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Various low cost biodegradable and synthetic adsorbents for defluoridation of drinking water

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Abstract

A comprehensive review of the literature reveals that fluoride removal through adsorption onto various materials is most promising in terms of cost of the medium and running costs, ease of operation, adsorption capacity, potential for reuse, number of useful cycles, and possibility of regeneration. Adsorption processes involve the passage of the water through a contact bed where fluorideis removed byion-exchange or surface chemical reaction with the solid bed matrix. In the past few years, surface adsorption is the most interesting area of research for removing fluoride from water. This method is frequently used in large scale by many water treatment plants as it can operating flow rates, can produce high quality of water without the generation of sludge, and removes contaminant from water very effectively. Several adsorbent material shave been tried in the past to identify an efficient and economical de fluorinating agent. Activated alumina, activated carbon, activated alumina-coated silica gel, calcite, activated saw dust, activated coconut shell carbon-activated fly ash, groundnut shell, coffee husk, rice husk, magnesia, serpentine, tricalcium phosphate, bone charcoal, activated soil sorbent, carbon, defluoron-1, defluoron-2, etc. are different adsorbent materials reported. The most commonly used adsorbents are activated alumina and activated carbon. The fluoride-removing efficiency of activated alumina gets affected by hardness, pH, and surface loading (the ratio of total fluoride concentration to activated alumina dosage). The adsorption process can remove fluoride up to 90%, and the treatment is very cost-effective. Regeneration is required after every 45 months as effectiveness of the adsorbent for fluoride removal is reduced after each regeneration cycle. Recent analysis of edulabad ground water performed at our Engineering Lab revealed that the fluoride concentration is 3.9ppm which is far greater than the WHO limit of < 1.5ppm. As social responsibility of suggesting economical and simple techniques of defluoridation for the residents, we have taken up the project of defluoridation techniques using natural, biodegradable and synthetic adsorbents.

Keywords: defluoridaton, biodegradable and synthetic adsorbents

Introduction

Traditional Techniques adopted for defluoridation of water: The fluoride removal from drinking water and wastewater has been successful by different techniques like coagulation and precipitation, adsorption, ion exchange, membrane separation, dialysis, electrodialysis, electrocoagulation and so on. Adsorption process for defluoridation has favored for the most part in developing countries as it is techno-economical viable method, environmental friendly and straightforwardness in operation.

The various adsorbents including activated carbon, activated alumina, metal oxides-hydroxides, bone chars and agricultural and industrial wastes were attempting to keep up the standard limit of fluoride concentration in drinking water by adsorption. Usage of plant materials and agricultural wastes as adsorbents are most adopted in developing country like India in light of the fact that they are low cost materials, available in abundance, biodegradable and natural existence. The industrial byproducts are additionally needed to reuse them in advantageous ways for defluoridation as accessible in huge amount and economical. One of the most popular techniques for defluoridation that is used in India is Nalgonda technique. In this technique, calculated quantities of alum, lime and bleaching powder are mixed with water followed by flocculation, sedimentation, filtration and disinfection. Disadvantage of this technique is that treated water has high residual aluminium concentration (2-7 mg/L) than the WHO standard of 0.2 mg/L. Alum coagulant can be used to remove fluoride selectively from aqueous solutions. Even at 60-70% removal, the left over fluorides were within the permissible limit for drinking water. Aluminum in drinking water poses possible risks to humans. Aluminium is strongly neurotoxic and may be involved in the development of Alzheimer’s disease.

The present review briefs the procedure and removal efficiency of various low cost bio-adsorbants like coconut shelt carbon, corn cob, groundnut shell, tamarind seeds and drum stick seed pulp.

These materials are available in huge amount, mostly biodegradable in nature are inexpensive and environmentally friendly disposable.

Fig 1: various low cost bio-adsorbents

Deflouridation Using Various Adsorbents

Coconut shelt carbon

The adsorbent is prepared by heating Coconut shelt at 400^0C in a furnace with controlled air supply. The obtained charcoal was activated with 25% NaOH, 25% $CaCl_2$, 2M H_2SO_4 and 2M HNO_3 respectively for 12 hours. The charcoal was then washed thoroughly with deionized water, oven dried and grinded to get granular activated carbon that was finally sieved at 4.75mm,2.36mm, 1.18mm, 600µ, 300µ and 150µ sieve sizes for the purpose of meeting different particle sizes. 58.4%, 14.2% for NaOH activated carbons, 23.8% for Nitric acid activated carbons, 18.7% for calcium chloride activated carbons and 3.33% for commercial purchased activated carbon [6].

