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DEFLUORIDATION OF DRINKING WATER BY VARIOUS BIODEGRADABLE ADSORBENT –A REVIEW

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Abstract:
Most of the developing countries like India depend on the groundwater as source of drinking water that may be contaminated by natural sources or by industrial effluents. One such contaminant is fluoride that possesses adverse effects on human health. When people consume water having fluoride (F^-) concentration >1–1.5 mg/L for a long period of time, various ailments that are collectively referred to as fluorosis occur. Various defluoridation technologies are being used to remove fluoride from water but still the problem has not been rooted out.
This paper describes the review on the utilization of agricultural wastes, plant biomass and readily available biodegradable materials for adsorption of fluoride from aqueous solution under different experimental conditions and their practical defluoridation efficiency.
Key Words: *Defluoridation, coconut shelt carbon, corn cob, cashewnut shelt carbon, tamarind seed pulp, drumstick seeds.*

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INTRODUCTION:

Water is equated to life as it regulates all body functions. Even though 75% of earth surface is covered with water only 0.3% is potable. Due to the pressure of human activity, urbanization and industrialization the ground water sources are degrading gradually; therefore pure, safe and healthy drinking water is a matter of concern. Access to fresh water is a human right, yet more than 780 million people, especially in rural areas, rely on unimproved sources and the need for finding ways of treating water is crucial. Some elements are essential in trace amount for human being while higher concentration of the same can cause toxic effects. Fluoride content is one such important factor that has adverse effect on human health. Due to rapid urbanization and growth of modern industries (anthropogenic source of fluoride) as well as geo chemical dissolution of fluoride bearing minerals fluoride concentration is increasing in the environment including water resources. Permissible limit of fluoride as per WHO [1-3] guidelines for drinking water is $\leq$ 1.5 ppm. American Public Health Association and as per Bureau of Indian Science specification permissible limit is 1.2 ppm. Fluoride is more toxic than lead, it also reduces IQ in humans. Traces of fluoride in minute amounts is an essential component for bones and for the formation of dental enamel in animals and human but its high concentration more than 1.5 mg/l causes irreversible demineralization of bones and tooth tissues i.e. skeletal and dental fluorosis, damage to the brain, harmful effects on kidney and liver, headache, skin rashes, bone cancer and even death in extreme case.

In India, endemic fluorosis affects more than one million populations and is a major problem in 17 of the 29 states. Similar health problems due to high fluoride content in ground water have also been reported worldwide and it is estimated that around 260 million people are adversely affected in 30 countries of the world. The Bureau of Indian Standard which is the main regulatory agency for drinking water in India specifies that the desirable limit of fluoride in drinking water is 0.6- 1mg/L.

Fluoride – An Indian Scenario

India is one among the 25 nations in the world, where fluoride contaminated groundwater is creating health problems. A report of UNICEF (1999) [4] confirms the fluoride problem in 177 districts of 20 states of India (WHO 2004).Andhrapradesh and Telangana are two of the fluorosis endemic states. Today India is the seventh largest, second most populous country in the world with a land mass of 3.29 million square km with more than 1.04 billion people [Planning Commission report]. Total population of India will exceed 1330 million in 2020 with water consumption expected to move up by 20-40%. India possesses 16% of the world's population but just 4% of its water resources. Though, surface water in India is scarce and groundwater is deep and difficult to reach (UNICEF 2002), almost 90% of drinking water needs are met with from groundwater only. The scarcity of groundwater and presence of excess fluoride can be treated as the two most critical issues in the Indian systems of sustainable drinking water supply.

