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Removal of Fluoride from Water by Bioadsorbents: A Review

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Abstract: Fluoride is major inorganic pollutant present in groundwater. Fluoride is two edge sword.In small doses, it prevents tooth decay and in higher doses it causes fluorosis. Its permissible limit is 1.5 ppm.Due to high toxicity of fluoride, there is urgent need to remove fluoride.There are many techniques to remove fluoride but common technique used to remove fluoride is adsorption.This review article is aimed at providing information on various bioadsorbents used for removal of fluoride from water. Different types of biological materials have been used such as plant biomass, microbial biomass ,industrial and agricultural waste biomass for removal of fluoride from water.
Keywords: Fluoride, Adsorption, microbial,Langmuir, Freundlich.

I. INTRODUCTION

Fluorine is 13th most abundant element of Halogen group and is one of the most reactive of all chemical elements. It is also most electronegative element and has a strong tendency to acquire negative charge to form fluoride ion in solutions[1]. Naturally it is found in rocks, soil and fresh water. Weathering of fluoride containing rocks and soils leads to leaching of these ions from the soil into ground water. Further it also makes its place into ground water due to dissolution from minerals/rocks like topaz, fluorite, fluorspar, cryolite, fluorapatite etc[2]. Fluorides in the form of salts with monovalent cations i.e. NaF and KF are water soluble but salts of fluoride with divalent cations such as CaF_2 are insoluble in water.

The pathway of fluorides in natural aquatic streams is mainly dependent on the geological conditions of rocks such as decomposition, dissociation and subsequent dissolution with considerably longer retention times that lead to the leaching of fluoride ions in water bodies[3].

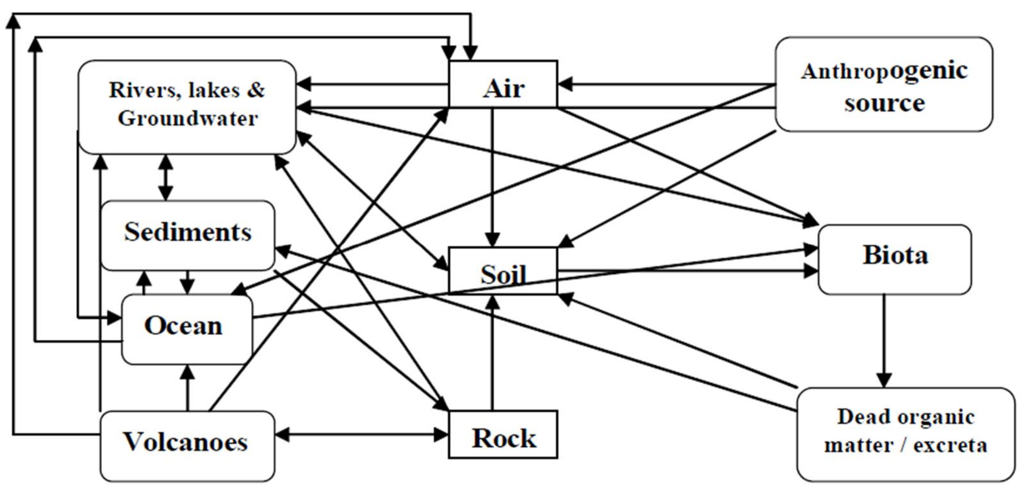

Figure1: Pathway of Fluoride Ions Accumulation in Natural Water Streams

Various industries such as glass and ceramic production, semiconductor manufacturing and electroplating further adds remarkably high amounts of fluorides in aqueous stream[3].Flouride ions also have a great tendency to get escape in the metabolism of living organisms. In industrial effluents specifically released from the processes of stainless steel pickling, herbicide production and glass itching, these ions exist in the form of weak and potentially toxic hydrofluoric acid having capacity to penetrate into tissues leading to everlasting damages. All these processes contribute to the excessive concentration of fluoride ions in nature and ultimately in

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food chain by one or other means. Excessive consumption of these ions has numerous profound health effects if intake is beyond permissible limits[4]. Many health problems such as dental fluorosis, skeletal fluorosis, poor thyroid functioning, neurotoxicity, reduced insulin secretions and noncarcinogenic effects on kidney are seen due to high concentrations of fluoride ions in metabolism [5]. Maximum permissible limits of fluoride ions defined by various health organizations.

