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EFFECTIVE BIO-ADSORBENTS FOR REMOVAL OF FLUORIDE FROM WATER: A REVIEW

Prerna Kumari

PG Scholar, Environmental Science & Engineering Group Birla Institute of Technology, Mesra, Ranchi (India)

ABSTRACT

Fluorosis is a public health problem in many parts of the world. The major source of fluoride intake is drinking water. Even though fluorosis once established is irreversible but it can be prevented through simple precaution. The permissible limit of fluoride concentration in drinking water is 1.5mg/L according to WHO guidelines. Thus, defluoridation of water and supply of safe drinking water is the immediate solution and best preventive measure. Cost is of major consideration in developing countries like India, hence, the adsorption process using bio- adsorbents that are abundant, easily available have been investigated. This review listed on efficiency of different biomass for the sequestration of fluoride from water by using adsorption technique. Utility of various plant materials, agricultural wastes or biomass as bio-adsorbents are overviewed and their efficiency of eliminating fluoride is studied with different parameters such as pH, agitation time, dose, temperature, initial fluoride concentration, and particle size.

Keywords: Fluorosis, Adsorption, Bio- Adsorbent, Sequestration, Drinking Water.

I. INTRODUCTION

Fluorine (F2) due to its high electronegative and reactivity cannot found in natural environment in elemental form. Fluoride (F^-) is a fluorine anion which has a great tendency to behave as ligand and easiness to form a great number of different organic and inorganic compounds in soil, rocks, air, plants and animals. These compounds are quite soluble in water, so fluoride is present in surface and groundwater as an almost completely dissociated fluoride ion [1, 2].

Drinking water is the major source of fluoride daily intake and continuous consumption of drinking water with heightened fluoride concentrations (>1.5 mg/L) can induces birth, reproduction and immunological defects [3, 4], dental and skeletal fluorosis [5–12]. There are lot of methods developed for removal of excess fluoride from drinking water, such as the use of ion exchange columns, coagulation, use of membranes and electrochemical methods, the high cost of these technologies makes them unpractical for developing countries[13]. Among these techniques, adsorption discovered as an effective, environmentally friendly economical one [14]. Adsorption process using conventional adsorbent like activated carbon is used to remove fluoride ion metals from water. However, adsorbent-grade activated carbon is expensive and the regeneration of the used carbon is often difficult, resulting in low feasibility. Thus, considerable attention has been given to the use of agriculture waste materials as an alternative to replace the conventional adsorbents. Agriculture waste materials (bio- adsorbents) are inexpensive, available in large quantities and remain unused; they can be disposed without concerning expensive regeneration process. Furthermore, plant materials like Citrus limonum (lemon) leaf [15], Ficusreligiosa (Peepal Leaf Powder) [16], Banana peel dust [17], Devdaru (Polyalthia Longifolia) leaves [18], Neem charcoal [19]. Neem (Azadirachta indica) and Kikar (Acacia arabica) leaves [20] are reported as fluoride sequestration and hence applicable for defluoridation agents. Apart from these numerous waste, biomass sources on which some experimental adsorption properties have been reported e.g. Sawdust [21], Tea Ash [22], Maize Husk Fly Ash [23], Eggshell Powder [24], Algal Spirogyra Sp. I02 [25], Babool Bark [26], Tamarind seed [27], Banana peel, groundnut shell and sweet lemon peel[28]. Reports on extraction of fluoride using different biomass are also available in recent literatures.

II. LITERATURE REVIEW

V. Tomar et al. (2014) investigated the most inexpensive, easily available and eco-friendly analyte (fluoride) adsorbent of treated Citrus limonum (lemon) leaf has been developed and evaluated its feasibility for fluoride ion removal from aqueous environment. The adsorption of fluoride ion was affected by pH, adsorbent dose, contact time and initial fluoride concentrations. Batch experiments were performed to study the influence of various experimental variables such as pH of aqueous solution (2–8), 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. The optimal value of pH 2 was observed where the adsorbent showed the maximum defluoridation capacity of 70% of 2 mg/L fluoride ion. The experimental data revealed that both the Langmuir and Freundlich isotherm models fitted with the fluoride sorption process but very well followed Freundlich isotherm model [15].