The adsorbent was prepared by burning the coconut fiber in muffle furnace at 423 K for one and half hours. The ash was washed with distilled water, dried in sunlight and then dried in oven at 353 K for overnight. It was sieved through mesh size of 150 mm and kept in plastic air tied container for further use. This material considered as coconut fiber ash (CFA). The aluminum impregnation done by taking 100 g CFA, 500 mL of 0.6 M aluminum sulphate solution added and stirred at 180-200 rpm with 1.0 M sodium hydroxide solution till pH reaches 5-7.In this process, the addition of sodium hydroxide is very vital and it was controlled by proper checking of pH of the mixture. Desorption study with AICFA showed that the fluoride removal efficiency is 98% at pH 12. Furthermore, the coexisting anions had significant effect on fluoride adsorption. The relative performance of fluoride removal in the presence of anions increased in the order PO4 < SO4 < Cl < NO3 [7]

The adsorbent Zirconium ion impregnated coconut fiber carbon (ZICFC) was obtained by stirring 5% $ZrOCl_2$ solution (pH 1.6) with coconut fiber carbon (CFC) at room temperature (298 K) for 5 days. The impregnated adsorbent was subsequently filtered, rinsed, and dried in an oven at 333 K. ZICFC has its fluoride removal efficiency at 99.2% and 97.4% at pH 4 and 7 respectively8

Coconut Shell Activated Carbons (CSAC) in granular form with the average particle size of 2.26 mm diameter were bought from KWHB Company, and used directly without any further grinding and sieving. Highest removal efficiency was 94% that was achieved at the pH of 12 [9].

Corn cob

The adsorbent was prepared by micronizing the corn cobs to required size. IS sieve of size 63micron is provided at the primary course of filter media, in which micronized corn cobs powder is placed. The sample water is passed through these fabricated filters and analysed. The fluoride removal efficiency 81% [10].

A serial filtration frame was constructed to adopt layers of different filter media in a single portable laboratory setup. The frame used Corncob pieces, Corncob powder, Corncob charcoal and Lightweight Expanded Clay Aggregate (LECA) balls in consecutive filter layers respectively from top to bottom. Each layer is provided with quantitative filter paper also called as ash-free filter paper for better filtration. The fluoride removal efficiency70- 83% [11]

The adsorbent was prepared by deseeding the corn cobs and drying for 48 h in sunlight. Then the corn-cobs were powdered in an electric grinder to mesh size of 30-60. 5.0 g of the powder was washed with 100 mL distilled water to remove surface adhered particles, which was later kept in contact with 100 mL solution containing 1.0 g of $AlCl_3$ for 24 h. The solution was filtered and the solid material obtained was washed with 200-250 mL distilled water. The resultant powder was dried again at 900-100°C in the oven. Another adsorbent using the above procedure was prepare by treating with calcium chloride instead of $AlCl_3$.1.0 g of the powder was found to retain 0.193 g of Al and 0.35 g of Ca in the case of $AlCl_3$ and $CaCl_2$ respectively. The prepared powder was used in further experiments. The IR spectra of corn cobs powder showed the presence of hydroxyl and carbonyl groups. aluminium treated corn cob is up to 88% for 120 minutes and that of Calcium treated corn cob is up to 95% [12].

The adsorbent was prepared by drying corncobs and crushing them to granules of 1.2-0.7 mm grain sizes (CAAG).Impregnating the corncorb granules (1.20.7 mm grain sizes) was done by dissolving 25g of KOH pellets in 1 liter of distilled water, mixed with 100g CAAG, agitated on a magnetic stirrer at 80 for 1 hour. The pH adjusted to 7 and dried for 24 hours at 110±50C.The material is kept in the muffle furnace for carbonization process at 600 for 2 hour. After two hours of carboneous process the materials were taken out, cooled and then activated by keeping in a beaker which contains 3N HCl, for 30 min at 80 on a magnetic stirrer, after this once again the said material is washed with distilled water for 4 to 5 times or till one get pH of washed water near to7. Then the material is kept in oven for 12 hours under 110± 50C for drying 92% [13].

