DEFLUORIDATION TECHNIQUES - A CRITICAL REVIEW

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ABSTRACT

Fluoride in drinking water plays a vital role in dental health. Due to excessive fluoride in water, enamel loses its luster. At lower concentration, it guards our teeth against cavities but at higher concentrations imparts fluorosis in varying concentrations. Excess fluoride in drinking water is reported from more than 35 countries around the globe with India and China. In countries like India, the severe contamination of drinking water with excess fluoride acquired the dimensions of a social economic rather than a public health problem triggering defluoridation research.

Keywords: Defluoridation, Dental fluorosis, Skeletal fluorosis.

INTRODUCTION

Fluoride is considered as an essential element in human because of its role it plays in bone and dentin mineralization [1]. Fluoride is the 13th most abundant naturally occurring element which is reactive and electronegative [2]. Fluoride in drinking water has a beneficial effect on teeth in low concentration. Excessive fluoride exposure can cause irreversible demineralization of bone and tooth tissue, a condition known as fluorosis, and long-term damage to brain, liver, thyroid, and kidney [3,4]. In India, 60-70 million people are affected with dental and skeletal fluorosis [5-7]. At a higher concentration effects the teeth causing dental fluorosis [8]. To remove these excess fluoride content, defluoridation technique is adapted. In 1930's several nations began to investigate the negative effect of excess fluoride in drinking water and work on methods to remove excess fluoride [9] therefore this review focus on various methods of defluoridation.

FLUORIDE METABOLISM

Approximately 75-90% of ingested fluoride is absorbed. In an acidic stomach, fluoride is converted into hydrogen fluoride, and up to 40% of ingested fluoride is absorbed from stomach as HF. High stomach pH decreases gastric absorption by decreasing the concentration uptake of HF. Fluoride is later absorbed in intestine but is unaffected by pH at its site [10]. Once absorbed in the blood, fluoride readily distributes throughout the body, with approximately 99% of body burden of fluoride retained in calcium rich areas such as bone and teeth. In infants, 80-90% of absorbed fluoride is retained, but in adults this level falls to about 60%. Fluoride crosses the placenta and is found in mothers milk at low levels essentially equal to those in blood [11]. Levels of fluoride that are found in bone vary with the part of bone examined with the age and sex of the individual. Bone fluoride is considered to be a reflection of long-term exposure to fluoride (IPCS2002). Fluoride is excreted primarily via urine [12]. Urinary fluoride clearance increases with urine pH due to a decrease in concentration of HF.

FLUROSIS IN INDIA

Yama *et al*. 1999 and FRRDF1999 noted a total of 17 out of 32 States in India are reported to have endemic fluorosis in India [13,14]. In 1987, it estimated that 25 million people were suffering from fluorosis [13]. The prevalence of dental fluorosis has been investigated in Rajasthan by Choubisa *et al*. [15]. At mean fluoride concentration of 1.4 and 6 mg/l, dental fluorosis was seen in 25.6% and 84.4% of school children and 23.9% and 96.9% of adults.

Kodaly *et al*. reported dental mottling in 76% of children in 5-10 years age group and 84% of children in 10-15 years age group in Kodabakshupally, Armpit and Sivanagiren [16]. Yama and Lata examined the prevalence of dental fluorosis in Haryana, and over 50% children were examined to be affected by dental fluorosis [17]. Meanwhile, in Andhra Pradesh, Reddy and Prasad reported dental fluorosis level as 43%, whereas drinking water fluoride concentration ranges from 1.2 to 2.1 mg/l [18]. Endemic skeletal fluorosis was reported in India. It was first observed in Andhra Pradesh bullocks used for plowing. Short *et al*. observed years later the same disease in humans in 1937 [19]. In Andhra Pradesh Reddy and Prasad found 0.2-1% population affected with skeletal fluorosis were maximum drinking water fluorosis was 2.1 mg/l [18]. The prevalence was found higher in male and increased with increase in fluoride level in the water. The severe cases of fluorosis were observed in North-West India with fluoride concentration of 0.4-19 mg/l and South India with fluoride concentration of 0.2-20 mg/l, and moderated fluorosis was reported in Central India and Deccan Province with fluoride concentration of 0.2-10 mg/l and 0.4-8 mg/l, respectively [14,20]. Increased fluoride concentrations can also be due to anthropogenic sources. Aluminum smelters release fluoride as a by-product, leading to an increase in fluoride in nearby aquatic systems [21-26]. Phosphate fertilizers contain fluoride [27], and effluent from fertilizer plants [28] and runoff from agricultural areas [29] can contain elevated levels of fluoride.