Table1 : Permissible Limits of Fluoride ions in Potable Water Prescribed by Various Health Organizations

Name of the Health Organization	Permissible limits of fluoride ion (mg/l)
World Health Organization (International standard of drinking water)	0.6–1.5
US Public Health Standards	0.8
The committee on public health engineering manual and Code of practice, Government of India	1.0
Indian Council of Medical Research (ICMR)	1.0
Bureau of Indian Standards (BIS)	0.6–1.5

Different health effects of fluorides depending upon the intake concentrations are given in table.

Table 2: Different Fluoride Concentrations and Their Effects on Human Health

S. No.	Fluoride ion concentration (mg/l)	Effects on Human Health
1	Below 0.5	Dental Caries
2	0.5 to 1.5	Protection against dental caries. Takes care of bone and teeth.
3	1.5 to 3.0	Dental fluorosis
4	3 to 10	Skeletal fluorosis (adverse changes in bone structure)
5	10 or more	Cripping skeletal fluorosis and effects on other organs of body.
6	50	Thyroid malfunctioning
7	125	Kidney malfunctioning

1) *Bioadsorption:* Biosorption is a physiochemical process that occurs naturally in certain biomass which allows it to bind contaminants onto its cellulose structure. Bioadsorption technique has gained momentum in recent years due to high efficiency, stumpy cost and ease in operation and maintenance. This method has been found of significant utility to remove fluoride ions from aquatic water systems. As the process involves various kind of interactions such as ion-exchange, complexation, adsorption by physical force, precipitation and impingement in vacant voids it has been widely used by the researchers all over the world. Different types of biological materials were explored such as plant biomass, microbial biomass industrial as well as agricultural waste biomass.

A. *Microbial Adsorbents For Removal Of Fluoride*

Various Bacteria, algae and fungi moulds have been used to removal fluoride from water. Summary of some microbial bioadsorbents is given below:

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Sr.No.	Adsorbent	Optimized Conditions	Result	Reference
1	Anabaena fertilissima and chlorococcum humicola microalgal Biomass	pH=7.0	70% for Anabaena fertilissima and 30% for chlorococcum humicola	Monica Bhatnagar et. al. (2002) [6]
2	Algal Biosorbent (Spirogyra)	pH=7.0	65%	S. Venkata Mohan et. al. (2007) [7]
3	Aspergillus penicilloides fungal biomass and mucor racemosus fungal biomass	pH=7.0	30%	Rajneesh Prajapat et. al. (2010) [8]
4	Aspergillus nidulans fungal bioadsorbent	pH=4.0	36%	Ramchander Merugu et. al. (2013) [9]
5	Pleurotus eryngii fungus	pH=2.0	92%	Farah Amin et. al. (2014) [10]
6	Trichoderma harzianum fungal bioadsorbent	contact time of 60 min. and pH = 10	36% fluoride removal	Shalini khosle et. al. (2016) [11]
7	Chloerella vulgaries alga immobilized in Calcium Alginate beads	Flow rate of feed to column=5 ml/min, initial fluoride conc. = 10 mg/L	46%	Poornima G. Hiremath et al. (2017) [12]
8	Spriulina Platensis immobilized on calcium alginate beads	pH=5.2	82.65%	Sushma Kundari (2017) [13]

B. Agricultural Adsorbents For Removal Of Fluoride

Agricultural waste biomaterials are available in huge amount, biodegradable, inexpensive and environmentally friendly. This biomass material includes peels, seeds, shells, juices stems and leaves etc. Summary of some agricultural waste bioadsorbents have been used to remove fluoride from water is given below.

