

"Mitigating Health Risks of Fluoride: Low-Cost Bio-Adsorbents for Effective Fluoride Removal from Water"- Review

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Abstract. In India has fluoride and fluorosis issues are common in majority of places. The fluoride epidemic is spread among 19 states in India. The serious health problem is occurs in human beings due to contaminated drinking water for natural and man-made action. Therefore, various treatments applied for removal of fluoride, such as namely Nalgonda method, contact precipitation, bone charcoal, clay column, ion-exchange, Reverse osmosis, activated alumina, and Mgo. This review paper is discussed various parameters such as contact time, temperature, concentration, initial fluoride, pH. From this review paper discussed various bio-adsorbents materials and also seen fluoride removal efficient.

Keywords: Fluoride, bio sorbents, toxicity

Introduction

The high level of fluoride concentrations are generally occur due to contaminated ground water and causes fluorosis problem [1-2]. It causes serious health problem due to presence of fluoride leaching underground water. According to the Bureau of Indian Standard (BIS) is suggested fluoride concentration in drinking water from 0.6 to 1.2 ppm [3]. The World health organization (WHO) is suggested fluoride concentration in drinking water from 0.6 to 1.5 ppm [4]. The fluoride is belonging to halogen family and widely spread in nature. Also fluoride is an important constituent in soils and rocks. Fluoride is lead to causes skeletal and non-skeletal fluorosis due to fluoride consumption. Generally fluoride element is damage tissue, ligaments and muscles [5]. Fluoride generally exists in rock minerals such as Fluor apatite, fluorspar, and mica etc., [6]. The presence of fluoride concentration in drinking water is due to physical and geological structure of aquifers [7].

The presence of small amount of calcium and alkaline concentration (pH 7.5-8.5) is responsible for suspension of fluoride from host rocks[8]. The bio-materials are a useful for cleaning impurity from drinking water that is till utilizing in countryside of Latin America. The purpose of bio-material has newly revived by water supply researcher and developed with more research rigor [9-10]. The fluoride removal has such as Nalgonda technique, electro-dialysis, Activated alumina, bone charcoal, contact precipitation, reverse osmosis, ion-exchange, clay column, bio-adsorbents [11-13]. India has a rich biodiversity in medicinal plants. The maximum number of study revealed that defluoridation capacities of the following plant materials-Ramacham (*Vetiveria zizanioides*), Tamarind seed (*Tamarindus indica*), Clove (*Eugenia carryophyllata*), Neem (*Azadirachta indica*), Acacia (*Acacia catechu willd*), Nutmeg (*Myristica fragarns*), and coffee

husk (*Coffea arabica*) [14]. According to world health organization said an estimated that 25 million people in 19 states were India and union territories have already been affected and another 66 million are at risk, including 6 million children below the age of 14 years [15]. In some parts of India, the fluoride levels are less 0.5 mg/l, whereas in other places, fluoride levels are as high as 35 mg/l [16]. Bio-sorption gives benefits of low operating cost, minimization of the capacity of chemical or biological sludge to be disposed, high efficiency in dilute effluents, no nutrient requirements and environmental sociability and financial feasibility. This provides a cost-effective solution for industrial wastewater management [17]. Bio-sorbents techniques are used for numerous bacterial sources, leaf-based adsorbents and water hyacinth has been reported by various investigators [18].

The low cost materials such as activated alumina, activated carbon that produce problem in regeneration. Bio adsorption technique is a cheap process and utilizes for removal of fluoride from contaminated drinking water through native bio-adsorbent materials. The biomass of bio-materials can be used for measure an effective removal of fluoride and resolving their sludge removal problem. The upcoming adaptableness and techno-economic feasibility are also check by their field performance of bio-adsorbent bed column.

FLUORIDE REMOVAL BY BIOSORBENTS

PLANTS:

S.Venkata Mohan et al, investigated fluoride contaminated water used waste fungal biomass (*Pleurotus ostreatus*) resulting from laccase fermentation method. Batch adsorption studies are accomplished and an effects visible that bio sorbent established capacity to fluoride adsorbed from aqueous solution. The adsorption process followed pseudo first order rate equation. Langmuir isotherm adsorption model well fitted with Isothermal model [19].