METHODS TO ESTIMATE FLUORIDE CONCENTRATION IN WATER

Several methods were reported for analyzing the total fluoride in water, but preference was given only to International or National Standard methods. The analytical method which was determined as follows [31]:
1. Ion chromatography (IC): Chemical suppression of Eluent Conductivity Method (EPA 300.0, ASTMD4327-91 and Standard method 4110E, ISO10359-1).
2. Ion-selective electrode (pH meter): Ion-selective electrode method (ASTMD1179-93B and Standard methods 4500F-C).
3. Colorimetric: SPANDNS method (Standard method 4500F-D).

Table 1 shows the Permissible limit of fluoride in drinking water prescribed by various organizations.

IC [32]

To analyze samples using IC, the calcium carbonate must be dissolved in a strong acid, such as hydrochloric acid. The carbonate is converted to carbonic acid, and then to carbon dioxide gas. As a result, the carbonate ion concentration is only slightly higher than the amount normally

Table 1: Permissible limit of fluoride in drinking water prescribed by various organizations [30]

S. No.	Name of organization	Permissible limit of fluoride ion (mg/l)
1.	World Health Organization (International standard of drinking water)	0.6-1.5
2.	US Public Health Standards	0.8
3.	The Committee on public health engineering manual and Code of practice, Government of India	1.0
4.	ICMR	1.0
5.	BIS	0.6-1.5

ICMR: Indian Council of Medical Research, BIS: Bureau of Indian Standards

found in aqueous solutions, due to dissolved carbon dioxide from the atmosphere. The resulting solution will contain calcium ions, carbonate, chloride (from the acid), and any other ions that were incorporated into the calcium carbonate structure.

Fluoride ion-selective electrodes [33]

It is used to measure a wide variety of matrices however, they are not ideal for studying fluoride in a calcium carbonate matrix. After the dissolution of the shell with a strong acid, the solution would have a pH ~1. Determination of fluoride at this pH using an electrode will be inaccurate because most of the fluoride is in the form of HF, HF2⁻, and [HF] n and will not be detected by the electrode, which only measures fluoride, [34]. This problem can be overcome by adding a total ionic strength adjustment buffer, which serves to buffer the solution at an optimal pH, releasing fluoride from complexes with captions such as iron and aluminum, and it ensures that all measurements are done at the same ionic strength [35-37]. Unfortunately, this technique does not allow for the identification of other ions present in the matrix.

The colorimetric method

The sodium 2-(para sulfophenyl azo)-l,8-dihydroxy-3, 6 naphthalene disulfonate method, is based on the reaction between fluoride and a dark red zirconium dye lake, forming a colorless complex anion (American Public Health Association [APHA], American Water Works Association [AWWA], and Water Pollution Control Federation [WPCF] 1985). This method results in a bleaching of the red color in an amount proportional to the fluoride concentration. As the amount of fluoride increases, the resulting color becomes lighter. Color then is determined photometrically using a filter photometer or spectrophotometer [38].

The colorimetric and fluoride ion-specific electrode methods are currently the most common methods employed (APHA, AWWA, and WPCF 1985).