Summary of Various Researches conducted on removal of fluoride by using Agricultural Bioadsorbents

S.No.	Adsorbent	Optimized Conditions	Results of %age removal of fluoride	References
1.	Leaves of Neem (Azadirachta indica), Peepal (Ficus religiosa), Khair (Acacia catechu wild) Trees	The highest fluoride ion concentration (15 mg/l), the fluoride ion level in the effuent gradually decreased to 0 mg/l within 180 min. at 29 0.5°C when thedose of adsorbent is 10 g/l.	90%	A.V. Jamode et al. (2004) [14]
2.	Tamarind Seed	pH=7	90%	M. Murugan et al. (2006) [15]
3.	Alminium chloride and calcium chloride treated powdered corn cob	pH=5.0-6.5	85%	Parmar et al. (2006) [16]
4.	Thermany activated neem (Azardirachta indica) leaves carbon (ANC) and thermally activated kikar (Acacia arabica) leaves carbon (AKC)	The optimum pH was found to be 6 for both adsorbents. The optimum dose was found to be 0.5 g / 100 ml for ANC and 0.7 g / 100 ml for AKC. The optimum time was found to be one hour for both the adsorbent.	68%	Kumar et al. (2008) [17]
5.	Zirconium impregnated cashew nut shell carbon (ZICNSC)	pH=7 and contact time of 180 min	80.33%	Alagumuthu et al. (2010) [18]
6.	Zirconium impregnated ground nut shell carbon (ZIGNSC)	contact time of 180 min and fluoride saturation capacity is 1.26mg F^{-}/g at room temp.	92%	Ganpaty Alagumuthu et al. (2010) [19]
7.	Cynodon dactylon (Bermuda grass)	Contact time of 105 min	83.77%	Ganpaty Alagumuthu et al.

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		And pH =7		(2010) [20]
8.	Egg Shell Powder	Maximum adsorption at pH=2.0-6.0 and contact time of 120 min.	94%	R. Bhaumik et al. (2011) [21]
9.	Zirconium loaded Garlic peel	pH=2.0	97.2%	Kai et al. (2011)[22]
10.	Rice Husk	Fluoride removal from 5mg/l of Fluoride solution requiring an equilibrium time of 3 hours	83%	C.M. Vivek Vardhan et al. (2011) [23]
11.	Tea Leaves (after making of tea)	pH=6 and contact time of 150 min.	91%	S. Jenish et al. (2011) [24]
12.	Aluminium hydroxide coated Rice husk ash	pH=5.0	80%	Ganvir et al. (2011) [25]
13.	Betel Nut coir charcoal	Contact time of 180 min	92%	Sutapa Chakrabarty et al. (2011) [26]
14.	Acacia Farnesiana (Sweet Acacia) Carbonized material (AFC)	pH=6.9	70%	Hanumantharao et al. (2011) [27]
15.	Neem Charcoal Powder	pH=5 and contact time of 180 min.	94%	Sutapa Chakrabarty et al. 2012 [28]
16.	Tea Ash	pH=6 and contact time of 180 min.	83%	Naba Kumar Mondal et al. (2012) [29]
17.	Bark of Babool	pH=6-8 and contact time of 8 hours.	77.04%	Bhagyashree M Mamiwar et al. (2012) [30]
18.	Zirconyl-Impregnated Activated Carbon prepared from Lapsi Seed Stone	pH=3-4 and contact time of 180 min.	75%	Sahira Joshi et al. (2012) [31]
19.	Mangrove plant leaf powder (MPLP), Almond tree bark powder (ATBP), Pineapple Peel powder (PPP), Chiku leaf powder (CLP), Toor plant leaf powder (TPLP) and Coconutcoir pith (CCP)	The optimum contact time was 60 minute and the percentage removal at pH 2	Uptake of fluoride was in order MPLP > CCP > TPLP > > CLP > PPP > ATBP.	Patil Satish et al. (2012) [32]
20.	Guava Seeds	pH=5-8	75%	S.A. Valencia-Leal et al. (2012)[33]
21.	Fresh leaves, dry leaves and stem of Basil (Tulsi Leaves, Ocimum sanctum, Lamiaceae)	pH=9 for fresh basil leaves pH=6 for fresh basil stem pH=6 for dry leaves pH=7 for dry stem	The maximum removal of 94% (by fresh basil leaves), 75% (fresh basil stem), 78% (dry leaves) and 74% (dry stem) achieved from 5 ppm of fluoride solution	Kamble et al. (2012) [34]
22.	Activated carbon prepared from almond shell with KOH activation	pH=4	68%	Bhagawati et al. (2012) [35]
23.	Phosphoric acid activated vetiver root	pH=6	75%	Puthenveedu Sadavisan Pillai Harikumar et al. (2012) [36]
24.	Tamarind fruit shell	pH=2 and contact time of 90 min.	85%	V. Ramanjaneyulu

