A Short Review: Various Approaches For The Elimination Of Fluoride From Groundwater

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Abstract:

The existence of fluoride in humans trigger multiple health complications. There is a need to *extract fluoride-ions from water to decrease these health complications. This paper explains the elimination of fluoride from ground water by various processes, such as coagulation & precipitation, membrane filtration, ion-exchange, electro-coagulation, adsorption process, etc. Due to the high establishment or supporting price, membrane filtration and ion exchange techniques are not frequent among these processes. In the numerous methods used for water de- fluoridation, the adsorption process is commonly used which delivers reasonable outcomes and is more desirable for all accounts for removal of fluoride in terms of quality, outline simplicity andactivity.*

Keywords: Adsorption, Defluoridation, Fluoride removal, Ground water, Ion exchange, Membrane filtration.

Introduction-

The world's demands for water are fulfilled by land and groundwater reserves and supplies. Groundwater related risks have been rising on a regular basis owing to the increase in community and demands. As the need for groundwater for residential, industrial and agriculture requirementshave risen, the burden on this resource has become immense. In certain areas on earth, groundwater is regarded as the main resource of drinking water. Generally, citizens use freshwater without anychemical and physical preparation for drinking purpose & household uses, mainly cooking. However, owing to the absence of a treated water piped delivery in certain areas of developed nations, this activity should not be prevented. Groundwater has been reported with a fluoride content above the allowable level in many areas of the world. As established by the WHO, the value is 1.5 mg/l. The WHO reported in 1984 that more than 260 million citizens worldwide drinking water having fluoride content greater than 1 mg/l (WHO, 1984).

Most citizens impacted by elevated ground water fluoride concentrations exist in tropical countries where the intake of water is more attributable to the prevalent atmosphere. People drink 3 to 4 liters of water in areas such as Ghana, which is significantly greater than the WHO average of 2 l/adult/day [1]. In these areas, the chance of fluorosis is greater. However, the prevalence of fluorosis has been identified in individuals residing in many other areas of the world. In the two densely populated countries in the world, namely India and China [2], the severity of the fluorosisissue is quite serious. In certain instances, the host rocks that are naturally abundant leads to increased fluoride in the groundwater. The intensity of fluoride rises as a consequence of rock water contact, long residence period and evapotranspiration.

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Most reports suggest a rise in the concentration of fluoride in groundwater with an increase in land surface depth [3]. This, though, is not always popular [4]. High-fluoride groundwater geochemistry is sometimes correlated with acidic to alkaline pH, lower calcium and high amounts of sodium and bicarbonate. There are alsoelevated nitrate events in certain situations where high fluoride levels are found in groundwater [5]. Saxena and Ahmed (2001) [5] suggested that alkaline circumstances of pH between 7.6 and 8.6 are suitable for the degradation of fluorite minerals from the host rocks. Waters of the sodium bicarbonate form are characteristic of high fluoride water. Fluoride shows a favorable sodium and bicarbonate interaction, while the inverse calcium relationship is expanded. Handa (1975) found that groundwater with regard to fluorite is usually undersaturated, although in certain situations it is saturated or over-saturated [6].

2. Methods of Removal of Fluoride

1-Coagulation & precipitation method

This process requires addition of Al-salts, bleaching powder followed by quick blending, sedimentation, flocculation, lime and filtration. Al-salt is responsible for de-fluoridation from wastewater. If the alkalinity & fluoride amounts in water rises, the dosage of aluminum salt rises too. Empirically, the dosage of lime is 1/20th of a dosage of aluminum. For accelerated settlement, lime encourages the creation of denser flocks. Bleaching powder is applied to raw water for disinfection at a dose of 3 mg/l [7]. Many mechanisms this process is not reliable in higher fluoride concentrations in drinking water.

A limestone reactor of two columns has been constructed by Wang and Reardon to decrease the fluoride contamination in waste water [8]. In one analysis, it is stated that the original fluoride intensity of 109 mg/l was decreased by this method to less than 4 mg/l. In this process, the fluoride in the first section is precipitated. While in first section the calcite dissolves and precipitation is observed in the second section.