Vardhan and Karthikeyan et al., used rice husk and *Moringa Oleifera* (drum stick) was modified by MnCl_2 and MnSO_4 . The adsorbent could remove fluoride from contaminated water. Those adsorbent dependent on pH and also fluoride removal efficiency was observed to be greater at lower pH. The various parameters are studied such as initial fluoride concentration, contact time, effect of co-ions and adsorbent dose. The fluoride removal capacity was influenced by co-existing HCO_3^- and CO_3^{2-} ions. The fluoride adsorption is respect with variation of initial fluoride concentration, adsorbent dose, heat treatment and presence of interfering ions and pH were studied [20].

Maheswari et al have studied defluoridation from contaminated drinking water used newly prepared dry fruits collected from plants *Enterolobium Saman* (ESC), *Acacia Arabic* (AAC), *Prosopis Juliflora* (PLC) [21]. Ardekni et al have found removal of fluoride from water used Na_2CO_3 modified bagas and chitosan. The fluoride removal capacity decreases with initial concentration of fluoride solution but increases with increasing dosage of adsorbent and contact time [22]. Liang et al have studied using rare earth modified chitosan and found removal efficiency of fluoride. At pH 7.0, the adsorbent dose of 2g/lit., at 60 minutes equilibrium time that maximum fluoride adsorption reached [23].

S. Venkata Mohan et al, have been examined removal of fluoride used algal bio sorbent (*Spirogyra IO2*). From the batch study result found that bio sorbent established to remove of

fluoride. The fluoride removal was affecting various parameters such as fluoride concentration, the dosage of adsorbent, the pH of aqueous solution, the temperature on removal of fluoride. The interaction of bio-sorption of algal species showed fluoride removal tendency that followed pseudo first order rate equation. From experimental data fitted with Langmuir's adsorption isotherm model. [24].

Pandey et al. have developed fluoride removal capacity used plant of *Tinospora cordifolia* powder 1.18 μm particle size after washed and dried in the sun and hot air oven at 60°C. At pH 7.0, the initial dose of adsorbent 7g/ml, the maximum fluoride removal 70% attained of starting time 120 minutes. The experimental data revealed that Freundlich isotherm, Langmuir models and Pseudo first order kinetic model fitted well as compared to second order kinetic model [25].

Rao et al. have succeeded defluoridation of drinking water used activated *Dolichos Lablab* carbon found by pyrolysis of dry parts of *dolichos* in electric furnace at 600°C for 4 hours then washed and dehydrated and subsequently altered by 0.1M of HNO_3 . The *Dolichos Lablab* carbon has fluoride removal efficiency 83.6% at contact time 30 minutes with 5 ppm initial fluoride solution and 3g/l adsorbent dosage. The exhausted adsorbent was regeneration with 0.1N of NaOH solution [26].

Kumar et al have studied defluoridation from water used by thermally prepared carbon from *Neem* (*Azadirachta Indica*) and *Kiker* leaf (*Acacia Nilotica*). At pH neutral fluoride removal maximum efficiency is obtained. From the experimental data Freundlich adsorption isotherm and a first order kinetics were obeyed for the adsorption process [27].

Rayappan *et al* have examined defluoridation from water used to possible of *Cissus Quadrangularis* (CQ) powder. The contact time 60 minutes, 120 rpm trembling speediness and dose of 10mg/50ml, the removal efficiency of fluoride 90% obtained. At optimal pH and 1mg/50ml of dose of adsorbent and maximum removal were 7.0. The occurrence of PO_3^{2-} ions exhibited a tremendously adverse effect for the adsorbent that abridged the defluoridation from 89% to 7%, while the SO_3^{2-} , Cl^- and NO_2^- and CO_3^{2-} , exhibited little adverse effect on adsorption. From the experimental data was best fitted Freundlich and Langmuir adsorption isotherm [28].