Shubha Dwivedi et al. (2014) focused on the bioadsorption of fluoride from aqueous solution by Ficusreligiosa (peepal) leaves. Removal of fluoride has been investigated as a function of pH, bioadsorbent dose, temperature, time and equilibrium initial fluoride ion concentration. Batch study exposed that the bioadsorption of fluoride on Ficusreligiosa (peepal) leaves were strongly pH dependent, and maximum fluoride removal was found to occur at equilibrium pH of 7. Optimum adsorbent dose, temperature, time and initial concentration were found 10 g/L, 30°C, 45 min and 20 ppm respectively. Characterization of peepal leaves, before and after adsorption were studied by Scanning Electron Micrograph to get a well understanding into the mechanism of adsorption. Freundlich isotherm gives well prediction of the equilibrium adsorption (R^2 = 0.995). 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 is found to be 2.24 mg/g. When the initial fluoride concentration is 5mg/L, the removal efficiency of peepal leaf powder is 85.7% so that the fluoride concentration at the treated water is below the permissible limit [16].

Ria Bhaumik • Naba Kumar Mondal (2014) highlighted the effective application of banana peel dust (BPD) for removal of fluoride (F^-) from aqueous solution. The effects of operating parameters such as pH, initial concentration, adsorbent dose, contact time, agitation speed and temperature were analysed using response surface methodology. The significance of independent variables and their interactions were tested by the analysis of variance and t test statistics. Experimental results revealed that BPD has higher F^- adsorption capacity (17.43, 26.31 and 39.5 mg/g). Fluoride adsorption kinetics followed pseudo-second-order model with high correlation of coefficient value (0.998). On the other hand, thermodynamic data suggest that adsorption is favoured at lower temperature, exothermic in nature and enthalpy driven. The adsorbents were characterised through scanning electron microscope, Fourier transform infrared spectroscopy and point of zero charges (pHZPC) ranges from pH 6.2–8.2. Finally, error analysis clearly demonstrates that the adsorbents are well fitted with Langmuir isotherm compared to the other isotherm models. The reusable properties of the material support further development for commercial application purpose [17].

Bharali and Bhattacharyya (2014) prepared Devdaru (*Polyalthia longifolia*) leaf powder, DLP, from mature, dried *Polyalthia longifolia* (Devdaru) leaves was used as biosorbent for removal of fluoride from aqueous solutions using the batch adsorption process. The biosorbent was first characterized with respect to surface area, surface functional groups by FTIR, cation and anion exchange capacities, surface topography by SEM technique and then effects of pH, agitation time and fluoride concentration, adsorbent amount and temperature on adsorption of fluoride were investigated to determine optimum adsorption properties. The biosorbent was found to be effective around pH 7.0 and the maximum fluoride removal capacity of this adsorbent was about 77 % at 303 K. Kinetic study showed that the fluoride sorption on DLP was predominantly chemical in nature and its mechanism was a complex one involving both surface adsorption and intra-particle diffusion. Thermodynamic study showed that sorption process was exothermic in nature and was found to be favourable at lower temperature[18].

Sutapa Chakrabarty et al. (2012) studied for defluoridation on neem charcoal powder showed that the adsorbent were highly influenced by temperature, pH of the solution, and initial fluoride concentration. The neem stem charcoal is found to be an efficient adsorbent for the defluoridation of contaminated drinking water sources. The biosorbent was successful in removal of fluoride ions from aqueous solution of 10mg/l fluoride concentration with about 94% efficiency. Biosorption equilibrium was achieved within 180 minutes. It was observed that the adsorption was pH dependent with maximum adsorption achieved at pH 5.0. Both Langmuir and freundlich isotherm models fits well to the adsorption mechanism. Although regression coefficient of both pseudo first order and pseudo second order plot indicates adherence of both the rate laws but higher regression value of second order plot than the pseudo first order reaction indicates that the adsorption follows the second order rate law [19].

Kumar et al. (2008) investigation deals with fluoride removal from aqueous solution by thermally activated neem (Azadirachta indica) leaves carbon (ANC) and thermally activated kikar (Acacia arabica) leaves carbon (AKC) adsorbents. In this study, neem leaves carbon and kikar leaves carbon prepared by heating the leaves at 400°C in electric furnace was found to be useful for the removal of fluoride. The adsorbents of 0.3 mm and 1.0 mm sizes of neem and kikar leaves carbon was prepared by standard sieve. Batch experiments done to see the fluoride removal properties from synthetic solution of 5 ppm to study the influence of pH, adsorbent dose and contact time on adsorption efficiency. The optimum pH was found to be 6 for both adsorbents. The optimum dose was found to be 0.5g/100 ml for ANC (activated neem leaves carbon) and 0.7g/100 ml for AKC (activated kikar leaves carbon). The optimum time was found to be one hour for both the adsorbent. It was also found that adsorbent size of 0.3 mm was more efficient than the 1.0 mm size. The adsorption process obeyed Freundlich adsorption isotherm. The validity of langergren equation consequently first order nature of the process involved in the study. Results indicate that besides intraparticle diffusion there may be other processes controlling the rate which may be operating simultaneously[20].