Groundnut shell carbon

The adsorbent was prepared by collecting groundnut shell with silica content, washed with water and dried in oven for 24 hrs at 110^{0}C.The dried seeds were grinded and sieved in 75µm mesh size particles. 92.8% was obtained with the dose of 4.5g/L [14].

The adsorbent was prepared by crushing 100 g of groundnut shell material, heating for about 3 h in a low temperature muffle furnace at 300-400^{0}C, at which all of the material was completely carbonized. The carbonized material was then taken out of the muffle furnace, cooled, powdered and kept in a beaker of 2 L capacity and 200 mL of concentrated sulphuric acid was gradually added to it, stirring the contents of the beaker continuously to ensure thorough mixing. The beaker was kept in a hot air oven at 100^{0}C for 5 h. The activated charcoal was then cooled and left overnight and washed free of acid and dried at 110^{0}C for 2 h, then sieved using 40 and 100 meshes 99% [15].

Ground nut shell carbon (GNSC) was prepared by taking 100 g crushed ground nut shell, kept for 3 h in a low temperature muffle furnace at 573– 673 K. This carbonized material was taken out of the muffle furnace, cooled, powered and 23 kept in a beaker and 200 ml of concentrated sulphuric acid was gradually added to it, contents were stirred continuously to ensure thorough mixing. GNSC was then cooled 2 and left overnight and washed free of acid and dried at 383K for 2 h, then sieved using 3 various mesh size. Subsequently, the obtained material was further used as the desired adsorbent for impregnation. Zirconium ion impregnation was carried out by addition of 5 5% $ZrOCl_2$ solution and GNSC (solution/solid ratio = 2:1) and the mixture was kept for 3- 6 days at room temperature (298 K). The zirconium impregnated carbonized ground nut shell recorded a maximum percentage of fluoride removal, 84.00%, when compared with carbonized ground nut shell which showed fluoride removal of 63.67% [16].

Tamarind seeds

The adsorbent Tamarind fruit covers (TNFC) were prepared from the ripe tamarind fruit. It was washed with distilled water for discoloration and dried in an oven at 100 ^{0}C for half an hour. The dried biomass was powdered and sieved to get produced uniform sized powders. Half of the virgin biomass was pretreated with 1M HCl in order to examine the possible increase in adsorption capacity of TNFC. Fluoride adsorption investigations were made on TNFC (virgin) as well as TNFC (treated). The pre-treatment of TNFC is carried out to increase the metal uptake efficiency. The virgin TNFC was soaked in 1 M HCl for 24 hrs and kept on the water bath (70°C) for half an hr. It is cooled and is neutralized with 50 ml of 1N NaOH. Finally it was washed with distilled water several times and dried in an oven at 80ºC for 6 hrs and cooled at room temperature in desiccators. The fluoride removal percentage of virgin TNFC is 74% and that of acid treated THFT is 80% respectively [17].

The adsorbent was prepared from tamarind seed that were washed well with tap water and then with double distilled water, dried in an air oven at 110^{0}C for an hour, micronised in a flour mill and sieved to get particles of sizes 75, 150 and 300 meshes ASTM. Desorption studies were carried out by agitating 100 mg of fluoride-loaded tamarind seed in 50 ml of 0.1 M HCl for a period of 2 hrs. The fluoride removal efficiency 90% [18].

The shell was removed from ripened tamarind fruit, washed thoroughly with distilled water and completely dried under sun. Then this substrate was powdered, sieved with a standard screen mesh no. 52/75 to get a uniform sized powder. Dip in bags made up of cotton cloth containing 25 mg of absorbent were prepared and dipped in each sample of 100 ml drinking water for overnight and then the cloth bag was removed in the morning from the plastic container. The tamarind fruit shell gave slight brown colour to the water sample which was decolourized by adding hydrogen peroxide (H_2O_2) drop by drop (around 8 – 10). The fluoride removal efficiency 50% [19].