Pratibha Pandey, Fahad Khan et al have studied plant of *Azadirachta indica* and *Moringa oleifera* for fluoride removal from ground water. Those plants extracts have strong potential efficiency for removal of fluoride and co existing ions. The experimental data found that have strongly possible of extract on dissimilar water quality parameters including total dissolved solids (525–201), pH (8.4–7.1), total hardness (253–127), turbidity (15.70–5.10), F- concentration (2.80–1.00), and *Escherichia coli* count (325–30) in contrast to *Azadirachta indica* and *Moringa oleifera* leaves extract at the alike doses (0, 25, 50, and 100 mg/L) [56].

LEAVES:

Mondal et al used Tea ash as a bio-sorbent for removal of fluoride from contaminated drinking water. The fluoride removal mechanism was followed pH, contact time and dosage of adsorbent. The bio-adsorbent was 90% successfully remove the fluoride content from water [29].

Sheo Prasad Shukla et al used eco-friendly biosorbent, *Psidium guajava* leaves and found fluoride removal efficiency. The batch experiment reveal that various parameters affect the bio sorption such as bio sorbent dose (1–7 mg/50 mL), pH (2–10), contact time (30–210 min) and initial fluoride concentration (5–12 mg L⁻¹) for the adsorption of fluoride at room temperature. At pH 6.5 and initial fluoride concentration 5 mg/50ml and contact time 180 minutes, the fluoride removal efficiency was 88% achieved. From the experimental data well fitted Freundlich isotherm and found multilayer adsorption of fluoride on treated *Psidium guajava* L. The maximum bio sorption capacity was found 62.50 mg g⁻¹. *Psidium guajava* leaves (L), showed the pseudo-first-order rate equation [30].

Veeraputhiran and Algomuthu et al have used *Phyllanthus Albica* for removal of fluoride from contaminated water. [31]. The fluoride removal was achieved at maximum when smaller particle size while increasing dose of adsorbent F⁻ elimination ability also increases. The interference of co-ions on F⁻ elimination was found to be in the following order HCO₃⁻ > SO₄²⁻ > Cl⁻ > NO₃⁻

Aziz Fazli et al have studied the F⁻ elimination from contaminated drinking water used *Dodonaea viscosa* leaf powder as an adsorbent. At pH 2-8 and fluoride initial dose 1-10 g and contact time 5-145 minutes, the maximum fluoride removal efficiency obtain 45%. From the acidic environment, Langmuir adsorption was well fitted in this study. The *Dodonaea viscosa* leaf powder was low cost adsorbent and F⁻ ions removal also effective and environment friendly method for the defluoridation of drinking water [32].

K.P Singh et al have examined used three plants for fluoride toxicity, namely hyacinth, money plant and water duckweed. It is observed that vastly basic nature of groundwater (pH ≈ 9) within 10 days, hyacinth and duckweed plant died. However without nutrient the money plant sustain 2 month when removal of both total dissolved solid and fluoride from contaminated water. The physico-chemical analysis shows that money plant can store dissolved heavy metals and anionic contaminants, sulfate and chloride. This is potential plant for the low cost method, sustainable F⁻ treatment for fluoride-contaminated water [33].

Peng et al have deliberate for removal of fluoride used chitosan modified natural zeolite. The chitosan modified zeolite had almost three times greater fluoride removal ability than that unmodified zeolite. At pH 4.5 to 5.5 the high level of F⁻ consumption occurred. A pseudo second order kinetics and the intra particle diffusion model were followed by the adsorption kinetics of fluoride on chitosan modified zeolite [34].

Hanumantharao et al have examined the leaf *Acacia Farnesiana* for elimination of F⁻ from aqueous solution. The co ion does affect the fluoride adsorption by CO₃²⁻ ions. At pH 6.9 maximum fluoride removal efficiency obtains. The experimental data well fit for kinetics adsorption process was found to be second order [35].

Aju Mathew George et al have investigated the F⁻ elimination from water used by novel *Ficus benghalensis* leaf bio sorbent. At pH 7.0, initial fluoride concentration 5mg/l, contact time 120 minutes, temperature 30°C, adsorbent dose 10g/l, the high F⁻ elimination competence achieved. From experimental data showed that the Langmuir isotherm with coefficients values obtained as

2.183 mg/g and 0.667 L/mg and fitting well with pseudo second order kinetic model. The thermodynamic studies showed that spontaneous and endothermic nature. The Experiment shows that 90% fluoride removal efficiency and the concentration of treated water fulfills drinking water standards for fluoride [36].

