Comparative Study of Defluoridation from Water using Waste Materials as Adsorbents – A Review

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Abstract- Fluoride is one of the most abundant constituents occurring in the groundwater in many parts of the world, posing a potential threat to human health. The permissible limit of fluoride concentration in drinking water is 1.5mg/L according to WHO guidelines. In India, 25 million people in 19 states and union territories have already been affected and another 66 million are at risk, including 6 million children below the age of 14 years [1]. Among various methods used for defluoridation of water Adsorption method is relatively simple, economical, and appropriate for drinking water treatment, especially for small communities. In this review, a list of various adsorbents and their adsorption capacities for fluoride are overviewed with various affecting parameters.

Keywords – Adsorption, Adsorption Isotherms, Removal efficiency, Waste adsorbents.

I. Introduction

Fluoride is one of the most abundant constituents occurring in the groundwater in many parts of the world, posing a potential threat to human health. It is one of the three important chemicals, in addition to arsenic and nitrate, which cause large-scale health problems through drinking water exposure. Fluoride-related health hazards are considered to be a major environmental problem [2]. The presence of naturally occurring fluorides or added fluoridated salts in drinking water allows its easy entrance in the body via the gastrointestinal tract [3, 4]. The epidemiological studies reveal that 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 [5,6], dental and skeletal fluorosis [7–14]. Besides drinking water, fluoride can also enter into the human body through food, industrial exposure, drugs, cosmetics, *etc.* [15]. Depending on the concentration and the duration of continuous uptake, the impact of fluoride in drinking water can be beneficial or detrimental to mankind. Fluoride in drinking water has a narrow beneficial concentration range in relation to human health. Small amounts in ingested water are usually considered good to have a beneficial effect on the rate of occurrence of dental caries, particularly among children [16]. On the contrary as mentioned earlier, excess intake of fluoride lead to various diseases such as osteoporosis, arthritis, brittle bones, cancer, infertility in women, brain damage, Alzheimer syndrome, and thyroid disorder [17-18]. Fluorosis is a common symptom of high fluoride ingestion manifested by mottling of teeth in mild cases and softening of bones and neurological damage in severe cases [19]. Defluoridation of drinking water is the best practicable option to overcome the problem of excessive fluoride in drinking water, where alternate source is not available. During the years following the discovery of fluoride as the cause of fluorosis, extensive research has been done on various methods for removal of fluoride from water and wastewater. Among all available methods adsorption is considered to be simple, economical and globally pursued technique.

II. ADSORPTION

Adsorption is typically used in wastewater treatment to remove toxic or recalcitrant organic pollutants (especially halogenated but also non-halogenated), and to a lesser extent, inorganic contaminants, from the wastewater. Researchers in recent year argue that the adsorption technique is economical efficient and produces good quality water.

A typical adsorption process has four steps:
1. Addition of feed solution and the adsorbent solid particles.
2. Good mixing to enhance the transfer of solutes from the liquid phase to the surface area of the adsorbent solid particles.
3. Separation of the adsorbent plus adsorbate (solute) from the bulk of solution.
4. Removal of adsorbed solute from the surface of the solid adsorbent by using a different solvent. This is called elution of solute and the solvent is called elution solvent.
5. At steady-state adsorption conditions there is a physical equilibrium between the concentration of solute in the liquid.

Adsorption studies pointed most important characteristics which determined adsorbent suitability for practical application: adsorption capacity, selectivity for fluoride ions, regenerability, compatibility, particle and pore size, and cost while fluoride removal efficiency always depends on raw water quality profile, *i.e.*, initial fluoride concentration, pH, temperature, contact time and adsorbent dosage [19-24].
Processed materials like activated alumina, activated carbon, bone char, defluoron-2 (sulphonated coal) and synthetic materials like ion exchange resins have been extensively evaluated for defluoridation of drinking water. Among these materials, bone char, activated alumina and calcined clays have been successfully used in the field; [25-27].

III. OBJECTIVE OF REVIEW

Removal Fluoride from water is expensive process. Hence this study reviews the development of waste materials adsorbents as non conventional alternative for Fluoride removal.

IV. REVIEW OF WASTE ADSORBENTS

Dobaradaran, Nabipour, Mahvi, Keshtkar, Elmi, Amanollahzade, Khorsand (2014) Studied Fluoride removal from aqueous solution using shrimp shell waste by batch experiment. Investigator reported that, for an initial F concentration of 8 mg/L, the removal percentage of F increased with increasing adsorbent dose from 3.2 g/L to 64 g/L. The maximum removal of 80% was obtained at pH 11with15 minutes of contact time and adsorbate dose of 8mg/l. The investigator reported that shrimp shell waste exhibited a high defluoridation capacity within few minutes of contact time as compared to other adsorbents [28].

AashMohammad and CB Majumder(2014) Investigated 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. The banana peel, groundnut shell and sweet lemon peel removed 94.34, 89.9 and 59.59 % of fluoride 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. 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 [29].

