

Methods of fluoride removal-A Review

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Abstract - This article describes how the elimination of fluoride from consuming water may be carried out with the aid of using exceptional techniques, for example, coagulation-precipitation, membrane separation, ion trade, adsorption strategies, etc. Among those techniques, ion trade and membrane techniques aren't definitely widely recognized because of the excessive value of help and implementation. Two exceptional strategies are very famous in India. The Nalgonda approach is one of the broadly identified strategies generally implemented to water defluorination in growing countries. Among the unique strategies implemented to lessen fluoride in water, adsorption method is broadly implemented and offers the preferred impact and surpassed all of the payments being a greater appealing approach to put off fluoride in phrases of value, easy definition and operation. An evaluation of the literature has exposed new possibilities to put off fluoride in numerous ways. In any case, this superior technique ought to be used correctly at some point of the employer and ought to result in changes in contamination control.

Keywords: fluoride content, Defluoridation methods, health effects.

INTRODUCTION

Nowadays, the immoderate presence of ions in water assets reasons outstanding hobby and draws growing attention; so, it's miles important to get rid of too many ions which includes fluoride from consuming water. Fluoride may be found in sedimentary formations in minerals, rocks, soils, clays and anthropogenic activities [1]. Fluorine is the anionic form of fluorine. Groundwater sources, mainly borehole water, are considered the largest source of fluoride in drinking water, although water from salt lakes is known to have the highest concentrations of fluoride. Excessive consumption of fluoride ions from food and water weakens the appearance of teeth, known as dental fluorosis or mottled enamel [2]. Fluoride is a detail that

happens clearly withinside the earth's crust. It isn't always vital for the increase and improvement of human beings or different organisms. Most fluoride happens as insoluble fluoride, however ionized fluoride exists in soil and groundwater [3]. Large population observations have suggested that people who drink fluoride-rich water have better oral health and less tooth decay than those who consume less fluoride while drinking. As a result, some countries have added fluoride to drinking water as a measure to protect public health [4]. Some suggest that it is not necessary because most people get enough fluoride from other sources. These now include widely used fluoride toothpastes and mouthwashes. Fluoride is found in all-natural water sources in certain concentrations [5]. Sea water contains fluoride at approximately 1 part per million, equivalent to drinking water containing fluoride. The three main fluorinated compounds commonly used for fluoridation of water are: sodium fluoride, hydrofluorosilicic acid (hexafluoro silicic acid), and sodium silicofluoride. All of these mixes completely in water resulting in the availability of fluoride ions to prevent tooth decay [6].

Health effects

The repercussions of long-standing beverage customs on individual health in terms of fluoride quantity

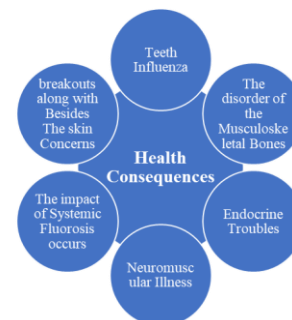


Fig.1 Health Consequences

Materials with a fluoride content of 0.7 ppm are currently considered optimal for oral health. Concentrations above 4.0 ppm can be dangerous. It does no longer have an effect on the fitness of the teeth. Early symptoms of bone fluorosis include stiffness and pain in the joints, impaired bone structure, and calcified ligaments leading to muscle weakness and pain. [1]. A rising quantity of fluoride might damage the glands that control parathyroid hormone in specific circumstances. This can lead to high thyroid hormone levels or excessive levels of parathyroid hormone output. This might result in less calcium within the bone structure and better than normal blood calcium levels. Lower calcium levels in joints make joints more prone to breaking. Fluoride levels were measured in 299 pregnant women. They tested cognitive abilities at the ages of 4, 6, and 12. Higher quantities of fluoride are linked to diminished IQ assessments. Higher quantities of fluoride are linked to diminished IQ assessments. Fluoride was identified as a neurotoxic that could harm children's development, among ten other industrial contaminants such as the elements lead, arsenic, solvent, and mercury methyl form. [4]

Non-skeletal/systemic fluorosis effects

Gastrointestinal diseases, Neurological sign and symptoms, Muscle weakness & stiffness, Skin Allergies, inflammation [2]. Urinary tract infections are arising. During pregnancy or breast feeding if the mother consumes high concentrations of fluoride can also damage a foetus. Fluoride accumulates at the erythrocyte membrane, which in flip loses calcium content material and cause low haemoglobin degrees in sufferers chronically unwell because of fluoride toxicity [7].

