, Ind. Eng. Chem., 1934, 26(8), 849-850.
169. Suzuki T., Nakamura A., Niinae M., Nakata H., Fujii H. and Tasaka Y., Immobilization of fluoride in artificially contaminated kaolinite by the addition of commercial-grade magnesium oxide, Chem. Eng. J., 2013, 233, 176-184.
170. Venkataraman K., Bull. Cent. Pub. Hlth. Engg. Res. Instt., 1960, 2, 77.
171. Bhargava D.S. and Killedar D.J., Batch studies of water de-fluoridation using fishbone charcoal, Wat Polln Control Fed, USA, 1991, 63,848-858.
172. Arulanantham A., Ram Krishna T.V. and Bala Subramanian N., Studies on fluoride removal by coconut shell carbon, Indian J. Env. Prot., 1992, 12, 531.
173. Dahiya S. and Kaur A., Studies on removal of fluoride by coconut coir pit carbon, Indian Journal of Environmental Protection, 1999, 19(11), 68-71.
174. Karthikeyan G., Muthulakshmi N., Saravana G., Sundar., De-fluoridation property of burnt clay, J. Indian Water Works Association, 1999, 31, 291.
175. Sivabalan R., Rengaraj S., Arabindoo B. and Murugesan V. Fluoride uptake characteristics of activated carbon from agriculture-waste, J. Sci. Indst. Res., 61, 1039-1045.
176. Uday Simha L., Panigrahy R.K. and Ramakrishna S.V., Prelimanary studies on fluoride adsorption by water hyacinth, I J E P., 2002, 22(5), 506-511.
177. Kishore M., Studies on Delonix Regia Tree Dry Fruit Carbon in de-fluoridation of potable water, Acharya Nagarjuna University, Andhrapradesh, India, 2004.
178. Daifullah A.A.M., Yakout S.M. and Elreefy S.A., Adsorption of fluoride in aqueous solutions using $KMnO_4$ modified activated carbon derived from steam pyrolysis of rice straw, Journal of Hazardous Materials, 2007, 147, 633-643.
179. Kumar S., Gupta A. and Yadav J.P., Fluoride removal by mixtures of activated carbon prepared from neem (Azadirachta indica) and kikar (Acacia arabica) leaves, Ind J Chem Tech., 2007, 14, 355-61.
180. Karthikeyan G. and Siva Elango S., Fluoride sorption using Morringa Indica-based activated carbon, Iran. J. Environ. Health. Sci. Eng., 2007, 4(1), 21-28.
181. Saisathish R., Raju N.S.R., Raju G.S., Nageswara Rao G., Anil Kumar K. and Janardhana, C., Equilibrium and Kinetic studies for Fluoride adsorption from water on Zirconium impregnated Coconut shell carbon, Seperation Science and Technology, 2007, 42, 769-788.
182. Somasekhararao K., Kaza's carbons are low cost adsorbents for de-fluoridation of potable water - a review, Asian J. Chem. Env. Res., 2008, 1(1), 1-12.

183. Rao V. B. M., Rao M.S., Prasanthi V. and Ravi M., Characterization and de-fluoridation studies of activated Dolichos Lab Lab carbon, Rasayan J. Chem., 2009, 2, (2), 525-530.
184. Chakrapani Ch., Suresh Ch. B., Vani K.N.K. and Rao K.S., Adsorption Kinetics for the removal of Fluoride from aqueous solution by activated carbon adsorbents derived from the peels of selected Citrus Fruits, E-J. of Chemistry, 2010, 7(S1), S419-S427.
185. Alagumuthu G. and Rajan M., Equilibrium and kinetics of adsorption of fluoride onto zirconium impregnated cashew nut shell carbon, Chem. Eng. J., 2010a, 158, 451-457.
186. Alagumuthu G. and Rajan M., Kinetic and equilibrium studies on fluoride removal by zirconium (IV)-impregnated ground nutshell carbon, Hem. Ind., 2010b, 64 (4), 295-304.
187. Alagumuthu G., Veeraputhiran V. and Venkataraman R., Fluoride sorption using Cynodon dactylon based activated carbon, Hem. ind., 2011, 65(1), 23-35.
188. Alagumuthu G. and Veeraputhiran V., Sorption equilibrium of fluoride onto Phyllanthus emblica activated carbon, Int. J. Res. Chem. Environ., 2011, 1(1), 42-47.
189. Dinesh M., Rupa S., Singh, V.K., Pittman Jr. U., Fluoride removal from water using bio-char, a green waste, low-cost adsorbent: Equilibrium uptake and sorption dynamics modeling, Ind. Eng. Chem. Res., 2012, 51, 900-914.
190. Hanumantharao Y., Kishore M. and Ravindhranath K., Characterization and de-fluoridation studies using activated Acacia Farnesiana carbon as adsorbent, Ele. J. Environ. Agricul. Food Chem., 2012b, 11 (5), 442-458.
191. Hanumantharao Y., Kishore M. and Ravindhranath K., Characterization and adsorption studies of Lagenaria siceraria shell carbon for the removal of fluoride, Inter. J. of Chem Tech Research., 2012c, 4(4), 1686-1700.
192. Hanumantharao Y., Kishore M. and Ravindhranath K., Characterization and de-fluoridation studies of active carbon derived from Typha Angustata Plants, J. Analytical Science & Technology, 2012d, 3(2), 167-181.
193. Yadav A.K., Abbassi R., Gupta A., and Mohammad D., Removal of Fluoride from aqueous solution and groundwater by wheat straw, sawdust and activated bagasse carbon of sugarcane source, Ecological engineering, 2013, 52, 211-218.
194. Nava C.D., Rios M. S. and Olguin M.T., Sorption of fluoride from aqueous solution and well drinking water by thermally heated hydrocalcite, Sep. Sci. Tech., 2003, 38(1), 131-147.
195. Rao S.G., A Study on the removal of fluoride from water using carbonized coconut coir, Institution of Public Health Engineers, India, 2003, 3, 5-81.
196. Gogoi P. K. and Baruah R., Fluoride removal from water by adsorption on acid activated Kaolinite Clay, Ind. J. Chem. Tech., 2008, 15, 500-503.

*****