Fluoride Removal from Water by various techniques: Review

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Abstract

This paper depicts the fluoride removal from drinking water can be accomplished by different methods, for example, coagulation-precipitation, membrane separation process, ion exchange, adsorption techniques and so on. Among these procedures, membrane and ion exchange processes are not extremely regular because of its high establishment and support price. Other two tecniques are extremely regular in India . Nalgonda procedure is one of the well known strategies generally utilized for defluoridation of water as a part of developing nations, for example, India, Kenya, Senegal and Tanzania. Among different tecniques utilized for defluoridation of water, the adsorption procedure is broadly utilized and offers acceptable results and is by all accounts more appealing technique for the removal of fluoride regarding expense, straightforwardness of outline and operation. It is apparent from the literature study that different methods have indicated novel potential for the removal of fluoride. Be that as it may, in any case there is a need to figure out the reasonable utility of such developed procedures on a business scale, prompting the change of contamination control.

Keywords: water treatment, fluoride removal, adsorption, ion exchange, review

1. Introduction

Nature of drinking water is a major task in advanced days because of expansion in pollution of water bodies [1]. Fluoride is one such pollutant that undermines living life forms, specifically people [2]. Fluoride is an vital in little amount for mineralization of bone and assurance against dental caries, higher intake reasons decay of teeth enmel called fluorosis. Fluoride enters aqueous environment by weathering of fluoride rich minerals and as through anthropogenic actions, for example, industrial drains [3]. The issue of fluoride in water bodies is serious for tropical nations, for example, such as India, Kenya, Senegal and Tanzania. The best way to pypass this issue is defluoridation. Various methods are accessible for the removal of fluoride from water, for example, precipitation-coagulation, membrane-based processes, ion –exchange and adsorption process. The precipitation-coagulation method makes vast amount of sludge and may include leaching of undesirable components; membrane procedure are lavish and fouling is an inescapable issue. Adsorption procedure have their own particular poimts of interest, for example, ease and minimized water disposal. In this review, a widespread list of procedures literature has been assembled. It is apparent from a literature study

of around 200 latest papers that minimal effort methods have exhibited extraordinary removal capacities for fluoride.

2. METHODS FOR DEFLUORODATION OF WATER

To conquer the hazardous wellbeing impact of fluorosis, different approaches for defluoridations are exists like coagulation – precipitation, membrane separation processes, ion exchange, adsorption techniques and others (electro-dialysis and electrochemical). Each approaches have their advantages and limitations and worked productively under ideal condition to remove fluoride to more noteworthy range. All the above approaches are examined briefly with their advantages and limitations.

2.1. COAGULATION AND PRECIPITATION METHOD

Lime and alum are the most usually utilized coagulants for Nalgonda technique for defluoridation of water. Expansion of lime prompts precipitation of fluoride as insoluble calcium fluoride and raises the pH value upto 11 – 12. As the lime leaves a leftover of 8.0 mg F^-/l, it is constantly connected with alum treatment to guarantee the best possible fluoride removal. As a first step, precipitation happens by lime dosing which is trailed by a second step in which alum is added to bring about coagulation. At the point when alum is added to water, basically two reactions happen. In the first reaction, alum reacts with an alkalinity's portion to deliver insoluble aluminium hydroxide $[Al(OH)_3]$. In the second reaction, alum reacts with fluoride ions in the water. Best fluoride removal is proficient at pH range of 5.5 – 7.5 [4]. Nalgonda technique created by NEERI is coagulation – precipitation method includes an expansion of aluminium salt, lime and bleaching powder took after by quick mixing, flocculation, sedimentation and filtration. Aluminum salt is utilized to remove fluoride from water. The dosage of fluoride relies on upon the concentration of fluoride proportionately. The dosage of lime is by and large 1/20th of the dose of alum. Lime serves to shape bigger and denser flocs for fast settling. Bleaching powder is included for cleansing at the rate of 3 mg/l [5]. It is the most generally utilized defluoridation method especially at community level [6-8]. The bucket defluoridation system based Naldonda technique has also been developed for

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household utilize [9]. The process is suitable for 20 litres of water for one day utilization. The process produces water with leftover fluoride somewhere around 1 and 1.5 mg/l [10]. Fill and draw type defluoridation system based on Nalgonda technique has also been account for [9]. Nevertheless, co-precipitation methods in view of aluminium salts have a few point of interest and restriction:

Interest

1. Generally utilized technique.
2. Technique is more practical when contrasted with other defluoridation technique.
3. Technique is easy to understant.