Suman Mann et al. (2014) studied the adsorption behaviour of sawdust in order to consider its application for fluoride removal. The batch adsorption method was employed: Laboratory investigation of the potential of sawdust to remove fluoride from aqueous solution has been studied. The effects of various experimental parameters, such as pH (3-11), adsorbent dosage (0.5-2.5 g/l), particle size (90µm to 300µm), contact time (30-150 min) and initial concentration(5 to 30mg/l) were investigated . The equilibrium data have been analyzed by the Langmuir, Freundlich and Tempkin isotherm models, the experimental data were better fitted to the Langmuir equation. The adsorption kinetics also investigated by the pseudo-first-order, pseudo-second-order, intraparticle diffusion and Elovich model . The deflouridation process followed pseudo second order model [21].

Naba Kumar Mondal, RIA Bhaumik, Tanmoy Baur, Biswajit Das, Palas Roy and Jayanta Kumar Datta (2012) studied the removal of fluoride using Tea ash as adsorbent through batch studies. The authors reported that the adsorbent was efficient for the uptake of fluoride at pH 6 and contact time 180 minutes. Tea ash was found to me more efficient at an initial concentration of 5mg/l and temperature 303 k. The authors also reported that the data nicely fitted with Langmuir adsorption isotherm indicating monolayer adsorption and adsorption of fluoride decreased with increase in temperature in the range of 303-333 K. Again the adsorption process was observed to follow a pseudo-second-order kinetic model [22].

Jadhav A S[1], Jadhav M V[2] study batch adsorption process was conducted to evaluate the suitability of Maize husk fly ash (MH fly ash) as an adsorbent for removing fluoride from water. During the experimental work, effects of some of the major parameters of adsorption, viz. Contact time, pH , Adsorbent dose and Stirring rate on removal efficiency were studied and optimized. The equilibrium was attained in 120 minutes, Maximum removal efficiency was obtained at Ph value of 2, optimum adsorbent dose was found to be 2.0 g/50 mL, optimum stirring rate was obtained at 250 rpm. Maximum fluoride removal was observed to be 86% at optimum conditions. Freudlich, Langmuir, Temkin and Redlich-Perterson isotherms were plotted and best suited model was found [23].

R. Bhaumik, N. K. Mondal, B. Das , P. Roy, K. C. Pal, C. Das, A. Banerjee, and J. K. Datta (2011) Studied the role of eggshell powder as an adsorbent for removal of fluoride from aqueous solution using batch technique. The maximum adsorption was achieved at pH 2.0-6.0. The investigators achieved around 94% removal of fluoride at initial metal ions concentration of 5 mg/l at optimum dose of 2.4 g/100ml and optimum time of 120minutes. Experimental data provided best fit with the Langmuir isotherm model, indicating monolayer sorption on a homogenous sur. The adsorption kinetics followed pseudo-second-order kinetic model indicating towards chemisorption. Intra-particle diffusion was not the sole rate controlling factor [24].

S. Venkata Mohan et al. (2007) pertaining to the adsorptive studies carried out on fluoride removal onto algal biosorbent (*Spirogyra IO2*). Batch sorption studies were performed and the results revealed that biosorbent demonstrated ability to adsorb the fluoride. Influence of varying the conditions for removal of fluoride, such as the fluoride concentration, the pH of aqueous solution, the dosage of adsorbent, the temperature on removal of fluoride, and the adsorption–desorption studies were investigated. Sorption interaction of fluoride on to algal species obeyed the pseudo first order rate equation. Experimental data showed good fit with the Langmuir's adsorption isotherm model. Fluoride sorption was found to be dependent on the aqueous phase pH and the uptake was observed to be greater at lower pH. Maximum fluoride sorption was observed at operating 30°C operating temperature. Adsorption–desorption of fluoride into inorganic solutions and distilled water was observed and this indicated the combined effect of ion exchange and physical sorption phenomena. Significant changes in the FT-IR spectra was observed after fluoride sorption which is indicative of the participation of surface function groups associated with hydrogen atoms in the carboxylic groups in sorption interaction. From X-ray photoelectron spectroscopy (XPS) analysis a marginal increase in the area for the binding energy peak at 287.4 eV was observed which could be due to the formation of –C–F– bonds. Thermo gravimetric (TGA) analysis of the fluoride loaded sorbent showed that the biosorbent underwent three steps decomposition process when heated from 25 to 100°C. The maximum weight loss was observed to be between 200 and 400°C and 700 and 800°C[25].

