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Review Paper

Review on removal of fluoride from water by some bio adsorbents

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Abstract

Fluoride (F^-) is an anion of fluorine and which acts as the contaminants as in the form of anionic and it originate in additional in ground or surface water due to geochemical reactions or anthropogenic activities. For animals and human beings the content of fluoride may be requirement under within the desirable limit. As per WHO in drinking water the desirable limit of fluoride should not exceed 1.5 ppm (in the absence of alternate, the fluoride maximum limit is 2.0 ppm). If excess amount of fluoride in water consumed by human beings in long-term, people suffers an adverse effects. As per the available epidemiological studies clearly shows that human beings suffers fluorosis, dental fluorosis, skeletal fluorosis, immunological defects and brittle bones etc. If excess fluoride present in the drinking water rather than desirable limit it must be removed by suitable method, then only it is proper/ safe for drinking. Among various available methods around the global, adsorption process has relatively significant technique for defluoridation in water than precipitation, coagulation, ozonization, electrolytic treatment, reverse osmosis and ion exchange. Due to importance of fluoride in water, people at least know in the aspect of quality and quantity of fluoride in their drinking water and also should know the available remedy methods. In this paper a review study has been taken up on defluoridation from water by some selective biomass as adsorbents and their adsorption capabilities are discussed under various optimum experimental conditions.

Keywords: Fluoride, water, adsorbents, removal of fluoride, Bio adsorbents, defluoridation.

Introduction

Water is most abundant and is an essential compound and people believed that it is always accessible sufficiently as a gift from nature. Depending upon the various chemical constituents present in water it decides suitability of drinking or domestic, Industrial or agricultural purpose. Drinking water affects the health of public due to the existence of chemical constituents & also many diseases associated directly or indirectly depend upon quality of drinking water. Water is contaminated may be natural sources or released effluents from industry or manmade activities. Water is contaminated by fluoride is one of the main important factor because of geochemical reactions or anthropogenic activity and here a slight introduction is given related to occurrence of fluoride in water as follows.

Fluoride ion is capable to form a number of compounds in the earth, air and living beings to form type of compounds as inorganic and organic characteristic nature. Among these formed compounds some of them have a property of quite soluble in water and to be present as in the form of dissociated fluoride ion in surface and ground water[1,2]. In surface and ground water, occurrence of fluoride depends on various causative factors like pH, TDS (Total Dissolved Solids), alkalinity, hardness and geochemical composition of aquifers[3-8].

Natural water is striking enhanced with fluoride is due to geological processes and industry activities. Around the worldwide projecting fluoride amount levels are increased in the water is due to fluorine polluted waste water releases by various industries like fertilizer industry[9,10], ceramic and glass manufacturing processes[11,12], Zinc and Aluminum smelters[13-15], oil refiners[14,16-18], PV cells industry[19,20], high tech-semiconductors production[21-24], electroplating operations, metal etching with HF acid, steel production, thermal power stations, removal of Beryllium from materials, wood preservatives and burning of waste fluorinated plastic materials by Municipalities etc. are triggered in many areas.

As point of human beings Fluoride is an important constituent and would be its affect is mainly based on the total amount consumption by a person. The absorption of fluoride into human body from potable water with the level of 1.0mg/L improves both development and prevents dental carriers and the maximum tolerance limit of fluoride as per WHO[25]in potable water is 1.5 mg/L. If excess amount of fluoride was taken by human beings, it causes dental or skeletal fluorosis, softening of bones and neurological damage are the severe problems. Stephen Peckham and Niyi Awofeso[26] conclude that fluoride has a potential to cause foremost adverse effect on heath of human beings. To overcome by these health problems due to excess fluoride consumption by human beings, defluorinated water is the best practice for drinking purpose.