VARIOUS DEFLUORIDATION TECHNIQUE

Fluoride at excess level in drinking water in developing country is an emerging problem. There are sources available for defluoridation of water to prevent fluorosis. There are several different types of defluoridation method. The following are defluoridation method; bone charcoal, contact precipitation, Algona, activated alumina, ion-exchange technique, membrane filtration, nanofiltration, and clay. Advanced treatment technologies are reverse osmosis (RO), electrodialysis, and distillation.

BONE CHARCOAL

Since 1940's bone charcoal is the oldest known defluoridation technique used and successfully removes arsenic from water [39].

This method of fluoride removal is still followed in the USA mainly because of its large scale use in sugar industry [40]. The first domestic defluoridators were developed in 1960's [41] and in 1988 the ICOH filter type was launched by WHO [42]. Bone charcoal is produced by calcification of animal bones or carbonizing bone at a temperature of 1100-1600 degree. Bone charcoal consists of calcium phosphate and carbonates. Using bone charcoal defluoridation technique has become simple and regenerated without significant loss of binding capacity for fluoride [43]. Today bone charcoal is replaced by ion-exchange resins

and activated alumina but at domestic level bone charcoal seems to work well as defluoridation.

Limitations [44]

1. There is no special acceptance for bone char because of some religious and cultural objections.
2. Bone charcoal harbours bacteria, and hence it is unhygienic.
3. It is technique sensitive method, the efficiency of bone char as an absorbent for fluoride is a function of charring procedure which should be done cautiously, and when the material is exhausted, the fluoride uptake is ceased.

CONTACT PRECIPITATION

Contact precipitation is a method for defluoridation which requires addition of calcium and phosphate compound and bringing water in contact along with bone charcoal medium. In a solution containing calcium, phosphate, and fluoride, the precipitation of calcium fluoride is easily catalyzed in contact bed that acts as a filter for precipitation [45]. From bed, the fluoridation water follows continuously by gravity to shallow clean water tank. The flow from the raw water tank to clean water tank is constrained by a narrow tube or valve to allow sufficient contact time in bed. The constant time of 20-30 minutes is reported to show excellent operation. The filter resistance is compared with flow resistance through tube and valve by Dahi in 1998 [46]. It was reported that this method allows high removal efficiency, has low operating cost, no overdose or any health risk and low daily working load [46].

NALGONDA TECHNIQUE

Initially, Aluminum salts are added as an alum and responsible for fluoride removal from water which caused mottling of teeth and lead to health concern in the USA [47]. Later Algona technique was developed in India by National Environmental Engineering Research Institute (NEERI). Later there was a study by Parthasarathy *et al.*, which showed a combination of calcium salts and polymeric aluminum hydroxide are used for treatment in Algona technique [48]. The advantage of using polymeric aluminum hydroxide over alum is that less concentration of former is required and results are good. The process in aluminum sulfate based coagulation-flocculation sedimentation for removal of excess fluoride from water. Aluminum sulfate is dissolved and added to water to ensure proper mixing. As a result, aluminum hydroxide micro-flocs are produced and gathered as large settling flocs. The mixture now is allowed to settle, and during this flocculation many kinds of microparticles and negatively charged ions including fluoride are removed by electrostatic attachment to flocs [49]. Another study was reported by Mameri *et al.*, suggested an efficient defluoridation process where aluminum bipolar electrodes were used [50]. The electrocoagulation process with aluminum bipolar electrodes permitted defluoridation of Sahara water without adding salts to treated water. The technique is resourceful and can be used for large communities, for small communities as well as rural and for domestic defluoridation [51]. The Algona technique at NEERI resulted in three main achievements by Bulusu *et al.* [52].

1. Understanding non-stoichiometric co-precipitation mechanism for removal of excess fluoride,
2. Knowledge on solving the treatment problems at low cost, and
3. It shows the required dosage of aluminum sulfate for given values of water and fluoride concentration.