Jamode *et al* have used (Azadirachta indica), (Ficus religiosa), (Acacia catechu wild), leaf powder neem trees for removal of fluoride from aqueous solution. From the investigational records was fixed with Langmuir adsorption isotherm with regression coefficient of 0.9409. At pH 2.0 and bio adsorbent particle size 1.4mm, the removal of fluoride was 50% and bio adsorbent 600 micron particle size adsorbent might eliminate 90% [37].

Kumar *et al* have studied used neem (Azadirachta indica) leaves and kikar (Acacia arabica) leaves for removal of fluoride from contaminated water. At optimum pH and equilibrium time 6.0 to 60 minutes and initial fluoride concentration 5 ppm, the removal efficiency was maximum. The size of adsorbent material which is 0.3mm was more removal of fluoride than size 1.0 mm size of adsorbent. The optimal dosage of neem and kikar leaves is 0.6g/100ml and 0.8g/100ml respectively. The investigational records was fixed with Langmuir first order equation and Freundlich adsorption isotherm[38].

Kamble has examined defluoridation from aqueous solution used Basil (Ocimum sanctum, Lamiaceae) leaves. At pH 9.0, 6.5, 6.0 and 7.0, equilibrium time 20 minutes, initial fluoride concentration leaves dosage of 70 mg/100 ml, 110 mg/100 ml, 250 mg/100 ml and 260 mg/100 ml and removal efficiency of fluoride was 94%, 75%, 78% and 74% obtains. The leaf Basil (Ocimum sanctum, Lamiaceae) is low price effective and environmentally-friendly to react to F⁻ polluted water from country and city area [39].

Bharali and Bhattacharyya have explained fluoride removal from aqueous solution by used Devdaru (Polyalthia longifolia) leaf powder as bio sorbent. At neutral pH and temperature 303K the fluoride removal efficiency 77% was obtains. The maximum rate of adsorption when optimum dose 3.0gm/l. From the experimental records exhibited that the (Polyalthia longifolia) adsorption mechanism involved both intra-particle diffusion and chemo sorption process[40].

Tomar *et al* have examined for F⁻ elimination from aqueous solution by used Citrus limonum (lemon powder). At pH 2.0 and initial fluoride concentration 2 mg/l, the F⁻ elimination competence 70 % achieves. From the experimental data showed greater with Langmuir isotherm than Freundlich isotherm model [41].

Sudarshan *et al.* examined fluoride removal from contaminated water used Tulsi (Holy Basil) leaves. At equilibrium time 20 minutes 95 % F⁻ elimination competence achieved. The F⁻ elimination does not reduction when reach the original time from the experiment data showed that F⁻ solution of 5.0 to 7.0 mg/l was abridged to 1.0 or 2.0 mg/l by bio adsorbent powder, after 2 or 3 hours the F⁻ concentration improved again and got to its original level [42].

BARKS

Karthikeyan and Ilango have studied defluoridation of aqueous solution by used morringa indica bark activated carbon. The high F^- elimination was found at 25 minutes by minimum contact time. At pH 2.0 and F^- original concentration was 3 mg/l, fluoride removal efficiency obtained 80% achieved. The fluoride adsorption obeyed both Freundlich isotherms and Langmuir. Therefore the adsorption reaction was endothermic in nature [43].

Kamble et al has studied fluoride removal from chitin, chitosan, lanthanum-modified chitosan. At pH 6.7 the fluoride removal from contaminated water attain maximum. The equilibrium adsorption numbers has fixed in the Langmuir isotherm model. The F^- elimination was extremely affected co-ions SO_4^{2-} , CO_3^{2-} , and HCO_3^- concentrations. Their competing effect of other co ions adsorption process indicating that fluoride uptake was more in distilled water than from fluoride contaminated ground water [44].

Merugu et al has examined defluoridation from water used fungal bio-sorbent prepared from *Aspergillus Niger*. In this study demonstrated effectiveness of calcium and alkaline treated biomass for F^- elimination. At pH 8, fluoride removal has maximum. The presence of co-ions does not disturb F^- elimination competence such ions Cl^- and SO_4^{2-} but decreased with increasing HCO_3^- concentration [45].