Many methods or technologies are accessible for defluoridation such as coagulation/precipitation, Reverse Osmosis (RO),

treatments of electrochemical processes, electro coagulation, Ion–exchange and Adsorption with advantages and disadvantages[27]. Adsorption is a better process and useful for the removal of constituents like heavy metals, phenols, ions like chloride, nitrate, sulphate, phosphate and fluoride etc. from water, waste water and industry effluents. It is a worthy method and it works with effectively over the above mentioned methods because of relatively simple, simplicity design, high efficiency, easy handling and availability of different adsorbents as in the form of natural and synthetic materials, agricultural and industrial wastes, and also Nano adsorbents can be employed with minimum cost. Another advantage is suitable to elimination of constituents present even at low concentrations in the samples. In recent years, the search for low-cost adsorbents that have pollutant–binding capacities has intensified around the worldwide. Based on these advantages taken into consideration by number of researches intensified their research in the direction of removal of fluoride or various constituents from drinking water/waste water/industrial effluents by using different types of adsorbents. In addition the adsorption studies are extended to greatly for removal of heavy and toxic metals and various constituents form different types of Industry waste samples or effluents. Overall in the adsorption study, the activated carbon plays a vital role for removal of constituents form samples.

Most of the researchers[28,29] ensure that technique of adsorption technology is economically competent and possibility to produce good quality water. The removal of constituent from sample through adsorption process is on to solid adsorbent leads in a way to three phases. Adsorption process depends upon on species involved in the sample with their bonding sites and carried through weak intermolecular forces[30]. In the selection of adsorbents for removal of constituents; the main points are to be considered in particular to suitability/availability of raw materials (as adsorbent), with stand capacity for removal, regeneration ability, and competency, size of pore and particle, design and cost. The efficiency of defluoridation from sample by adsorption studies depends on quality of raw water, i.e. concentration of fluoride, pH, temperature changes, time of contact, influence of other ions in the sample and adsorbentdosage[17,31-33,29].

Objective of review: To identify the good bio adsorbents for removal of fluoride in the samples with minimum cost and to consolidate their maximum utilization possibilities in adsorption studies in water treatment.

Review and discussion about adsorbents for their removal capacity of fluoride from samples

Several authors have been investigated and reported successfully on defluoridation using different type of adsorbents from naturally occurring, various synthetic materials, oxides and hydroxides, Bio adsorbents, geomaterials, carbonaceous materials, agricultural waste and its by–products, Industrial products and by-products etc. In this paper a review is mainly focused on capacity of defluoridation from samples by some biomassadsorbents.

Easily accessible and eco-friendly mixed adsorbent was prepared by Jamode et. al[34] in the form of powdered sieved particle sizes viz. 600µ, 710µ and 850µ, 1mm and 1.4mm from leaves of trees from neem (Azadirachta indica), Pipal (Ficus religiosa) and kikar (Accia catechu Willd.). The adsorption studies were accompanied at room temperature under the impact of variables like pH (2.00 to 10.00) of aqueous solution, contact time (up to 180 minutes), adsorbent dose (between 0.5 and 12g/L), initial adsorbate concentration (between 2 and 15 mg/L) and adsorbent particle size (600 µ to 1.4 mm). As per their experimental results they have concluded that capably 80% defluoridation was noticed at pH (2.00) in sixty minutes and 70% removal in the case of treated bio sorbents with an initial adsorbate concentration 10 mg/L with 10 g/L of mixed adsorbent dose. Based on the experimental results in this adsorption study they found that treated bio sorbents obeys Langmuir isotherm.

Jain J.K. and Gupta Nidhi[35] prepared bio adsorbents such as seed powder of Black Berry (Syzygium Cumini) (BB), leaf powder of Guava (Psidium Guajava) (GL), Neem Bark (Azadirachta Indica) (NB), Neem leaf powder (NL) and Rice Husk (RH). Furthermore mixed adsorbents of (GL+BB) and (NL+NB)} are also prepared in 1:1 ratio and sieved to get the particle size of 60-250µm are used in their adsorption studies. They have investigated the exclusion efficiency of fluoride by treated adsorbents through batch studies and found that fluoride removal capacity of these adsorbents is significantly high and also they identified mixed adsorbents showed that good adsorption capacity as compared to the individual adsorbents. The experimental results indicates that adsorption kinetics follow first order rate mechanism for Rice Husk(RH), leaf powder of Guava(GL) and Neem Bark(NB), but the above remaining adsorbents followed second order rate mechanism, finally adsorbents well fitted with Langmuir and Freundlich models in fluoride adsorption.