The Algona technique is modified using poly aluminum chloride because of its efficiency is high when compared with alum and less cost, less flocculation time is sufficient for poly aluminum hydroxyl sulfate than the alum.

Limitation [53-55]

1. The Algona technique is more time consuming and is more difficult than other defluoridation technique.
2. It is difficult to regulate the pH and the correct dose of chemical to be added.
3. It removes only 18-33% of fluoride in the form of precipitates and converts 67-80% of fluoride into soluble toxic Al^{3+}-F^- ions.
4. A large dose of aluminum sulfate, up to 700-1200 mg/l is needed. A larger dose results in large sludge disposal problem in water treatment and sulfate ion concentration crosses the maximum permissible limit of 400 mg/l which can cause health problems in human beings.
5. The residual aluminum in excess of 0.2 mg/l in treated water causes dangerous dementia disease as well as the neurological problem, structural, and biochemical changes.
6. It also affects musculoskeletal, respiratory, cardiovascular, endocrine, and reproductive system.

ACTIVATED ALUMINA

Alumina is highly porous material, granular contains aluminum oxide. When water passes through a packed activated alumina, pollutants are adsorbed onto the surface of the grains. According to Hao and Huang *et al.* activated alumina is used treat water with fluoride concentration from 4 to 15 mg/g, however another study by COWI (1998) showed practically the fluoride removal capacity is 1 mg/g [56]. Therefore, there is a difference in activation of alumina and it may be due to pH. The capacity of alumina dependent on pH is 5. While it is easy to adjust pH but at the same time it is necessary to depend on actual pH of raw water in the small community and domestic treatment [56]. The activated alumina process was evaluated for fluoride removal from underground mine water in South Africa in early 1980's, and it was reported that portable water could be produced by underground mine water with a fluoridation concentration of 8 mg/l. Two 500×10^3 l/day defluoridation plants were installed as a result [57,58]. In India, China and Thailand activated alumina may be affordable for low-income communities. Activated alumina is widely used in the industrial chemical. Hence, activated alumina has become low costly and more popular where it is manufactured. Table 2 shows various researches conducted on activated alumina .

Modified activated alumina

To enhance the adsorption efficacy of activated alumina, researchers have modified alumina surface in various forms given (Table 3).

ION-EXCHANGE TECHNIQUE

For defluoridation of water various types of anion and cation exchange resins have been used. Some of these are polyanion (NCL), Tulsion A-27, Deacedite FF (IP), Amberlite IRA 400, Lewatit MIH-59, and Amberlite XE-75. These resins have been used in chloride and hydroxy form. The fluoride exchange capacity of these resins depends on the ratio of fluoride to total anions in water. The capacity of Amberlite XE 75 was found to be approximately 88 g/m 3 when fluoride to total anion ratio was 0.05. The capacity increased with increasing ratio. Polyanion removed fluoride at the rate of 862 mg/kg and 1040 mg/kg with an initial fluoride concentration of 2.8 and 8.1 mg/l, respectively. Deacedite FF (IP) and Tulsion A-27 could treat 2270 L and 570 L of water bringing fluoride level from 2.2-1.0 mg/l [51]. This technique helps to remove 90-95% of fluoride. It helps to keep the retention of taste and color of water intact.

Limitations [99]

1. Its efficiency is reduced in the presence of other ions such as sulfate, carbonate, phosphate, and alkalinity.
2. A large volume of regenerate is required for regeneration of cation and anion exchange resin.

3. Regeneration of resin is a problem because it leads to fluoride rich waste, which has to be treated before final disposal.
4. The method is expensive because of the cost of resin.
5. Treated water has very low pH and high levels of chloride.

MEMBRANE FILTRATION PROCESS

RO and electrodialysis are two membrane filtration processes which can be used for the removal of fluoride. Similarly, use of RO membranes for fluoride removal from contaminated water sources has also been reported.