				et al. (2012) [37]
25.	Lagenaria siceraria shell carbon (LSSC)	pH=6	85%	Hanumantharao et al. (2012) [38]
26.	Activated bagasse carbon (ABC), Sawdust raw (SDR), and wheat straw raw (WSR)	pH=6 and contact time of 60 min	The CAC, ABC, SDR and WSR removed 57.6, 56.4, 49.8 and 40.2% respectively.	Ashish Kumar Yadav et al. (2013) [39]
27.	Zirconium impregnated walnut-shell carbon (ZIWSC)	pH=3	94%	M Rajan et al. (2013) [40]
28.	Guava leaf powder, Neem leaf powder, Neem bark powder, Black berry powder.	pH=7	----	Jain et al. (2013) [41]
29.	Khimp Plant Stem Powder	pH=6.7	97%	Shyam et al. (2013) [42]
30.	Timber of Aralu	pH=5	80%	Yadav et al. (2014) [43]
31.	Cissus Quadrangularis (CQ Powder)	pH=7 and contact time of 60 min	90%	Rayappan et al. (2014) [44]
32.	Date Palm Seeds	pH=7	94%	Mise and Gurani et al. (2014) [45]
33.	Citrus limonum (lemon) Leaf	pH=2	70%	V. Tomar et al. (2014) [46]
34.	Peepal leaves	pH=7 and contact time of 45 min	85.7%	Shubha Dwivedi et al. (2014) [47]
35.	Devdaru (Polyalthia longifolia) leaf powder DLP	pH=7	77%	Bharali and Bhatacharya et al. (2014) [48]
36.	Maize husk fly ash	pH=2 and contact time of 120 min and optimum adsorbent dose was found to be 2.0g/50ml	86%	A.S. Jadhav et al. (2014) [49]
37.	Silikha (Terminalia chebula) leaf powder	pH=6.8 and 303k temp.	74%	Bharali and Bhatacharya et al. (2014) [50]
38.	BananaPeel,groundnut shell and sweet lemon peel	Contact time of 1 hour	The banana peel, groundnut shell and sweet lemon peel removed 94.34, 89.9 and 59.59% respectively at doses of 14, 12 and 16 gm/l respectively	Aash Mohammad et al. (2014) [51]
39.	Bale Fruit Shell	pH=5 and contact time of 20 min	63%	G. Anusha et al. (2014) [52]
40.	Sawdust	pH=7 and contact time of 120 min.	70%	Suman Mann et al. (2014) [53]
41.	Activated cotton nut shell carbons	pH=7	80%	Rajan Mariappan et al. (2015) [54]
42.	Sal(Shorea Robusta) Leaf Powder	pH=7.5	63.6%	Kumari et al. (2015) [55]
43.	Chemically modified palm kernel Shell (CMPKS)	pH=6.0	75%	M.T. Bashir et al. (2015) [56]
44.	Aluminium impregnated coconut fiberash (AICFA)	pH=12	98%	Naba Kumar Mondal et al. (2015) [57]
45.	Carbonised Punica granatum Carbon (CPGC)	Contact time of 75 min	78.1%	Sudhanshu Kanaujia et al. (2015) [58]
46.	Coconut Husk	pH=5	78%	Islamudin et al. (2016) [59]

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47.	Bark of Phyllanthus Emblica (Amla)	pH=6-8	80%	R.N. Patil et al. (2016) [60]
48.	Sugar Cane Bagasse	Maximum removal of fluoride from the drinking water of 5 g/l dose at 323K temperature.	86% fluoride removal	Nusrat Ali et al. (2016) [61]
49.	Horse gram (Macrotyloma uniflorum) seed powder	Optimum dose and Optimum contact time for adsorption process is 0.8 gm and 20-30 min respectively.	80%	N. Gandhi et al. (2016) [62]
50.	Activated Bamboo Charcoal	pH=5-9	87.5%	Wendimu et al. (2017) [63]
51.	Activated carbon prepared from Bael Shell (ACBS)	contact time of 60 minutes, adosrbent dose of 2 g/l.	52% fluoride removal	Kalpana Singh et al. (2017) [64]
52.	Cabbage tree(Moringa Stenopetala)	contact time of 60 minutes, at pH=7	80% fluoride removal	Seid Tiku Mereta et al(2017) [65]