Hanumantharao et *al.*[36] studied defluoridation of water through batch adsorption dynamics and equilibrium studies at room temperature by used as an adsorbent of Acacia Farnesiana Carbonized material (AFC) [Base material is dry fruits of Acacia Farnesiana (in English called as sweet acacia, In Telugu as Nagatumma)]. They have identified that adsorption of fluoride was attained between the pH range of 6.50 to 7.00 and maximum adsorption was noticed around at pH of 6.90. Based on their results at optimum pH, fluoride adsorption follows the Freundlich isotherm indicates that AFC is highly heterogeneous. As per the data obtained from XPS, FTIR, SEM and EDS, they have identified that the result of formation of inner sphere complexation. Fluoride sorption on the AFC surface via inner-sphere complexation accompanying increased hydrogen bonding and surface oxidation has been identified. Finally they

said that adsorbent of Acacia Farnesiana Carbonized (AFC) could be worked as one of the alternate adsorbent to present available commercially activated carbon adsorbents for defluoridation.

Natural bio fiber of Bermuda grass was collected by T.K. Rout *et al.*[37] and it was processed under suitable experimental conditions to prepare adsorbent for their adsorptions studies. In this process the dried grass was converted into form of fibrous powdered without any acid and it is subjected to thermal treatment under CVD (Chemical Vapor Deposition) method in the presence of flowing Nitrogen and finally gets thermally activated carbon without using any catalyst and sieved to particle size 74 to 44 µm. The micro carbon grass powder (1part) is treated separately with 100 parts of Nano materials of TiO_2 and Fe_2O_3 in the Chemical Vapor Deposition setup under suitable conditions for obtaining Fe_2O_3 andTiO_2based carbon nanofiber composites. These Nano composite adsorbents are used for fluoride uptake from sample solution by researchers. In this study the experiments was conducted through batch adsorption by mixing 0.5 g of adsorbent with 10 mg/L of NaF as initial concentration in 100 ml flask. In this research study the defluoridation was performed under optimum experimental conditions such as time of contact, initial fluoride concentration, dose of adsorbent and fluoride uptake with fixed dosage by varying pH at 30 ± 5^{o}C. Finally based on the experimental results they observed that Iron Oxide Based Nano composite (IBNC) is performing quite well for removal of fluoride ion (97%) at a contact time of 60 minutes at pH 4.0. They also noticed that under the same experimental conditions 92% and 88% of fluoride removal was observed with Titania Based Nanocomposite (TBNC) and micro carbon grass adsorbent respectively. Finally the experimental data exposed that fluoride ion uptake is realized by Freundlich adsorption model as well as adsorption capacity and intensity for Iron Oxide Based Nanocomposite (IBNC) was higher than for Titania Based Nanocomposite (TBNC) and micro carbon adsorbent.

S. Jenish and P. Amal Methodis[38] were investigated defluoridation studies through batch experiments by used Tea leaves (after making of tea) with surface modifications as in the form of adsorbent with size in the range 250-500 µm. They found that used tea leaves possibly will be fruitfully used for removal of fluoride over a wide range of concentrations. This study revealed that experimental conditions like: an optimum adsorbent dose (12g/L), 5 mg/L of initial fluoride concentration at pH (6.00) and contact time 150 minutes was lead to 91% fluoride removal and satisfy with Langmuir adsorption Isotherm.

R.N. Patil *et al.*[39] have taken easily available with eco-friendly adsorbent as a bark of Phyllanthus Emblica (Amla) with the object of defluoridation from samples of aqueous medium. Batch experiment studies were performed for identifying the removal capacity of fluoride by this adsorbent, the study was performed under the influence of various experimental conditions such as pH of aqueous solution (2.0 to 10), initial fluoride concentration (3-20 mg/L),adsorbent dose(0.5 to 5.0 g/L), contact time (720 min) and particle size (75-600 µm). The researchers reported that utmost fluoride removal was noticed at pH of 6.0 to 8.0, and the results showed that this adsorbent revealed maximum defluoridation capacity is 5.0 mg/L by use with 1.5 g/L of adsorbent dosage. From the experimental results the data has well fitted with Langmuir and Freundlich isotherm models in sorption process.