RO: In RO, the hydraulic pressure is exerted on one side of the semi-permeable membrane which forces the water across the membrane leaving the salts behind. The relative size of the pollutants left behind depends on the pressure exerted on the membrane. Recent work by Fox KR, (1981) and Huxstep MR , (1981) has shown RO to be effective in reducing traced concentration of these contaminants. The improvements in design and materials of the membranes have made the water treatment process economically competitive and highly reliable [100,101].

Electrodialysis: In electrodialysis, the membranes allow the ions to pass but not the water. The driving force is an electric current which carries the ions through the membranes. The removal of fluoride in the RO process has been reported to vary from 45% to 90% as the pH of the water is raised from 5.5 to 7. The membranes are very sensitive to pH and temperature. The units are also subject to chemical attacks, plugging, fouling by particulate matter, and concentrated and a large quantity of wastes.

The waste volumes are even larger than the ion exchange process. Sometimes, the pretreatment requirements are extensive. Electrodialysis is highly energy intensive and expensive [100].

Limitation [55,102]

1. The membranes are sensitive to temperature, pH and arises maintenance problem because of plugging, fouling by particulate matter, concentrated with a large quantity of wastes.
2. Both processes are expensive and very complicated.
3. Removes all the ions present in water including some essential minerals for proper growth and hence remineralization is required after treatment.
4. High energy consumption and large amount of water gets wasted as brine.

DONNAN DIALYSIS (DD)

DD is also a separation process based on membrane filtration that utilizes counter diffusion of two or more ions through an ion-exchange membrane to achieve a separation. Donnan who described the equilibrium that resulted when a semi-permeable membrane separated two solutions of electrolytes, NaA on one side and KA on the other. DD is highly efficient in treating fluoride contaminated water and used for the treatment of low-concentration waters [103]. A hybrid process that combines the adsorption on conventional solid adsorbents such as aluminum and zirconium oxides with a specific DD procedure is applied to treat groundwater with an excessive fluoride concentration of 4 mg/l resulting from phosphate mining in Morocco [104].

Limitations

1. Expensive
2. Reduced efficiency in saline waters.

NANOFILTRATION

Nanofiltration is a process which takes in the upper end of RO, and the lower end of ultrafiltration. Permeability of Nanofiltration membranes is higher than those of RO. Nanofiltration membranes have a high retention of charged particles. It requires less pressure and capital than RO and it is widely applicable especially for drinking and waste water treatment and is used in defluoridation studies [105-108].