A basic technique named "the Nalgonda technique" was developed by Bulusu et al.[9] to extract fluoride in drinking water. This approach includes introducing chlorine, alkali, aluminum-sulfate and aluminum-chloride, agitation or combining, and precipitation to eliminate fluoride from drinking water. Unless the fluoride content is over 20 mg/l, this method is reliable. Banuchandra and Selvapathy [10] stated that by incorporating alum and lime, the amount of fluoride can be decreased to 20 mg/l from 1 mg/l. In conjunction with poly-aluminum chloride and poly-acrylic acid at decreased dosages, Chang and Liu [11] studied coagulation flocculation of calcium fluoride precipitates. Nath and Dutta [12] have shown that an inclusion of fluoride precipitation and adsorption can be more efficient.

1) Membrane filtration process

The method of membrane filtration is best utilized for the defluoridation of groundwater, wastewater disposal and desalination of sea water [13]. In the method of membrane separation, particles are separated using exceptionally composed semi-permeable membranes on the basis of their molecular size and form. A thin, non-porous or porous polymeric paper, ceramic or metal substance, or sometimes a liquid or gas, are the available semi-permeable membranes. Ideally, the membrane must not degrade, split or disintegrate [14]. Reverse osmosis, nano-filtration, Donnan-dialysis and electrodialysis are the most widely used membrane isolation methods for fluoride elimination.

a. Reverse osmosis (RO)

In this process, water is pushed into a semi permeable membrane under pressure and solutes are separated from the solution. 85-95 per cent of fluoride may be extracted from water through this method. The RO is the safest way to eliminate both fluoride and arsenic, but it has higher wastage by 20-40 percent, heavy electricity use and high capital costs. In certain instances, treatment may be required for pre-filtration and post- pH / alkalinity change.

The usage of RO in the effective removal of fluoride and phosphate from the fertilizerindustry was investigated by Dolar et al [15]. Reverse osmosis was used by Schoeman et al. to control effluent from the stainless steel plant. Among other ions, fluoride will be easily eliminated [16]. Min et al. preferred RO for fluoride elimination [17]. Growing demands and water degradation and parallel stringency in the requirements of water quality criteria have contributed to the advancement of membrane technologies for water purification in recent years, and the issue associated with other approaches. The performance of the RO method is controlled by numerous variables, such as water characteristics, temperature, strain, periodic monitoring and maintenance, etc. Rejected water from the RO phase has been processed by Babu et al [18] using $CaCO_3$.

b. Electro Dialysis (ED)

In this process, for the isolation of ionic compounds in water, rather than pressure DC current is used. It is therefore that the water doesn't mechanically move across the ED membrane. These membranes are not formally treated as filters. Often, it would not kill particulate matter. 85-95% fluoride may be extracted from water through the ED method [19, 20]. The ED methodology depends on our existing method of the membrane-based separation processes. However, there are major water loses in the context of brine discharge, high electricity usage and high capital costs, which are very expensive. A pre-filtration and post-pH/alkalinity modification requirement can occur. Chemical handling facilities and professional operators are needed for membrane maintenance. On the basis of industrial and economic evidence, an economic assessment of fluoride removal by electro-dialysis was carried out by Lahnid et al [21].

c. Nano filtration membrane process

The latter breakthrough of all the membrane filtration methods used for water de- fluoridation is in the form of nano-filtration (NF). In the midst of NF & RO membraneprocess, the basic difference is, that NF has significantly greater pores than those usedfor reverse osmosis and provides fewer tolerance to both solute and solvent [22]. Nonetheless, the permeability of the nano-filtration membrane is relatively high than RO membrane, rendering the desalination efficiency of NF stronger for certain brackish water [23]. 99 percent of salt existing in water was discarded in RO membrane separation, causing the elimination of all fluoride ions, whilst the method of NF separation process produces incomplete water de-fluoridation and optimum fluoride content in water may be obtained through adjusting the conditions of activity.

Diawara et al. reported that in some areas of "Senegal", the nano filtration membrane has provided a fluorine retention rate ranging anywhere between 63.3% and 71%. NF- 90 used for suitable de-fluoridation for brackish ground-water in southern Morocco (Tan Tan City) [24] by Pontie et al. Nano-filtration (NF-90) and RO were used by **Bejaoui et al.** for the removal of fluoride ions. Fluoride retention in both membranes was greater than 90%. The Spiegler Kedem model was developed to evaluate the membrane (r) reflection coefficient and ion (Ps) solute permeability coefficient [25].