Mamliwar et al have studied used babul bark powder for removal of F^- from aqueous water. At pH 8.0 and F^- concentration was 5 mg/l, the F^- elimination competence obtained 77% with equilibrium time attain at 8 hours and temperature 303K. From experimental data showed both Freundlich isotherm and Langmuir isotherm well fixed therefore pseudo second order kinetic model was good fitted as related to the pseudo first order [46].

The natural adsorbent material prepared from bark of babool tree (*Acacia Nilotica*) for defluoridation by Mamilwar et al. The high F^- removal attains at pH 8 and equilibrium time for the process 8 hours. The experimental adsorbent material showed result was fixed with both pseudo first order kinetic model and Langmuir and Freundlich adsorption isotherm. The co- existing ions presence of nitrate ions the fluoride removal adsorption remain nearly constant but increases with increasing percentage of Mg^{2+} , SO_4^{2-} and Fe^{2+} ions [55].

ROOT

Harikumar *et al* have used Vetiver root powder for removal of fluoride from contaminated water. At pH 6.5 to 7.5 and F^- concentration 2 mg/l, the fluoride removal efficiency was 80 to 90% achieved. The equilibrium data occurred was fitted well with Freundlich isotherms and Langmuir [47].

STEMS

Shyam and Kalwania (2013) have used Khimp plant (*Leptadenia pyrotechnica*) stem powder for fluoride removal purpose. The bio adsorbent powder have calcium ions therefore adsorbed negative charged F^- ions. The following experiment was depending upon precipitation and adsorption method. The high F^- elimination competence was 97% attained at pH 6.0 with dosage of adsorbent 6.0g/l and equilibrium time 60 minutes. The experimental numbers followed both Freundlich isotherms and Langmuir[48].

Chakrabarty and Sharma used neem stem charcoal for removal of fluoride from aqueous solution. At pH 5.0 and dose of 10mg/l of fluoride, the removal efficiency was 94% achieved. The equilibrium of bio sorption was found in 180 minutes. The investigational numbers was fitted both Langmuir and Freundlich isotherm model and high Langmuir removal ability 1.27 mg/l [49].

Suneetha *et al* have examined used stem of an Abutilon Indicum plant for removal of F^- from contaminated water. At pH 7.0 and dose of fluoride 5.0mg/l the removal efficiency of fluoride was 80% . The equilibrium of bio sorption was obtained at 60 minutes with the particle size 45μ and temperature $30\pm 1^\circ C$. The experimental data showed monolayer adsorption and well fitted pseudo-second order kinetic model and Langmuir isotherm model. The sequences of the order of co-existing ions $PO_4^{3-} > HCO_3^- > SO_4^{2-} > NO_3^- > Cl^-$. The presence of phosphate ions is responsible for the reduction of F^- elimination efficiency from 83% to 68.7% with 50 mg/L concentration [50].

PEELS

Gandhi *et al* have deliberated the removal ability of certain low cost adsorbents like various fruits peels used. Fluoride elimination percentage using cheap cost adsorbent materials consuming 12mg/initial F^- absorption for dose of 1g/100ml bio-adsorbent with 24 hours of contact time was found from fruits peels. The percentage of elimination efficiency was $(86\%) > (79\%) > (75\%) > (71) > (65\%) > (56\%) > (53\%)$. The removal of fluoride efficiency attain 80% by using concrete adsorbent for 60 minutes contact time the dose of 0.6g with 10mg/l of fluoride bearing sample. The Langmuir isotherm model for fruits peel adsorbent was best fitted to investigational figures as related to Freundlich isotherm model [51].

Aash Mohammad and Majumder used synthetic industrial wastewater for removal of fluoride from using banana peel, groundnut shell and sweet lemon peel. At pH 6, 7 and 4, different contact time 60, 40, 75 minutes, aqueous solution 20mg/l and fluoride removal efficiency was 93.34, 89.90 and 59.55%. The investigational figures were good fixed to the pseudo second ordered kinetic model with Langmuir isotherm model. The existence of additional ions in the water had slight adverse effects [52].