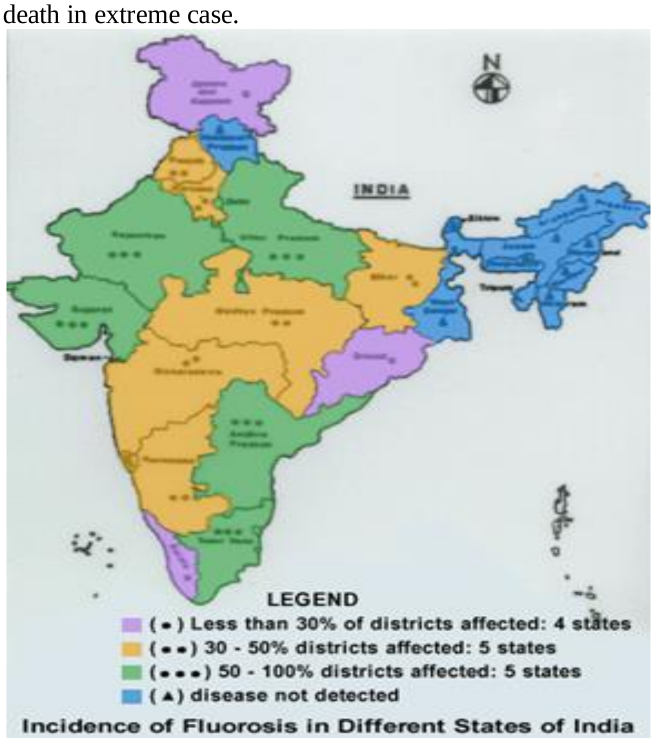

Fig 1: Fluoride contamination in India

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, electro-dialysis, electrocoagulation and so on. Adsorption process for defluoridation [5] has favoured 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 by-products 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.

Deflouridation using coconut shelt carbon:

Mateso Said et al [6]

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.

The fluoride removal efficiency of sulphuric acid activated carbon was 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.

Jayanta KumarDatta et al[7]

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 $PO_4 < SO_4 < Cl < NO_3$.

Satish et al [8]

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 respectively.

Cecilia Rolence et al[9]

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.

Deflouridation using corn cob:

Lavanya H D et al [10]

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 was 81%.

Sarayanna et al[11]
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 efficiency was around 70- 83%.

S. Parmar et al[12]
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.
Fluoride absorption efficiency of aluminium treated corn cob is up to 88% for 120 minutes and that of Calcium treated corn cob is up to 95%.

C.B.Shivayogimath et al[13]
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 (1.2-0.7 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±5^{0}C.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± 5^{0}C for drying.
Experimental data on batch study reveals that CAAG shown maximum fluoride removal of 92 %.

Defluoridation using groundnut shell carbon:

Buddharatna Godboley et al[14]
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.
The highest defluoridation capacity of 92.8% was obtained with the dose of 4.5g/L.

C Janardhana et al[15]
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.
The fluoride removal efficiency is around 99%.

Dr. G. Alagumuthu[16]
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%.

Defluoridation using tamarind seeds:

Phani kumar et al[17]
The adsorbent Tamarind fruit covers (TNFC) was 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.

Murugan et al[18]
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 defluoridation efficiency was 90%.

Anagha et al[19]
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).
50% reduction was seen in fluoride concentration by air dried powdered tamarind.

Ramanjaneyulu et al[20]
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 maximum adsorption of fluoride for tamarind fruit shell powder was found to be 85% at pH 2.

Defluoridation using drum stick seeds:
Ravikiumar et al[21]
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, achieving this way 92% of fluoride reduction in the treated water.

A.S. Parlikar et al[22]
The adsorbent was prepared by taking 40 gm of drumstick seed powder treated with 400 ml of 1N HNO_3 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.
Reported fluoride percentage removal is between 75 to 80% at optimum pH of 10.

Ravi kumar et al[23]
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 filter.
The defluoridation efficiency is 92.5%.

Anandu aravind et al[24]
Dry moringa oleifera(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.

The peak removal efficiency of 98.1% is obtained for a bed depth of 4cm and contact time of 30 minutes.

CONCLUSION:

Most of the countries depends on the groundwater as drinking water .This paper describes the review on the utilization of agricultural wastes and plant biomass as low cost materials and readily available biodegradable materials for adsorption of fluoride from aqueous solution under different experimental conditions and their practical applicability on field by column study. The efficiency of fluoride removal increases with increase in dose and smaller particle size of adsorbent. The modification of adsorbent with suitable chemicals or composite adsorbents was also remarked for the enhancement of efficiency of fluoride removal from water. The equilibrium studies were fitted well either with Langmuir or Freundlich isotherm model. The adsorption kinetics were suitable to pseudo-first order or pseudo-second order with or without intraparticle diffusion model depending on types of adsorbent used for fluoride uptake. The nature of the reaction was in term of spontaneous or non-spontaneous as well as exothermic or endothermic depending on adsorbent tried for defluoridation. The column performance of some adsorbent was also explained for field trials. The future research should be focused on the economy and efficiency of industrial and agricultural wastes to replace commercial adsorbent for developing countries like India.

Megha Institute of Engineering and Technology for Women is situated in Edulabad village, Ghatkesar Town. Recent analysis of edulabad ground water performed at Environmantal 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 adsorbents.

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