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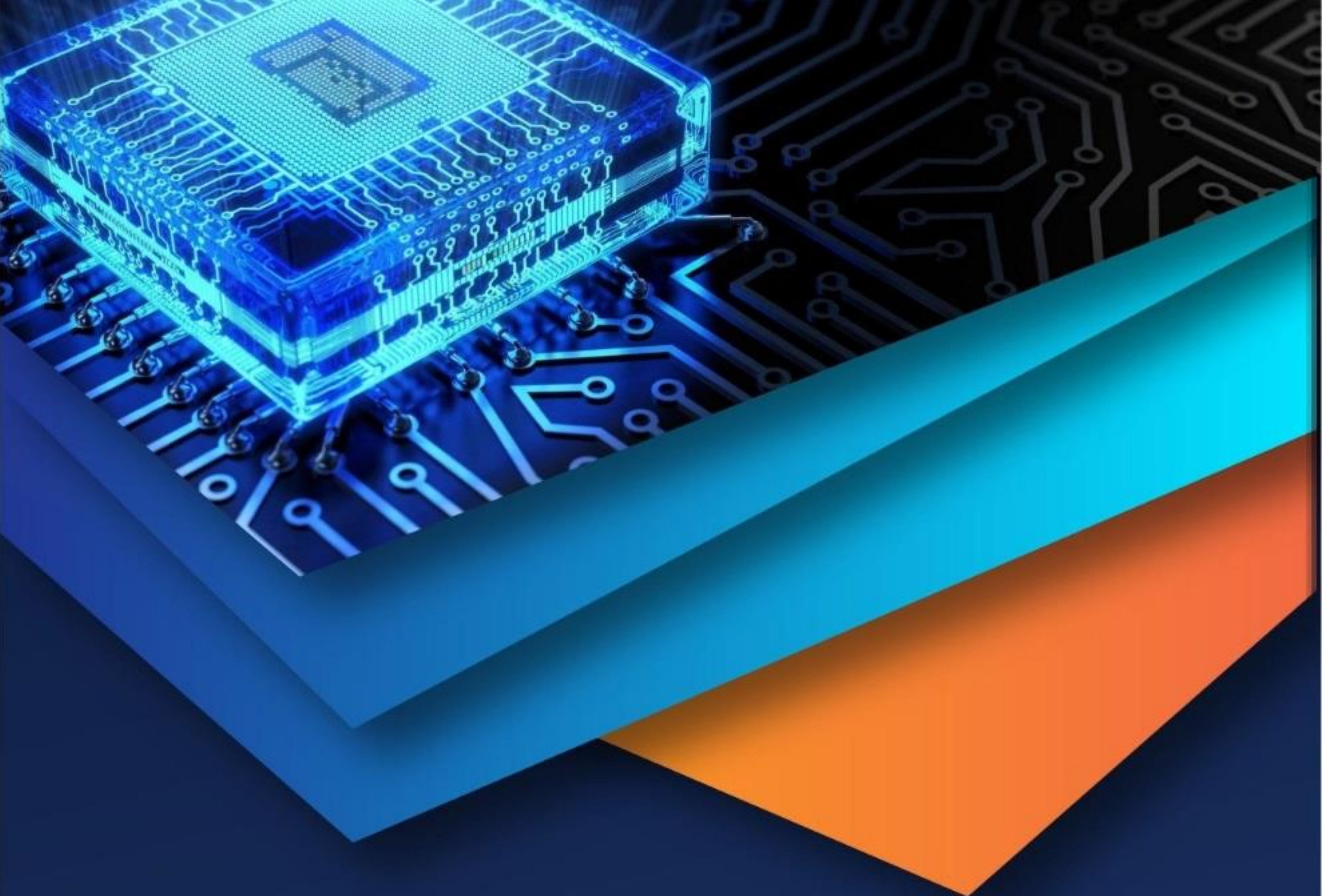

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