Jha *et al.* investigate the fluoride removal from drinking water used zirconium (IV) loaded carboxylated orange peel (ZCOP). Fluoride elimination competence attain at pH from 3.0-8.0. At pH range at 7.0, 0.7 g/l adsorbent dosage that maximum fluoride removal efficiency was 97.2% while initial concentration of fluoride limits 10mg/l with contact time of 50 minutes. The pH at 6.0 the maximum adsorption capacity was 5.605 by Langmuir model. The adsorption process was pseudo second order kinetic model. The adsorption process was spontaneous and endothermic nature. The fluoride removal disturbed existence of coexisting ions, PO_3^{2-} and HCO_3^- but NO_3^- , Cl^- and SO_4^{2-} did not disturb fluoride elimination competence [53].

Christina and Viswanathan have examined orange peel residue in alginate matrix and then matrix was loaded with 5% of $FeCl_3 \cdot 6H_2O$ and those composite prepared from orange peel used for fluoride removal purpose for the contaminated water. From the study orange peel residue showed equilibrium adsorption was best fixed with pseudo second order kinetic model with Langmuir

isotherm model. The high adsorption ability of orange peel residue was 7 mg/g. The attraction of carbonyl and hydroxide groups with fluoride has ability to remove the fluoride from contaminated water. The bio-waste of orange peel is active elimination competence of fluoride from aqueous solution[54].

The process of adsorption of F^- by bio-adsorbents can be summarized diagrammatically in the following figure.

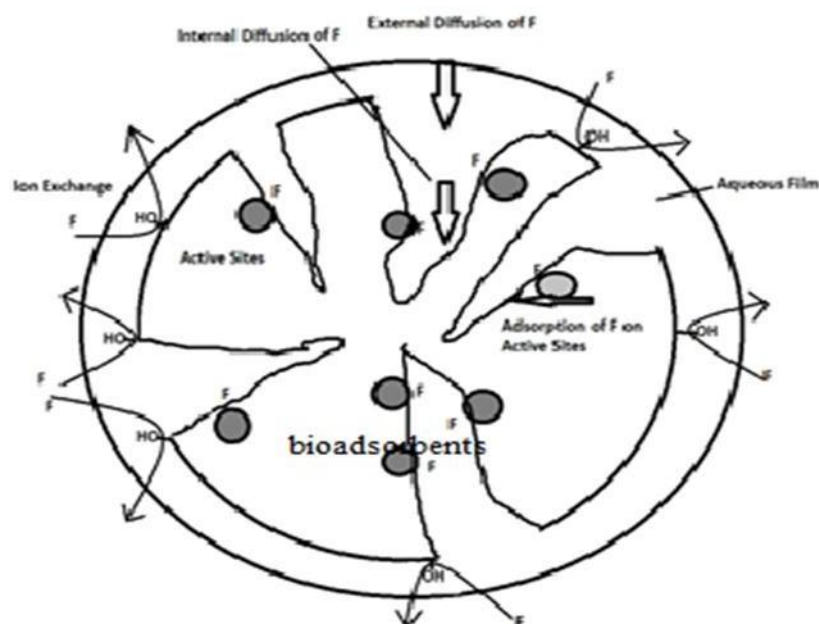


Figure 1. Diagrammatically representation of fluoride adsorption by bio-adsorbents

Conclusion and Future Work Prospects

In India among 19 states has serious health problem due to fluoride contaminated water. The fluoride removal studies measures the different methods and defluoridation techniques. From this review, we have summarized reasonable status of various bio-adsorbent methods using fluoride removal from the contaminated water. Those bio-adsorbent methods observed from defluoridation literatures and along with pros and cons of fluoride removal of contaminated water. Also in this review paper we discussed several parameters that how could affect the defluoridation method have been explained. Further we also discussed chemical and physical nature, temperature, pH, contact time and fluoride removal efficiency. The modified bio-materials were used and different percentage of fluoride removal efficiency was seen from different method. Bio-materials simply exist and usage in defluoridation is harmless and further emphasis for the investigation of bio sorbents. Future research will concentrate on the adaptation and development of bio based green material adsorbents with high fluoride removal capacities and kinetics rates from fluoride-contaminated water that are only marginally influenced by temperature, concentration, pH, and inorganic salts.

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