Bhagyashree M Mamilwar, A.G.Bhole, A.M.Sudame (2012) investigated the adsorption of Fluoride using bark of babool as adsorbent. The investigators conducted batch studies and used Freundlich and Langmuir isotherms to understand the adsorption mechanism. Optimum dose of bark of babool was found 5g/L for removal of fluoride concentration of 5 mg/L. Adsorption capacity was more in the pH range of 6-8.Optimum time of contact was found 8 hrs. The removal increased with time and adsorbent dose, but with higher initial concentration decreased with time and adsorbent dose. The study on defluoridation using bark of babool shows that the equilibrium data fits better to Langmuir isotherm as compared with Freundlich isotherm. The pseudo-second-order kinetic model fitted well as compared to pseudo first-order model [26]**.**

M. Murugan and E. Subramanian (2006) used Tamarind seed for the sorptive removal of fluoride from synthetic aqueous solution as well as from field water samples. Batch sorptive defluoridation was conducted under variable experimental conditions such as pH, agitation time, initial fluoride concentration, particle size and sorbent dose. Maximum defluoridation was achieved at pH 7.0. Defluoridation capacity decreases with increase in temperature and particle size. Further, defluoridation follows first order kinetics and Langmuir adsorption isotherm. Desorption was carried out with 0.1 N HCl and is 90 per cent. The surface and sorption characteristics were analysed using FTIR and SEM techniques. All these results indicate the involvement of energetic forces such as coulombic interaction in sorption. For domestic and industrial applications, defluoridation with 100% achievement and subsequent regeneration of adsorbent was performed with a household water filter and fixed bed column respectively [27].

Aash Mohammad et al. (2014) investigates the feasibility of three low-cost biomass based adsorbents namely: banana peel, groundnut shell and sweet lemon peel for industrial waste water defluoridation at neutral PH range. Action of these adsorbents on fluoride was compared with commercially available adsorbents. It was found to be much better, high removal efficiency at higher concentration (20 mg/l) of fluoride in industrial waste water. The banana peel, groundnut shell and sweet lemon peel removed 94.34, 89.9 and 59.59 %respectively. Contact time for banana peel, groundnut shell, and sweet lemon peel are 60.0, 75.0, and 40 min respectively at doses of 14, 12 and 16 gm/l respectively. Mechanism of adsorption kinetics was found pseudo-second order reaction, and the mechanism of fluoride removal on adsorbents was found to be complex. The surface adsorption as well as intra-particle diffusion contributes to the rate-determining step [28].

M. A. T. Ajisha et al. (November 2013–January 2014) performed new and cost effective defluoridation method using Pyrolysed Cocos nucifera Midribs (PCM) which is abundantly available and inexpensive raw material from an agricultural tree coconut palm. PCM was characterized both physically and chemically. PCM has very good surface area of 255.0968 m^2/g for an effective particle size of 150μ. Effect of pH, dosage of PCM, contact time, initial fluoride ion concentration, temperature and interfering Co-ions were determined using the batch studies. PCM was further characterized by FTIR, SEM studies. In FTIR analysis stretching at 2879.52, 2979.82 cm^{-1} and formation of new peaks at 1625.88, 540.03, 441.67 cm^{-1} shows the adsorption fluoride ion due to C–H, C–O stretching and C=C groups of various oxides. SEM analysis shows the presence of tubular and circular micro pores.

Equilibrium data obtained was best fixed to Freundlich isotherm. Freundlich was more suitable confirming the multilayer sorption. The sorption nature was studied using thermodynamic parameters Standard free energy change $\Delta G°$, enthalpy change $\Delta H°$ and entropy change $\Delta S°$. Thermodynamic study showed spontaneous, endothermic, irreversible and stable adsorption. Pyrolysed Cocos nucifera Midribs exhibited the potential adsorbent for application in treatment of aqueous solution of fluoride ions [29]

III. CONCLUSIONS

This paper provides an overview of various bio-adsorbents in place of expensive commercial adsorbents for the removal of fluoride from water. 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, etc. The removal capacity increases by increasing dose of the adsorbent and decreasing size of the adsorbent. The equilibrium data fitted well to the adsorption isotherms like Langmuir and Freundlich. A review of various biomass presented here shows a great potential for the fluoride removal. The use of commercially available adsorbents can be replaced by the inexpensive and effective bio-adsorbents. The future research should be concentrated in evaluating the efficacy of adsorbents in terms of cost and feasibility for the removal of fluoride.

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AUTHOR

Prerna Kumari, PG Scholar, Environmental Science & Engineering Group Birla Institute of Technology, Mesra, Ranchi (India),