Restriction

1. Required chemical dosages are high ($Al(OH)_3$ upto 700 – 1200 mg/l).
2. Sludge transfer issue.
3. Cannot accomplish the passable furthest reaches of fluoride.
4. Prerequisite of talented labor
5. Release of aluminium in treated water which may bring about Alzheimer's syndrome.
6. Final concentration of fluoride in the treated water significantly relies on upon dissolvability of precipitated fluoride and calcium and aluminium salt.
7. The utilization of aluminium sulfate as coagulant expands the sulfate ion concentration greatly which prompting catharatic impacts in human

Nalgonda technique, disregarding introductory achievement, did not take off in light of some intrinsic issues as depicted previously. A contact precipitation defluoridation technique utilizing bone char combined with sodium dihydrogen phosphate and calcium chloride has been account for [10]. Narthasarthy et al. contemplated the joined utilization of calcium salts and polymeric aluminium hydroxide for the defluoridation work. Chang and Liu examined coagulation –flocculation of calcium fluoride precipitates in combination with polyaluminium chloride and polyacrylic acid at lower dosage. To beat the restriction of chemical precipitation, granular calcite is utilized to catch fluoride present in water as calcium fluoride in a fluidized bed reactor by Aldaco et al. [31].

2.2. MEMBRANE PROCESS

The membrane separation process is more well known from industrial viewpoints for defluoridation of groundwater, wastewater treatment and sea water desalination [12]. In a membrane separation process, particles are isolated on the premise of their molecular size and shape with the utilization of extraordinarily composed semi-permeable membrane. The semi-permeable membrane is frequently a thin, nonporous or porous polymeric film, ceramic, or metal material or even a liquid

or gas. The membrane must not dissolve, disintegrate or break [13]. The most normally utilized membrane separation processes for removal of fluoride are reverse osmosis, nano-filtration, Donnan-dialysis and electrodialysis.

2.2.1 REVERSE OSMOSIS (RO)

RO is a physical process in which the anions are removed by applying pressure on the feed water to direct it through the semi permeable membrane. RO works at higher pressure with more prominent rejection of dissolved solids. The membrane rejects the ions taking into account the size and electrical charge. RO membrane process is the reverse of natural osmosis as a consequence of applied hydraulic pressure to the high concentration side of the solution, it forces solvent filter through the membrane, against a pressure gradient into the lower-concentration solution. In RO, utilizing a mechanical pump, pressure is applied to a solution via one side of the semi-permeable membrane to overcome inalienable osmotic pressure. The process likewise removes soluble and particulate matter, incorporating salt from seawater in desalination [14]. In the 80's, RO membrane separation technique was effectively connected for the treatment of industrial wastewater particularly for the removal and recovery of fluoride from its effluents. More than 90% of fluoride can be removed regardless of initial fluoride concentration using RO membrane separation process [15].

Ndiaye et al. was utilized RO separation process for defluoridation of industrial wastewater observed that the rejection of fluoride ion was regularly higher than 98%, considering that the RO membrane was completely recovered after every arrangement of analyses [15]. Berhanu Assefa et al. concentrated the fluoride retention of RO membranes of Ethiopian Rift Region were in the range of 94 to 99 % [16]. Diawara et al. utilized low pressure reverse osmosis for removing fluoride and salinity of brackish ground water of Senegal village where 97 to 98.9% of fluoride rejection happened [17]. Gedam et al. study uncovered that 95 to 98 % of fluoride was removed from ground water of Moradgaon village of Chandrapur district by using Polyamide RO membrane [18]. Schoeman had used RO for defluoridation of some provincial territories of South Africa expressed that fluoride can be removed with low pressure RO in the feed water concentration range from 10 to 17 mg/l to more or less 0.2 mg/l in the RO permeate and with high pressure RO from roughly 17 mg/l in the feed water to approximately 0.2 mg/l in the RO permeate [19]. Briao et al. used reverse osmosis for desalination of water from the Guarani Aquifer System for drinking purpose in southern Brazil. The rejection of 100% of fluoride, 97% of total dissolved solids (TDS) and 94% of sulphate ions was achieved by RO at 2MPa pressure and 1.61 m/s of cross-