Tamarind seeds

The adsorbent was prepared were reported by taking air dried tamarind fruit shell, sieved with standard screen mesh no. 52/75, followed by chemical digestion using both acid and alkali treatment. The acid treatment incurred heating the tamarind shell powder with 1N aqueous solution of HNO_3 for 20 min followed by washing with distilled water until the entire colour had been removed. The alkali treatment involved heating the tamarind shell powder with 1N aqueous solution of NaOH for 20 min, again followed by washing with distilled water until the entire colour had been removed. The adsorbent was dried to remove the moisture content. The fluoride removal efficiency 85% [20].

The aqueous extraction of drumstick seeds was prepared by using 200ml of tap water and 25 g of seed powder, mixed by a magnetic stirrer for 60 minutes and settled for 20 minutes. This aqueous extract is finally filtered through 20µm paper filter. The best coagulation condition was reached using coagulant dose of 2.5 g/L with 10 mg/L of fluoride in the water 92% [21]

The adsorbent was prepared by taking 40 gm of drumstick seed powder treated with 400 ml of 1N HNO3 for acid treatment and 0.5N NaOH for alkali treatment. The mixture was boiled for about 20 minutes. Washing of the powder sample was carried out by using distilled water until maximum color was removed and clear water was obtained. Finally, it was dried again in an oven at 50°C for 6 hrs The fluoride removal efficiency 75-80% [22].

Dry drumstick seed powder

The dry drumstick seed powder was prepared by adding ethanol. This was mixed with a magnetic stirrer for 30-45 min and subsequently separation of the residue from the supernatant was done by centrifuging for 45 min at 3000 rpm. The supernatant was decanted and the residual solid was dried (seed cake) at room temperature. The supernatant containing oil and ethanol when mixed with potassium hydroxide catalyst, the chemical reaction produces biodiesel and glycerol. Aqueous seed cake extract was prepared by using 200ml of distilled water and 25 g of MO seed cake powder, mixed by a magnetic stirrer for 60 minutes and settled for 20 minutes. MO aqueous extract is finally filtered through 20µm paper filter92.5% [23].

Dry moringaoleifera (drum stick) pods were collected, pod shells were removed manually; kernels were grounded in a domestic blender and sieved through 600 micron stainless steel sieve 95.5 [24].

Activated alumina

Down-flow column studies further demonstrated the practical applicability and effectiveness of activated alumina in removing fluorides from an initial fluoride concentration of 5.0 mg FL-1at a flow rate of 955 L/h/m-2 to the permissible limits of < 1.0 mg FL -1. Experimental studies were also conducted toinvestigate the household applicability of activated alumina for removal of fluorides by filling the candles of domestic (candle) water filters with AA and found to be effective. permissible limits of < 1.0 mg FL-1 [25].

Carbon nano tubes

The broad pH range of 6.0-9.0 with high adsorption capability suggests that carbon nanotube supported alumina have great potential applications in practice.

Zeolites

The X-ray diffraction profiles show that zeolite structures are stable during modification processes. The Si/Al ratio of composites decreases as the amount of AO is increased due to increase in Al content. The PZC of STI, initially at 2.5, increased to 9.2 in STI-AO. Textural properties of ZY and ZY-AO proved to block certain microporosity by the AO. Optimum conditions to reduce an initial fluoride concentration of 10 mg L^{-1} to below the WHO guideline value of 1.5 mg L^{-1} were 2 g L^{-1} for the 2:1 systems and 4 g L^{-1} for the 6:1 system.

Biopolymer-based adsorbents

The re-moving fluoride from water using low cost adsorbents suchas Acacia Catechu, Cuminumcyminum, Coensvettriverodies, Eletariacardamomum, Phyllanthusembilica, Strychnospota-torum and Tamarindusindica.

Conclusion

Recent analysis of edulabad ground water the fluoride concentration is 3.9ppm which is far greater than the WHO limit of < 1.5ppm. As social responsibility of suggesting economical and simple techniques of defluorination for the residents, we have taken up the project of defluorination techniques using Various Low-cost Biodegradable and Synthetic Adsorbents for Defluorination of Drinking Water natural, biodegradable adsorbents.

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