Furthermore, problems with health

fluoridated water also contributes to the following health problems:

1. breakouts along with various dermatological complications
2. cardiovascular heart disease, which include hardening of the arteries and calcifying, elevated blood pressure, destruction to the cardiovascular cardiac muscle
3. Reproduction-related problems that include infertility as well as premature adolescence in girls

4. problems impacting joints and bones, such as cartilage degeneration, tumours of the bones, and multiplex joint issues [4].

Fluoride content within the legal limit

There is a limitation on the level of fluoride, its effects vary with temperature.

The Government of India has initiated the National Fluorosis Prevention and Control Program (NPPCF) for the prevention, diagnosis and treatment of fluorosis in endemic areas. [1]. The World Health Organization (WHO) recommends a common standard for fluoride concentrations in water, which is also the upper limit for fluoride in drinking water in some countries. such as Canada, China, India, Australia, and Europe [8].

Different organization with fluoride limit

S. No.	Organization	Permissible limit in mg/l	Reference
1	Bureau of Indian Standards (BIS)	0.6-1.2	[1], [9]
2	The United States Public Health (USPH)	0.8	[1], [2]
3	Indian Standard Institution (ISI)	1.5	[11], [12]
4	Indian Council of Medical Research (ICMR)	1.0	[1], [12]
5	World Health Organization (WHO)	1.5	[9], [11]
6	Public Health Engineering Manual and Code of Practice, Government of India	1.0	[13], [11]
7	The international standard for drinking water	0.5	[11], [4]

The technique used international to take away fluoride from water.

Precipitation/coagulation

In this process compounds are introduced into non-boiling water precipitation of fluoride salts as insoluble fluoroapatite, it is separated from water. Precipitation is the well-set up approach but the method has simplest mild performance and the excessive chemical dose is required. Excessive use of aluminium salts produces sludge and unfavourable fitness results via aluminium solubility [14],[15]. Precipitation through flocculation is a cheap and efficient water defluoridation system that neutralizes and flocculates charged suspension particles to

sediment. Lowering the reaction temperature will destabilize the reaction and form a semi-precipitate [16]. The precipitation technique is primarily based totally at the addition of chemicals and next precipitation of soluble fluoride salts as insoluble fluoroapatite. Alum, lime, polyaluminium chloride, polyaluminium hydroxysulfate, and brushite are a number of the substances generally used in defluorination with the aid of using precipitation methods [17],[19]. Abdolmajid Fadaei studied various techniques to do away with fluoride in water, collecting the values of adsorption capability and fluorine elimination performance of various adsorbents. Coagulation is the most cost-powerful technique, sedimentation is an excessive chemical cost, and batch technique is limited in quantity and flavour [18]. M.V.V. C. Lakshmi et al. found that the operating cost of this method was low and that it could remove up to 90% of the fluoride concentration. This method removes fluoride from water. Catalytic precipitation as a promising method for the fluoridation of water. Contact rainfall is reliable and no runoff efficiency monitoring is required. Filters used for contact deposition are much more expensive [20], [21]. The performance of fluorine elimination through electrocoagulation depends on the pH, alkalinity, anions gift and response characteristics. The defluorination overall performance is set to 100%, with the sum of hydroxide and fluoride ions (-OH and F) being set to this value. The distribution of fluorine for the duration of defluorination through electrocoagulation is split into 3 parts: closing in water, eliminated through electrodes and adsorbed on aluminium hydroxide flakes [22]. The Nalgonda method for fluoridation of water involves the use of lime and alum as coagulants. Lime activates the precipitation of fluorine as insoluble calcium fluoride and raises the pH to 11-12, while alum is introduced and coagulation occurs. This is the most commonly used approach for defluorination, suitable for 20 liters of water for further use. The use of aluminium sulphate as a coagulant greatly expands the consciousness of sulphate ions, causing a laxative effect in humans [10].