Ganvir *et al.*[40] studied the defluoridation from the water by using Aluminum hydroxide coated rice husk ash as an adsorbent. They found that fluoride adsorption capacity of adsorbent was 15.08 mg/g in batch and 9.5 mg/g in column study. The maximum fluoride removal was attained at pH (5.0±0.5) and showed that adsorption was not dependent on initial fluoride concentration. Based on the study they identified that Freundlich isotherm with multilayer adsorption and pseudo second order kinetic were best fitted.

Examined the removal capacity of fluoride by Mondal *et al.*[41] through batch techniques under various experimental conditions such as pH, initial fluoride concentration, adsorbent dose and contact time in presence of adsorbents like Activated Rice Husk Ash (ARHA) with 250 µm size and synthetic Activated Silica gel (ASiG). They observed that maximum adsorption of fluoride was noticed at pH 2.00 and according to Langmuir model maximum adsorption is noticed in ARHA (0.402 mg/g) rather than ASiG (0.244 mg/g) and fit for pseudo second order kinetic model for both the adsorbents. Finally they concluded that as per overall results in this adsorption process the adsorbent ARHA showed little better performance than ASiG.

Defluoridation capability of fresh leaves, dry leaves and stem of Basil (Tulsi leaves; Ocimum sanctum, Lamiaceae) was studied in aqueous solution through batch studies by Kamble[42]. 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. Moderate to high concentrations of fluoride present in groundwater samples from different locations are collected by Sudarshan *et al.*[43] in Nalgonda, Warangal and Karimnagar districts in Andhra Pradesh and adsorption studies was carried out by separately in the above collected samples in presence of tulasi paste, tulasi liquid and tulasi solution (pancha tulasi) with regular intervals. The efficiency of fluoride removal from the collected samples (in100 ml) was studied separately by using 75 mg of tulasi leaves as a first set and add 0.50g and 1.0g of tulasi paste in all samples in the second and third set respectively and in the fourth set three drops of pancha tulasi was added. Based on their results they concluded that the removal of fluoride concentration in ground water samples by tulasi leaves is not much significant. Also tulasi paste and liquid was not much effect (negligible) for defluoridation in ground water samples.

Activated Bamboo Charcoal was prepared from waste bamboo and it is impregnated with Aluminium chloride and Ferric

chloride by Wendimu et al[44], it acts as an adsorbent (AIAABC-Aluminium-iron-amended activated bamboo charcoal) for fluoride removal from aqueous solution. The experimental results showed that fluoride was associated with the Aluminium Hydro (Oxide) (AO) particles of 23.7 mg/g and maximum adsorption capacity was noticed in the adsorbent (AIAABC)) of 21.1 mg/g. The cost of AIAABC is lower and is 87.5% effectively regenerated and compared to commercially available Aluminium hydroxide–based adsorbents; AIAABC is a highly promising material for defluoridation of drinking water.

S. Kanaujia *et. al.*[45] studied defluoridation in groundwater by used with CPGC adsorbent (Carbonized *Punica granatum* Carbon) and it showed that high capability of defluoridation as compared to other available conventional bio adsorbents. The powdered sample of Punica *granatum* (Pomegranate) was dried at 378 - 383 K for 24 hours and it is thoroughly washed with doubly distilled water to remove the free acid completely and again it was dried at above mention temperature for 3 hours to reach complete dryness. After that the dried powdered sample of *Punica granatum* was carbonized at the temperature of 1073 K to 1084 K in Muffle furnace and subsequently the activated adsorbent was sieved to the different particle sizes namely<55, 55 - 106, 106 - 150, 150 - 225 and 225 - 305 micron and these are kept in vacuum desiccators until required for treatment. Through batch adsorption studies de-fluoridation was carried out in the samples under various optimum experimental conditions like contact time, initial fluoride concentration, adsorbent dose, particle size and effect of co-ions with fixed dosage. In this investigation the equilibrium time for removal of fluoride concentration was noticed as 75 min and the fluoride saturation capacity of adsorbent (CPGC) is 1.68 mg fluoride ion per gram at room temperature. Based on the experimental results indicated that Freundlich ($R2 > 0.980$) adsorption isotherm model is best fitted and it specifies that fluoride bio-adsorption onto CPGC is characterized by chemisorption on heterogeneous surfaces. Furtherly they have identified that the particle size of CPGC adsorbent was also an important parameter that affects the sorption for fluoride. Finally the authors concluded that CPGC adsorbent employed as good adsorbent for defluoridation in the water samples and it was easily available as low cost adsorbents within compared to the other conventional adsorbents.