Table 2: Researches conducted on activated alumina from 1934 to 2013

S. No.	Authors	Study done on activated alumina
1.	Boruff (1934), Fink and Lindsay (1936), Swope and Hess (1937) and Savinelli and Black (1958)	Fluoride removal capacity of activated alumina increases directly with fluoride capacity [47, 59-61]
2.	Bishop and Sansoucy (1978)	Fluoride removal is using fluidized activated alumina as an adsorbent [62]
3.	Hao *et al.* (1986)	Suggested adsorption characteristics of fluoride onto hydrous alumina [63]
4.	Schoeman and MacLeod (1987)	Suggested that the rate of fluoride adsorption of small particles is more than large sized particles with favorable pH of 5-6 [64]
5.	Farrah *et al.* (1987)	Investigated the interaction of F$^-$ ion with naturally occurring amorphous aluminum hydroxide, gibbsite and aluminum oxide over a pH ranging from 3 to 8 and fluoride concentration from 1.9 to 19 mg/l; and most of AlF dissolved at 6pH. Max fluoride uptake occurred with pH of 5.5-6.5. The gibbsite removal capacity was less compared to others [65]
6.	Karthikeyan *et al.* (1997)	Studied activated alumina by Calibration method. The min contact time for defluoridation was 30 minutes where fluoride adsorption capacity was 3 mg/g and 20 mg/g in alkaline and acidic water, respectively, and at neutral condition fluoride capacity was 5.6 mg/g [66]
7.	Bahena *et al.* (2002)	Studied fluoride adsorption on to apha-Al_2O_3 and its effect on zeta potential at alumina aqueous. Maximum fluoride removal was achieved between pH of 5-6 at 25°C [67]
8.	Ku and Chiou (2002)	Studied about maximum fluoride at a pH of 5-7 which was 16.3 mg/g using activated alumina as an adsorbent. It was observed that adsorption of fluoride retarded in acidic solutions because of electrostatic repulsion between them. If the equilibrium solution was greater than pH 7, fluoride adsorption is reduced by alumina [68]
9.	Ghorai and Pant (2004)	Investigated the removal of fluoride using activated alumina (GradeOA-25) in batch and continuous operation and observed the adsorption capacity to be as 1450 mg/kg at pH 7. There was marginal decrease in uptake capacity after each regeneration cycle [69]
10.	Pietrelli (2005)	Suggested fluoride adsorption of MGA. It was observed the best removal was observed at pH 5-6 hence it is favorable to promote stable fluoro-alumina complex. The fluoride adsorption onto MGA decreased drastically at higher pH [70]
11.	Shimelis *et al.* (2006)	Studied adsorption capacity of UHA and thermally THA. The removal efficiency of fluoride increases with an adsorbent dosage. The increase in temperature treatment up to 200°C. The high defluoridation technique was achieved using both UHA and THA with a pH of 4.0-9.0 [71]
12.	Rao and Karthikeyan (2008)	Investigated fluoride adsorption capacity of gamma alumina for a fluoride solutions of 4 mg/l. They revealed about 85-95% removal was achieved in first 10 minutes after a sorbent dose of 8 g/l in pH 3-4 and remain fairly constant with pH 7 where adsorbent was used up to 10 cycles with alum [72]
13.	Wang *et al.* (2008)	Suggested fluoride removal potential of nano-scale aluminum oxide hydroxide. The maximum adsorption capacity of 3259 mg/Kg of fluoride occurred at pH of 7 [73]
14.	Tang *et al.* (2009)	Studied adsorption characteristic of fluoride on activated alumina. The fluoride adsorption was carried out a wide range of pH between 5 and 10.5. They observed the fluoride uptake decrease for an increase in pH. A spectation-based model was developed for pH 5-10.5 and wide surface loading range of 1-10 mg-F/g adsorbent [74]
15.	Yama and Yama (2009)	Studied performance of fluoride removal of soil pot made by soil with different amount of $Al_2(SO_4)_3$ and observed decrease in 10 mg/l fluoride concentration into 0.61 mg/l with 2 g/kg soil of $Al_2(SO_4)$ 3 in 3 hrs [75]
16.	George *et al.* (2010)	Investigated an activated alumina defluoridation model stimulator (AAD) predicted the dissolution of aluminum fluoride and aluminum complexes is more favorable to high fluoride concentration with low alumina dosage in pH ranging from 6.5 to 7.5, which leads to increase in residual aluminum in treated water [76]
17.	Sivasankari *et al.* (2010)	Reported that 98% removal of fluoride was achieved by 1.0 g of PAA and 1.4 g GPAA at pH 6 from 100 ml of 10 mg/l fluoride solution but fluoride uptake reduces at pH>7. The adsorption of GPAA was slightly faster than PAA [77]
18.	Lee *et al.* (2010)	Studied meso porous alumina prepared by using aluminum tri-sect-butoxide in the presence of either cetyltrimethylammonium bromide (MA-1) or stearic acid (MA-2) as structure directing agent has enhanced adsorption capacity and faster when compared to commercial activated alumina [78]
19.	Kamble *et al.* (2010)	Studied fluoride adsorption by alkoxide origin alumina. The maximum fluoride adsorption capacity occurred at pH 5 and 7 [79]
20.	Kumar *et al.* (2011)	Studied fluoride adsorption capacity of nano-alumina. The maximum fluoride removal capacity of 14 mg/g at pH of 6.15 at 25°C [80]

(Contd...)