2) **Ion-exchange process**

Solid base exchange resins either extract the fluoride along with anions on the hydroxyl- cycle or on the chloride-cycle. With extremely simple anion exchange resins comprising quaternary ammonium functional groups, fluoride may be extracted. The elimination happens in conjunction with the following reaction:

$$\mathbf{NR_3^+Cl^- + F^- \rightarrow Matrix - NR_3^+F^- + Cl^-}$$

The fluoride-ions substitute chloride-ions of resin. The process begins after all places on resin are filled. Furthermore, the resin which is super-saturated with NaCl is used to backwash with water. Fresh chloride-ions eliminate the fluoride-ions contributing to the resin being recharged and the phase restarts. The higher electro-negativity of fluoride-ions is accountable for guiding power for the substitution of chloride-ions of resin [26]. In order to increase the ability of ion exchange, mesoporous titanium oxohydroxide was prepared by Ho et al [27], utilizing dodecylamine as a guide.

3) **Electro-coagulation process**

In this process Aluminum electrodes are used. Al dissociates at anode. However, at the cathode, hydrogen gas passes. There are different aqueous aluminum species generated during the dissociation of Al at the anode, that depends on the solution chemistry. Throughmixing with the toxins, the Al species functions as a coagulant to form large-sized flocks.

Aqueous Al^{3+} species are formed by electrolytic dissociation of Al in the anode by oxidation in water [28]. The reactions to electrodes are described below:

$$\text{Anode:} \quad \mathbf{Al(s) \rightarrow Al^{3+} + 3e^-}$$

$$\text{Cathode:} \quad \mathbf{2H_2O + 2e^- \rightarrow H_2(g) + 2OH^-}$$

The bubbles of H_2 float & thus push mechanism of flotation. In addition, the Al^{3+} ions react as follows to create a solid $Al(OH)_3$ precipitate:

$$Al_3 + 3H2O \leftrightarrow Al(OH)_3(s) + \\ OH-$$

Fluoride is absorbed by precipitated Al $(OH)_3$. With its simplest type, an electro-coagulation reactor is fabricated of an electrolytic cell with a cathode & an anode [29]. Yang and Dluhy

[30] researched fluoride elimination through producing Al adsorbent in electro-chemical reactors. The aluminum sorbent was formed through anodic dissolution of Al-electrodes in diluted NaCl solution in a parallel plate electrochemical reactor. Through depositing Al water electro-chemical method, the NaCl in the solution efficiently decreased energy usage and stimulated adsorbent formulation. Newly produced Al adsorbent can reduce the intensity of fluoride to 2 from 16 mg/l in 2 minutes. By partial neutralization of the solution to pH 6.3, the final intensity was further decreased to 0.1mg/l. In a single electrochemical reactor, sorbent production and fluoride adsorption have been implemented. This process was able to reduce the intensity of fluoride from 16 to 6 mg/l after 2 minutes of therapy and to approximately 2 mg/l after 4 minutes. In order to get the fluoride concentration down to less than 1 mg/l, the effluent from the electrochemical device requires pH modification. Using a combination electrocoagulation and electro-flotation process, fluoride ions were electrochemically separated from the solution by Shen et al. [31].

4) Adsorption

a-Bio-adsorbent

The optimum contact time for the elimination of fluoride and the optimal adsorbent dosage have been defined. Tea ash was used by Mondal et al. [32] as a biosorbent. With initial fluoride solution concentration, the de-fluoridation potential decreases, but improves with increasing adsorbent dosage and contact period. Ardekaniet al. have used Na2CO3 adjusted bagas & chitosan for the fluoride removal [33]. Fluoride elimination is optimum at pH-7 at an adsorbent dosage of 2 gm/L and achieves equilibrium in 60 minutes.