Dwivedi Shubha, Mondal Prasenjit and Balomajumder Chandrajit (2014) Investigated the removal of fluoride using *Citrus limetta* in batch reactor. The Freundlich isotherm gives well prediction of the equilibrium adsorption (R2 = 0.996). The specific uptake increases from 0.089 mg/g to 1.35mg/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 1.82 mg/g. When the initial fluoride concentration is 5mg/L, the removal efficiency of mosambi peel is 82.5%. Adsorption kinetics is presented well by pseudo second order rate equation and the estimated equilibrium concentration falls within ~ 6 % error limit [30].

Pali Shahjee, B.J.Godboley, A.M.Sudame (2013) Investigated or checked efficiency of low cost adsorbent (Bleaching Powder) for the removal of excess fluoride from aqueous solution. The studies were conducted under batch adsorption. The investigators concluded that the bleaching powder is a good adsorbent for fluoride removal from aqueous solution. The optimal conditions for the effective removal of fluoride by bleaching powder were found to be at pH 10, contact time 8 hours and optimum dose of adsorbent was found 7.3gm/100ml for removal of fluoride of 5ppm concentration [31].

Mohammad Mehdi Mehrabani Ardekani, Roshanak Rezaei Kalantary, Sahand Jorfi, Mohammad Nurisepehr (2013) Compared the efficiency of Bagas, Modified Bagas and Chitosan for fluoride removal from water by adsorption. The pH value of 7, contact time of 60 min and adsorbent dose of 2 g/L were determined as optimum conditions for all three adsorbents. Chitozan and bagas did not show good capability for fluoride removal, but modified bagas showed more than 90% removal at optimized conditions, including the pH value of 7, contact time of 60 min and adsorbent dosage of 2 g/L. Both Langmuir and Freundlich isotherms show good correlation for description of results, but the Langmuir model with the correlation value of 0/99 is superior[32].

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 be 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. The adsorption process was observed to follow a pseudo-second-order kinetic model [33].

Patil Satish, Renukdas Sameer, Patel Naseema(2012) Performed batch study to investigate the efficacy of treated natural adsorbents such as Mangrove plant leaf powder (MPLP), Almond tree bark powder (ATBP), Pineapple peel powder (PPP), Chiku leaf powder (CLP), Toor plant leaf powder (TPLP) and Coconut coir pith (CCP). Researchers reported the effect of pH, contact time, adsorbent dose and initial metal ion concentration to remove fluoride ions from the aqueous solutions . Uptake of fluoride ions by adsorbents at equilibrium is found to be in the order of MPLP > CCP > TPLP > CLP > PPP > ATBP. The optimum contact time for all the adsorbents was 60 minute with a adsorbent dose of 10g/l for initial fluoride concentration of 5ppm. The highest percentage removal was found at pH 2. [34].

Naba Kr Mondal, Ria Bhaumik, Arnab Banerjee, Jayanta Kr Datta, Tanmoy Baur(2012) Conducted a comparative study for removal of fluoride using activated silica gel (ASiG) and activated rice husk ash (ARHA) as adsorbents through batch studies. The authors reported that both adsorbents were efficient for the uptake of fluoride at pH2.0 and contact time 100 minutes. ASiG was found to be more efficient than ARHA with an initial fluoride concentration of 5mg/l, percentage removal efficiency was 88.30 and 96.7 for ARHA and ASiG respectively.. The study on equilibrium sorption revealed that Langmuir isotherm model give best fit to experimental data [35].

R.Bhaumik, N.K. Mondal, P.Roy, C.Das, A. Banerjee And J.K.Datta (2012) Investigated egg shell powder as a medium of fluoride removal from aqueous solution. Fluoride adsorption was studied in a batch system. The researchers reported that The maximum adsorption occurred at pH 2.0-6.0. Experimental equilibrium data provided best fit with the Langmuir isotherm model, indicating monolayer sorption on a homogenous surface (maximum monolayer sorption capacity was 1.09 mg g^{-1} at 303 K). The activation energy of the adsorption process (Ea) was found to be 45.98 $kJmol^{-1}$ by using Arrhenius equation, indicating chemisorption nature of fluoride onto eggshell powder also. Thermodynamic analysis suggests that removal of fluoride from aqueous solution by eggshell powder was a spontaneous and exothermic process. The present findings suggest that such eggshell can be used as a waste adsorbent, it also can provide a simple, effective and low cost method for removing F^- form contaminated water [36].

S.T. Ramesh, R. Gandhimathi, P. V. Nidheesh and M. Taywade (2012) Investigated the adsorption potential of bottom ash for defluoridation of drinking water using batch and continuous fixed bed column studies. The optimum contact time for fluoride was found to be 105 minutes with the maximum efficiency of 73.5 % at 70mg/100ml bottom ash dosage. The optimum pH was found to be pH 6 with the maximum efficiency of 83.2 %.During the column studies; increase in fluoride ion uptake with an increase in the bed height was due to an increase in the contact time. A high degree of linearity of the BDST plot indicates the validity of the BDST Model when applied to continuous column studies [37].