Advantages

- ✓ Advantages of contact deposition are high reliability and no need to monitor flow efficiency.
- ✓ most widely used method.

- ✓ It is cost-effective, so it can be used at home and community level.
- ✓ Chemicals are Commercially available [23].

Disadvantages

- ✓ The downside is that the filters used for contact deposition are much more expensive.
- ✓ Possible water contamination due to excessive aluminium [13]. The sludge contains the removed fluorine in concentrated form and is toxic, making disposal a problem.
- ✓ Costly and efficiency depends on the presence of a pH-water combination, the need to adjust and re-adjust the pH, and increase the residual aluminium concentration [23].

Adsorption method

Modified Terminalia Chebula Barks (MTC) activated carbon was collected near the forest area of Chandrapur district, India, Maharashtra. It possesses the efficient properties needed for the defluorination process from groundwater. It is an effective, low-cost bio sorbent with good fluoride removal capacity during adsorption [24]. This technology works by adsorption of fluoride ions on the surface of the active substance. The material retains fluoride in a physical way. chemical or ion exchange mechanism. The adsorbent is saturating after some time of operation and must be regenerated. Adsorption can be performed using high performance, economical and locally available adsorbents. Inexpensive and locally available materials for use in this technique [14]. The formation of numerous fluoride complexes is an important component at the adsorption of fluoride via way of means of activated alumina from aqueous solution. Fluoride adsorption on activated alumina was probably due to electrostatic interactions between the alumina soil and the predominant fluoride species in solution and varied with Ph [6], [25]. Adsorption is divided into three types: chemisorption, physisorption, and ion exchange. Adsorption models are inversely proportional to particle size, and as the water temperature rises, the adsorption rate decreases. Continuous cycles of regeneration of fluoride-saturated aluminium oxide coated sand and charcoal (AOCC) become powerful [26]. Bone charcoal has the ability to improve the colour, taste and smell of water and remove fluoride from it. It is easy to set up and can

reduce fluoride concentrations to less than 1 mg/l. It is degenerate and not universally accepted [19]. The general characteristics of fluorine adsorption on columns adsorption capacity is highly dependent on the jet charge, inlet fluoride ions and matrix peaks, and is particularly important in the case of excessive contact time, reduced fluoride absorption. As the charge of the jet increased, the forward step curve became steeper and the decay coefficient time and recognition of adsorbed ions decreased [27], [28]. Himanshu Patel has researched ways to remove contaminants from man-made wastewater. Column adsorption studies are seen as higher adsorption due to their advantages, such as bigger, lighter, simpler, and more economical. Method parameters such as adsorbate, drift rate, mattress height, pH, adsorbent particle length, and preliminary determination of temperature have a decisive influence on this phenomenon. [29]. M. S. Sharma et al. studied the affinity of aluminium oxide for trapping fluoride ions in the defluorination process. Alumina alone cannot provide favourable adsorption properties and modified activated alumina can increase the adsorbent's defluorination capacity over the appropriate pH and temperature range. Bottom adjustment can also increase the reactivity of fluorine and hydroxide ions [5].

Advantages

- ✓ The major gain is its miles regionally to be had.
- ✓ This method can cast off fluoride as much as 90%
- ✓ Efficient and economical
- ✓ Minimum contact time required for maximum defluorination
- ✓ Locally accessible and reasonable.
- ✓ The refresh rate is much higher.
- ✓ Excellent accessibility
- ✓ budget
- ✓ Ease of operation, presence of a wide range of adsorbents [17].

Disadvantages

- ✓ Activated alumina technique has Low potential and the technique is slow.
- ✓ Its performance for elimination of fluorine os very low.
- ✓ The technique is extraordinarily pH based and works simplest in a pH variety of 5-6.