Table 2: (*Continued*)

S. No.	Authors	Study done on activated alumina
21.	Li *et al*. (2011)	Reported highly ordered meso porous alumina (MesoAl-400) and calcium doped alumina (Meso Al-10Ca) for removal of fluoride and arsenic. The highest defluoridation capacity is 300 mg/g and 450 mg/g at pH 6.5 and 298 K for 12 hrs [81]
22.	Biswas *et al*. (2012)	Investigated Hydrous aluminum oxide for fluoride adsorption. The maximum fluoride removal occurred at pH 6.5 [82]
23.	Gong *et al*. (2012)	Studied the adsorption of fluoride onto five unusual types of alumina synthesized at unusual pH and calcination temperatures. Acidic alumina has higher ion-exchange capacity than basic alumina without pH adjustment [83]
24.	Goswami and Purkait (2012)	Investigated acidic alumina has BET surface area of 144.27 m^2/g which removes maximum fluoride at pH 4.4 [84]
25.	Gupta *et al*. (2013)	Studied removal of fluoride with activated alumina in the presence of chloride. They observed the percentage removal of fluoride increases without an increase in dosage of activated alumina [85]

MGA: Metallurgical grade alumina

Table 3: Modified activated alumina

S.No.	Author's	Modified activated alumina
1.	Tripathy *et al*. (2006)	Al+Alum: Fluoride adsorption by alum impregnated activated alumina can remove 92.6% of fluoride at pH 6.5 at the dose of 8 g/l and 3 hrs time from water containing 25 mg/l. At pH above 6.5, the fluoride removal decreases sharply due to strong competition with hydroxide ions on the adsorbent surface [86]
2.	Nawlakhe *et al*. (1975)	Al+Calcium: Two chemicals of alum in the form of aluminum sulfate and potassium aluminum sulfate and lime as calcium oxide was rapidly mixed with fluoride contaminated water to form aluminum hydroxide, after stirring it was allowed to settle down to remove the maximum amount of fluoride [87]
3.	Lunge *et al*. (2012)	Al+Carbon: Alumina supported carbon composite prepared by waste of egg shell for removing fluoride from water. The Langmuir adsorption capacity of composite adsorbent was 37 mg/g at 303 K for a wide range of pH between 3 and 9 [88]
4.	Bansiwal *et al*. (2010)	Al+Carbon: Copper oxide coated alumina was synthesized by saturating alumina with copper sulfate solution followed by a calcination process in the presence of air at 450°C. The adsorption capacity for fluoride as the basis of the Langmuir model was 7.22 mg/g, which was 3 times higher than that of unmodified AA where value obtained was 2.232 mg/g. The significant increase in adsorption capacity was due to the increase in zeta potential. Marginal decreases in sorption capacities was noted at pH above 8 in alkaline conditions [89]
5.	Chubar *et al*. (2005)	Al+Iron: Studied ion exchanger based on double hydrous oxide for removal of fluoride, chloride, bromide, and bromate ions. The Langmuir fluoride adsorption capacity was 90 mg/g at pH of 4 [90]. Biswas *et al.* (2007) studied the adsorption capacity of iron (III) – aluminum (III) mixed oxides made by co-precipitated aluminum and iron hydroxides from a chloride mixture and the optimum pH for fluoride removal was 4-10 and equilibrium time required was 1.5 hr [91]. Dang *et al.* (2011) studied fluoride adsorption from water by Fe2(SO4)3 granular activated alumina (1-2 mm) [92]
6.	Maliyekkal *et al*. (2008)	Al+Magnesia: Magnesia amended activated alumina (MAAA) by calcination of magnesium hydroxide impregnated alumina at 450°C. More than 95% of fluoride removal (10 mg/l). An optimum fluoride removal was detected at pH range of 5.0-7.5s achieved within 3 hr of time at neutral pH [93]