Liang et al [34] used rare-earth enhanced chitosan for de-fluoridation application. The effects of different parameters, such as initial contact period of fluoride intensity co- ion effect, and adsorbent dosage, have been observed. The existence of HCO^- and CO^- ions has significantly affected the adsorption of fluoride by this substance. Vardhan and Karthikeyan [35] examined rice husk and seed extract of Moringa oleifera (drum stick) adjusted by $MnCl_2$ and $MnSO_4$. Fluoride adsorption was studied with regard to difference of initial fluoride concentration, adsorbent dosage, involvement of intervening ions in heat treatment, and pH. Veeraputhiran and Alagumuthu [36] wereused to extract fluoride from water with biosorbent produced from Phyllantus albicato. With average particle-size, higher fluoride adsorption is obtained and an improved dosage of adsorbent improves the potential of fluoride adsorption.

a. Bone-charcoal

Bone-charcoal has potential to intensify color, odor and taste of the water. It is most suitable for eliminate fluoride from the water. The key components of bone-charcoal are 6-10% calcium carbonate, 7-10% activated carbon, 57-80% calcium phosphate. The method of fluoride reduction is primarily the substitution of fluoride for the hydroxide groups in hydroxyapatite [20].

$$Ca_{10}(PO_4)_6\,(OH)_2 + 2F^- \rightarrow Ca_{10}(PO_4)6F_2 + 2OH^-$$

Strong propensity for adsorption of fluoride, local supply and low cost of the bones of animals are several of the benefits in utilizing bone charcoal. There are also no additives applied during the procedure of de-fluoridation. Due to moral views, however, the appropriateness of animal bones as sorption materials for drinking water is restricted. Any of the challenges to this methodology being common are large initial investments and expertise required to set up bone charcoal output, maintenance and periodic filter monitoring. It has to be regenerated or substituted until the bone charcoal is saturated. Water de-fluoridation utilizing bone charcoal has been investigated by Larsen et al. [37]. Bone-charcoal disintegration pattern was analyzed and used for de-fluoridationand crystalline carbon content. The de-fluoridation of water at varying inlet fluoride concentrations and flow speeds utilizing the bone charcoal column was examined by Mwaniki et al [38]. Christoffersen et al. [39] used $CaCl_2$ & $NaHSO_4$ at pH 3.0 to recreate a bone charcoal column used for de-fluoridation. Larsen and Pearce [40] alsoresearched the de-fluoridation of drinking water utilizing a fluorapatite precipitation method.

b. Activated Alumina (AA)

The Activated alumina surface is amphoteric and may exists as $AlOH^+$, AlOH and AlO^-

. It is hypothesized that fluoride attaching to activated alumina is attributable to the interchange of groups of surface hydroxyls, that could be illustrated by the reactions given below:

$$AlOH_2 + F \rightarrow AlF + H_2O$$

$$AlOH + F^- \rightarrow AlF + OH^-$$

Due to its compact size and wide surface region, activated alumina is a strong adsorbent. The potential of activated alumina to extract fluoride varies on concentration, alkalinity, pH, particle size of fluoride in water. For de-fluoridation, this technique needs minimal contact-time for polluted water. It is possible to quickly rebuild the depleted AA. AA is available and very inexpensive on an indigenous basis. For an increased amount of regeneration cycles, the AA's adsorption efficiency is decreased. At pH 5-7, the defluoridation potential of AA is maximal. AA is pretty simple to treat. It is important to substitute the triggered alumina very often, which extends the technique. Regeneration provides a high solvent in fluoride, creating problems for disposal. It is simple to produce defluoridation filters with AA and it does not need maintenance and energy. However, the existence of HCO^- ions decreases the potential of AA47 to eliminate fluoride. A term paper was submitted by Stewart [41] on the elimination of fluoride from water using AA. HCl, NaOH and H_2SO_4 respectively. Leyva-Ramos et al. [42] examined the fluoride kinetics elimination through AA synthesized from pseudo boehmite.

Conclusion –

Fluoride in ground water is becoming a serious issue for human community. Rocks that are high in fluoride are the primary source of fluoride in ground water. In sellaite, fluorite, cryolite, fluorapatite, apatite, fluoric acid, biotite, amphibole and many other minerals, fluoride exists. For good teeth and muscles, a certain level of fluoride is important for human. where they are found beyond the permissible limit by WHO & BIS, as the concentration rises, it causes moderate dental fluorosis to debilitating skeletal fluorosis. Due to the high establishment or supporting price, membrane filtration and ion exchange techniques are not frequent among these processes. In the numerous methods used for water de-fluoridation, the adsorption process is commonly used which delivers reasonable outcomes and is more desirable for all accounts for removal of fluoride in terms of quality, outline simplicity and activity.

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