- ✓ Presence of sulphate, phosphate or carbonate outcomes in ionic competition [23].
- ✓ The technique has low adsorption potential, negative integrity and wishes pre-treatment
- ✓ Needs renewal each 4-five month
- ✓ Disposal of fluorine-containing sludge is likewise a problem
- ✓ Qualified employees are required to perform the plant.
- ✓ Appropriate manufacturers are not to be had in underdeveloped countries.
- ✓ Activated alumina wishes to be regenerated every now and then due to the fact it's far used up over time [17].

MEMBRANE PROCESS

The RO membrane technique has emerged as a preferred opportunity to offer secure consuming water without posing the issues related to different traditional methods, RO is a bodily technique. Fluoride elimination efficiencies as much as 98% with the aid of using membrane processes [6]. The technique under study is the opposite of osmotic in the hub, removing all dissolved solids on the concentrated side of the membrane. RO operates at higher pressures, nanofiltration is a low voltage technique, and electro dialysis is an inexpensive way to remove fluoride from brackish water. Techniques without chemical pre-treatment are better suited to environmental needs. [13]. Membrane stress affects the length of fouling remaining, with the amount eliminated of fluoride ranging from 45-90% and the acidity level rising from 5.5-7. Temperatures and pH become significant, increasing the volume of refuse [21].

a. Electro-dialysis (ED)

Electrodialysis, a membrane method, and d. C. are used to separate ionic contaminants from water and can remove 85-95% of fluoride from water [19]. Abdolmajid Fadai has studied various methods and tactics for removing fluoride from water, including electro dialysis, which has an 86.2% output for defluorination and an 80-90% fluoride removal efficiency. It removes 50-60% of the fluoride within 6 minutes. [18]. Electro dialysis is a technique used to remove fluoride from water and wastewater. It can remove up to 85-90% of the fluoride content but

requires excessive energy consumption and moisture loss. It requires capital investment [20].

b. Reverse Osmosis (RO)

Reverse osmosis (RO) is a convenient way to remove fluoride and arsenic from water but requires chemical treatment by specialized operators and centres to successfully remove fluoride and phosphate from fertilizer production. Water loss, strength consumption, pre-filtration, and post-treatment pH/alkalinity adjustment may be required. RO membranes have a higher rate of fluorine removal than NF membranes, with a degree of fluoride removal ranging from 45% to 90% at pH 5.5 to 7 [19], [18]. Reverse osmosis is a technique used to remove fluoride from water and wastewater. It is governed by factors such as raw water quality, temperature, voltage, and daily care and maintenance requirements. The opposite of osmosis is excessive water loss due to silica concentration in the feedwater, which involves energy consumption and capital costs [20].

c. Nanofiltration

Abdolmajid Fadaei has learned tricks and methods to remove fluoride from water, collecting values of adsorption capacity and efficiency of various adsorbents. NF is a powerful method for water treatment, removing 98% of fluoride from contaminated water [18].

Advantages

- ✓ Reverse osmosis is the best way to remove both fluoride and arsenic present in water.
- ✓ NF/RO can provide maximum defluorination because these processes use different separation mechanisms such as solution diffusion, size displacement, charge repulsion and adsorption [23].
- ✓ Treatment of highly fluorinated water with better quality compared to adsorption and precipitation processes.
- ✓ Electrodialysis can remove up to 85-90% of fluoride [21].
- ✓ The process is highly efficient for fluoride removal.
- ✓ Because the membrane has a sufficiently long service life, regeneration or replacement problems are less frequent [13].

- ✓ There's not any confirmation of disruption caused by additional charges.
- ✓ Up to 90-95% Fluoride Removal
- ✓ Helps retain the taste and colour of water [20].