Easily available with minimum cost and environment friendly adsorbent was prepared from treated *Citrus limonum* (lemon) leaf by Tomar *et al.*[46] with the objective of defluoridation from aqueous environment. Batch experiment studies were executed to study the influence of some experimental variables like pH (2.00 to 8.00) of aqueous solution, adsorbent dose (1–10 g/50 mL fluoride solution), contact time (5–145 min), initial fluoride concentration (2–15 mg/L) and the presence of few competing anions on the adsorption of fluoride on *C. limonum* (lemon) leaf adsorbent. They reported that maximum efficient of fluoride removal was noticed at pH 2.0and 70% defluoridation capacity per 2 mg/L fluoride ion. Based on the obtained values obey Langmuir and Freundlich isotherm models in this study.

Kalpana Singh *et al.*[47] in their present study deals with the adsorption of fluoride from aqueous solutions by using Activated Carbon prepared from Bael Shell (ACBS). The batch adsorption experiments were carried out to find the fluoride adsorption capacity of ACBS from aqueous solution. The effect of various parameters viz. adsorbent dose, initial pH, contact time, initial concentration and temperature on the adsorption of fluoride have been studied. The study reveals that, the adsorption rate is found to be very fast and at the initial concentration of 4 mg/L, about 52% fluoride removal was found in 60 minutes contact time at the adsorbent dose of 2 g/L. The maximum uptake of 1.07 mg/g was found at lower concentration (4 mg/L) and 2.4 mg/g at higher concentration (8 mg/L) at the optimum dose and at 30°C. The adsorption isotherm data fits Redlich–Peterson, Langmuir, Radke-Prausnitz, Toth, Temkin and Freundlich isotherm in well.

Shubha Dwivedi *et al.*[48]investigatedbio adsorption of fluoride from aqueous solution by Ficusreligiosa (peepal) leaves. Batch study experiments were performed and noticed that the bio adsorption of fluoride on Ficusreligiosa (peepal) leaves were strongly pH dependent, and maximum fluoride removal was found to at equilibrium pH of 7.00 with optimum adsorbent dose (10g/L), temperature (30°C), time (45 min) and initial concentration (20 ppm). Freundlich isotherm gives well prediction of the equilibrium adsorption and the specific uptake increases from 0.09 mg/g to 1.48 mg/g with the increase in initial fluoride concentration from 1 mg/L to 20 mg/L. Maximum specific uptake obtained from Langmuir isotherm was found (24 mg/g) and 85.7% of defluoridation was noticed in this adsorption study.

Conclusion

Due to the consideration of effectiveness, simplicity of experimental design, the conditions of economic and environmental, adsorption study plays a vital role for removal of constituents present in the water. This paper provides briefly an indication about details of working conditions and capability of some bio-adsorbents in place of expensive commercial and conventional adsorbents for the defluoridation study in water or waste water treatment. A systematic study was impressed on these quoted references in this paper; at appropriate places the discussions were stated with respect to adsorption studies. An equilibrium study as well as pH of water is dominant influencing factors for fluoride adsorption on adsorbents. Majority of the researchers have been reported/quoted the rate of defluoridation increases from acidic to near neutral pH and then decreases with increase in pH (i.e. basic medium). In addition another significant factor in defluroridation studies is that the type and concentration of other ions present in treated water, which plays an important role.

Efficiency of bio adsorbents for fluoride adsorption mainly depends on nature of existing multifunctional group and alteration which has been piloted through increase adsorption capacity. In this review broadly understood the efficiency of different adsorbents in the removal of fluoride depends on dose of adsorbate, characteristics of adsorbent, pH, temperature, contact time, speed of agitation, increasing dose and decreasing size of the adsorbent etc. As per review papers the authors have been reported that the experimental data fitted sound to the different adsorption isotherms.

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