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 present 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 [38].

N.Gandhi, D. Sirisha, K.B. Chandra Shekar and Smita Asthana (2012) Conducted study on adsorbents like concrete, ragi seed powder, Red soil, horse gram seed powder, orange peel powder, chalk powder, pineapple peel powder and multhani matti. Experimental setup was bath studies. Result indicated fluoride removal efficiency of 86% for chalk powder and pineapple peal powder. 79% and 75% for horse gram seed powder respectively. Percentage removal for ragi seed and red mud was found to be 65% and 71%. Removal efficiency was recorded less for multani mitti and concrete which was 56 % and 53% [39].

S. A. Valencia-Leal, R. Cortés-Martinez, R. Alfaro-Cuevas-Villanueva (2012) Assessed Guava seed as waste bioadsorbent for removal of fluoride for aqueous solution. Authors concluded that maximum adsorption occurred between pH 5-8. Langmuir and Freundlich adsorption isotherm models were applied to evaluate the adsorption data. The pseudo-second order model describes the fluoride sorption kinetics using guava seed at different temperature. The Langmuir model best describes the isotherm‟s experimental data, which may indicate that the sorption mechanism of fluoride ions on guava seed is chemisorption on a homogeneous material [40].

Piyush Kant Pandey, Madhurima Pandey, Rekha Sharma(2012) This work was based on search of biomass - Tinasporia Cardifolia as bioadsorbent for removal of fluoride from waste water. The efficiency of the sorption process was investigated under batch different experimental parameters such as pH 7, standing time 120 min and biomass doses 7.0 g with 5 mg/L concentration of fluoride. Neutral pH was identified as the optimum condition of the medium and 120 minutes was the best contact time for maximum fluoride adsorption. The experimental data was found good fitting to Langmuir and Freundlich isotherm models. The maximum removal efficiency of 70% was reported at pH 7[41].

C.M.Vivek Vardhan and J.Karthikeyan(2011) Carried out investigations for removal of Fluoride from water employing physico-chemical processes of adsorption and coagulation employing abundantly available and low- cost materials like Rice Husk, seed extracts of Moringa Oleifera (Drum stick), and chemicals like Manganese Sulphate and Manganese Chloride. Rice husk of 6g/l accomplished a removal of 83% of Fluoride from a 5mg/l of Fluoride solution requiring an equilibrium time of 3 hours. Equilibrium Isothermal data fitted well into rearranged linearised Langmuir adsorption model. Moringa oleifera seed extracts, Manganese Sulphate and Manganese Chloride accomplished removal percentages of 92, 94 and 91 of Fluoride from a 5mg/l test solution at a dosage of 1000 mg/l. A slightly acidic pH of 6.0 was found favourable for Fluoride removal by Manganese sulphate, Manganese Chloride and MOE [42].

G.Alagumuthu, V. Veeraputhiran and R. Venkataraman (2010) Investigated the removal of fluoride from the water using *cynodon dactylon* as adsorbent. By conducting batch adsorption studies at constant temperature (25-32°C), the maximum removal of fluoride was 83.77 % while keeping 3.0 mg/L fluoride concentration and 1.25 g dosage of adsorbent at neutral pH., contact time 105 minutes. Various adsorption isotherm models were applied to evaluate the adsorption data. The adsorption of fluoride ions followed Redlich-Peterson isotherm as well as

Langmuir isotherms. The authors concluded that the adsorbent used in this work shows superior adsorptive efficiency than previously studied defluoridation works using natural adsorbents was most effective[43].

S.V. Ramanaiah, S. Venkata Mohan, P.N. Sarma (2007) Contributed result from the study of adsorptive capacity of waste fungal biomass (*Pleurotus ostreatus* 1804) in relation to fluoride removal. Batch sorption studies were performed and the results revealed that biosorbent demonstrated the ability to adsorb fluoride from aqueous phase. The sorption interaction of fluoride onto non-viable fungal species obeyed the pseudo-first-order rate equation. Isothermal data fitted well with the Langmuir isotherm adsorption model. Fluoride sorption was found to be dependent on the aqueous phase pH and the fluoride uptake was observed to be greater at lower pH. The maximum fluoride removal efficiency of 68% was achieved at pH 2 at contact time of 240 minutes [44].

V. CONCLUSION

This paper provides an overview of various waste materials as adsorbents used for the effective removal of fluoride from water. Most of the adsorbents performance is depend on parameters such as pH, contact time, adsorbent dose and temperature. The removal capacity increases by increasing dose of the adsorbent and decreasing size of the adsorbent. From the overview it is observed that defluoridation might be feasible with waste materials as adsorbents but there is need of more studies to establish waste adsorbents as non conventional potential source of defluoridation and also to make this technique more users friendly. It is hoped that it will encourage even more rapid and extensive developments for the treatment of fluoride from aqueous phase.

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