Disadvantages

- ✓ Reverse osmosis methods involve high energy consumption and high capital costs.
- ✓ pH affects flux and defluorination efficiency. Prefers slightly alkaline pH
- ✓ Raising the temperature of RO/NF inhibits defluorination [13].
- ✓ Energy consumption is high and the process requires large capital investment.
- ✓ Membrane maintenance requires chemical handling facilities and skilled operators.
- ✓ Takes away all ions from water. Remineralization is necessary following therapy because certain minerals are required for growth to be optimal.
- ✓ The water is acidic and requires pH correction.
- ✓ A large amount of water is consumed in the form of brine
- ✓ Salt water treatment is a problem [20].

Ion Exchange

Ion exchange is the exchange of ions between electrolytes or solutions and can be more efficient with simultaneous exchange of cations and anions. The downsides are high cost and low pH, but it removes up to 90% fluoride and preserves the taste and luster of the water [14]. Ion exchange is a process where water passes through a column filled with ion exchange resins. Once saturated, the filter media must be regenerated with a mild acid/base solution. Fluoride removal by this method depends on water quality and cost but has been successful with Cl⁻ and SO₄²⁻ as the main ions. Small volume, absorb other anions, fluoride concentration should be less than 10mg/L [19]. A strong basic anion resin was studied to remove fluorine from water. Fluoride excretion increased with increasing exposure time and dose, but the coexisting ion had an almost opposite effect on nitrate fluoride removal [30], [20]. The ability to remove fluoride from water was studied using a strong primary anion replacement resin containing quaternary ammonium groups. Fluoride ions regenerate the resin's chlorine ions. This method is very productive. This method is very luxurious and contains low pH and excessive

chloride content in the treated water. This requires a longer response period [13]. The ability of resins to change fluorine as a function of the ratio of fluoride to the total amount of anions in water was studied. Resins can corrode water garage inventory by increasing the chloride concentration and having an excessive pH. Regeneration methods of cation exchange and anion exchange resins require large amounts of regenerant and large waste, making them complex, susceptible to infection and expensive [21], [22]. Cation exchangers/chelating resins are used to absorb fluorides, and metal-containing cation exchangers contain inorganic cation exchangers. Ion exchange favours ions of higher valency, concentration, and smaller equivalent volume of hydration, with a removal rate of 90-95% while retaining taste and colour. However, the presence of sulphates, phosphates, bicarbonates can cause ion competition, higher cost, low pH, and high chloride content [2], [10]. SV Biradar has researched synthetic chemicals, such as anion and cation replacement resins, for fluoride removal. The ability of these resins to replace fluorine depends on the ratio of fluorine to total anions in water. An ion replacement method involves sodium ions being removed from a solution using a cationic material, while hydrogen fluoride was removed throughout the second stage. Without plumbing, it is difficult to adjust the flow of water through the unit and gradual depletion of living agents is not always detected without problems [17].

Advantages

- ✓ The ion trade method can effectively get rid of fluorine from carbonated water containing Cl^- and SO_4^{2-} as principal ions.
- ✓ Up to 90-95% Fluoride Removal
- ✓ Helps maintain the flavour and colour of water [31].

Disadvantages

- ✓ The primary dangers of the ion alternate process are excessive cost, low productivity, adsorption of different anions, and the fluorine attention ought to be much less than 10 mg/l.
- ✓ The presence of different ions along with sulphate, carbonate, phosphate and alkalinity reduces their effectiveness [13].
- ✓ Resin regeneration is tricky as it generates fluoride.

- ✓ Abundant waste that desires to be handled one after the other earlier than very last disposal
- ✓ Technology is high-priced because of resin cost.
- ✓ Purified water has a totally low pH and excessive chloride content [31].