7.	Teng *et al*. (2009)	Al+Maganese oxide: Redox process was used to coat hydrous manganese oxide from the surface of activated alumina. The max removal of fluoride was found at pH between 4 and 6. Adsorption of fluoride took place mainly by the anion exchange between hydroxyl ion and fluoride at acidic pH range. HMOCA was used for carrying out a study to reduce 5 mg/l initial concentration to less than 1.0 mg/l at flow rate of 2.39 m^3/m^2 hr [94]
8.	Tripathy and Raichur (2008)	Al+Manganese dioxide: The fluoride removal ability of manganese dioxide coated with activated alumina is up to 0.2 mg/l at pH 7 in 3 h with 8 g/l adsorbent dose when 10 mg/l of fluoride was present in 50 ml of water [95]
9.	Puri and Balani (2000)	Al+Lanthanum: The adsorption capacity for alumina impregnated with Lanthanum hydroxide was found as 0.340-0.365 mm/g, whereas for the original alumina it was 0.170-0.190 mm/g. The adsorbent was effective when the fluoride concentration decreases from 7 mm to 0.003 mm in the pH range of 5.7-8.0 where the Langmuir adsorption capacity was 0.82 mm F-/g of the adsorbent [96]. Shi *et al.* (2013) reported about lanthanum oxide impregnated granular activated alumina for fluoride removal. Five cycles of lanthanum impregnation on AA were carried out followed by the calcination at 573 K which was responsible for increasing the La content up to 19.1% and achieved the maximum fluoride adsorption at 16.9 mg/g [97]
10.	Karthikeyan *et al*. (2009)	Al+Polymer: The study was done on polyaniline/alumina (PANi-AlO) and polypyrrole/alumina (PPy-AlO) for the removal of fluoride. The maximum amount of adsorption was 6.6 mg/g and 8 mg/g for PANi-AlO and PPyAlO, respectively. The fluoride removal mechanism was based on formation of aluminium-fluoro complexes on the alumina surface and doping/dopant exchange of fluoride ions in the polymer. The pH study was under taken over the range of 3-9. In acidic pH, adsorption was high due to positive charged polymer/alumina composite to attract fluoride ions electrostatically whereas in the alkaline range, the hydroxide ion could compete effectively with fluoride ions leading to a lower defluoridation [98]

CLAY

Based on testing of capacity of clay to remove fluoride from water, different studies had different conclusion, Bulusu *et al.* reported that clay was not worth for defluoridation [109] and Zevenbergen *et al.* reported Andio soil appears to be efficient and economical method for defluoridation of water [110]. Both clay powder and fired clay can take up fluoride in sorption process. Due to its high density when compared with bone charcoal it will settle and drain off all the supernatant water. In a study by Padmasiri the domestic clay filter packed using clay chips resembles to filters used in Sri Lanka [111]. Bardsen and Bjorvatn reported the sorption isotherm of clay calcined at 600°C. It was found that sorption continuous to take place until 10 days but the capacity was low as 0.07 mg/g at 1 mg/g level [112]. Padmasiri showed the operational capacity of 0.08 mg/g for clay chips used in Sri Lanka [111]. Jinadasa *et al.* concluded the capacity is known to be optimum when the pH is 5.6 [113]. It was stated by Padmasiri that the clay process of defluoridation is the most cost-effective only if the burnt broken bricks are of good quality and a available at the same site. Nearly 80% of 600 clay column defluoridators used in the household of Sri Lanka were found in operating condition after being monitored from 2 years.

CONCLUSION

All the methods discussed above have proved to be efficient and have shown to have capacity to remove excess fluoride. But still, selection of appropriate technique, infrastructure and research experience in this field is essential to enhance safe defluoridation of drinking water.

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