NALGONDA TECHNIQUE

The Nalgonda process is a technique developed by the National Institute of Environmental Engineering, Nagpur, India. It involves the inclusion of alumina bases and bleaching agents, followed by quick interacting, coagulation, dissolution, purification, and irradiation. Salts of aluminium can be added in the form of aluminium sulphate, aluminium chlorine, or a mixture of both. It is responsible for excluding fluoride from water sources [14]. A commonly used coagulant in water treatment, hydrated aluminium salts are used to flocculate fluoride ions in water. Lime is added because processing under alkaline is first-rate. Bleach is added for disinfection. After agitation, the chemical coagulant dissolves into flakes and settles down [32], [20]. The technique of flocculation cannot remove enough fluoride, even when appropriate dosages are used. The remedy performance is restrained to 70%, making it less effective in cases of medium to excessive fluoride infection [19]. P. Renuka et al. researched the Nalgonda technique, which is low cost and easy to process. Polyaluminium chloride has better fluoride removal efficiency than alum at higher fluoride concentrations, so large amounts of alum had to be treated with fluoride. The pH of the treated water must be carefully controlled [21]. This method of defluorination involves adding alum and lime to the fluorinated water with constant stirring in a two-bucket defluorination machine. It was used for 1.5 years in 76 households and has a basic limitation that prevents processing of lower concentrations of fluoride unless excessive amounts of alum are used. It is environmentally friendly, simple design, easy to install and operate, easy to maintain, and less requirements for highly professional operators. However, it is time consuming, has problems controlling the pH of the treated water and the pH law of the lime, additional use of alum, and improper use of low-fluoride fluoride removal chemicals [2]. The Nalgonda technique is expensive to operate and not always automated, but research was conducted to improve its efficiency for eliminating fluoride by

adding charcoal powder and aluminium hydroxide. Manpower was needed to smooth out slime before other portions of the water facility. [3]. The Family Defluoridation Guide recommends applying a 10% alum solution in a 40-litre bucket to reduce 50% of fluoride, with no side effects and a price readily available to tribal villagers [8]. The Nalgonda technique, created by NEERI in 1961, is a cheap and straightforward way to remove fluoride. Remaining aluminium should be maintained below 0.2mg/L since it can have detrimental effects on one's health. Alum and moringa oleifera seeds can be used to remove aluminium from the cleaned water. [33], [1]

Advantages

- ✓ No media regeneration required
- ✓ Chemicals are readily available for municipal water treatment.
- ✓ Its economical, Suitable for home use. [17].
- ✓ can be used to purify large amounts of water for public use.
- ✓ Easy to design, structure, operate and maintain.
- ✓ Highly efficient fluoride removal from high to desired levels.
- ✓ Less water loss and less disposal problems [13].
- ✓ Minimal mechanical and electrical equipment required.
- ✓ Household equipment requires no energy other than muscle power.
- ✓ It provides defluorinated water of uniform quality.
- ✓ In addition to turbidity from fluoride, this method also removes colour, odour, pesticides and organics [34].

Disadvantages

- ✓ More sludge formation compared to electrochemical defluorination [21].
- ✓ A large amount of alum is needed to remove the fluoride.
- ✓ The pH of the treated water must be carefully controlled.
- ✓ Nalgonda method requires daily mixing of alum and lime [31].
- ✓ within side the system is necessary.
- ✓ The everyday operations require an educated operator
- ✓ Periodic evaluation of feed and water to determine best dose.

- ✓ Some authors report high residual aluminium in purified water [34].

CONCLUSION

Attempts have been made to provide a brief overview of various methods for removing fluoride from water to acceptable limits according to various organizations such as WHO, BIS, CPHEEO and ICMR. In the absence of alternative sources of fluoride in drinking water, defluoridation should be performed. Fluoride is known for both beneficial and detrimental effects on health. Drinking water is the primary supply of fluoride intake. Many methods are used to remove excess fluoride in drinking water, and each method has advantages and disadvantages. The ability to remove fluoride varies with chemical, geographic, and economic conditions, and certain processes are appropriate in certain regions. Precipitation is the most common method, is only moderately efficient and requires large amounts of chemicals. Adsorption can be performed in a highly efficient and cost-effective manner using locally available adsorbent materials. The membrane process is a highly effective technology and does not require chemicals. However, it requires a relatively high cost and a skilled workforce. Ion exchange removes up to 90-95% fluoride. Treated water may have a low pH and a high chloride concentration. High cost is a disadvantage. The Nalgonda technique consist of aluminium salts, lime and bleaching powder. This technology is highly efficient fluoride removal from high to desirable levels, with less water loss and less disposal issues.

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