section flow velocity. The recovery rate of 93% of drinking water was obtained by blended water created by mixing groundwater with permeate [20].

The point of interest and restriction of the RO membrane separation are given below:

Interest
1. Technique is profoundly compelling for fluoride removal.
2. RO membrane was completely recovered after every arrangement of examination.
3. This strategy can remove fluoride more than 90% regardless of initial concentration.
4. This strategy gives the synchronous removal of other dissolved solids.
5. It efforts under wide pH range.
6. No obstruction by different ions
7. No chemical oblige, least labor prerequisite and least operational expense.
8. The process allows the treatment and purification of water in one stage.
9. It guarantee steady water quality

Restriction
1. Non-attainable for rural regions.
2. Expensive technique.
3. Remove valuable minerals which are basically require for fitting development, remineralization is require after treatment.
4. Lot of water get squandered as saline solution and expendable of salt water is an issue
5. The water gets to be acidic and need pH improvement.

2.2.2 NANO FILTRATION MEMBRANE PROCESS

Nanofiltration (NF) is the later innovation among all the membrane processes utilized for defluoridation of water. The essential contrast in the middle of NF and RO membrane separation is that NF has somewhat bigger pores than those utilized for reverse osmosis and offer less resistance to entry of both solvent and of solute. As a outcome, pressures needed are much lower, energy prerequisites are less, removal of solute is substantially less thorough, and flow are faster [21]. Nanofiltration membrane removes essentially the larger dissolved solids when contrasted the RO making the process more prudent. Notwithstanding over, the permeability of nanofiltration membrane is higher than RO membrane, making the performance of NF in desalination more best for some brackish water [22]. In RO membrane separation 99% of salt present in water was rejected prompting the disposal of all the fluoride ion while NF membrane separation process give incomplete defluoridation of water and optimal fluoride concentration in water can be accomplished by changing the operation conditions.

Diawara et al. observed that NF membrane has given a fluorine retention rate differing somewhere around 63.3% and 71% in certain regions in Senegal [16]. Tahaikt et al. observed the attractive execution of two modules (NF90 and NF400) for fluoride concentration under 6 mg/l in single pass while double pass was important for higher fluoride concentration to convey down as far as possible [23]. Pontie et al. used NF90 for acceptable removing fluoride from brackish ground water of South of Morocco (Tan Tan City) [24]. Bejaoui et al. used nanofiltration (NF-90) and reverse osmosis (RO-SG) to lessened fluoride ions and total salinity of a metal packaging industrial effluent. The retention of fluoride was more than 90% by both the membranes. The Spiegler-Kedem model was to determine reflection coefficient of membrane (r) and solute permeability coefficient of ions (Ps) [25].

The point of interest and restriction of the NF membrane process are given below:

Interest
1. High productivity.
2. No chemicals are needed.
3. It lives up to expectations under wide pH range.
4. Interference because of the presence of other ion is not observed.
5. This process gives an effective barrier to suspended solids, all inorganic toxin, organic micro pollutants, pesticides and microorganism.

Restriction
1. Highly expensive technique when contrasted with other defluoridation techniques.
2. Prone to fouling, scaling or membrane degradation.
3. It removes all the ions present in water some of which are key for the ordinary development and henceforth remineralization of treated water is needed.

2.2.3 DIALYSIS

Dialysis separates solutes by transport of the solutes through a membrane instead of utilizing a membrane to hold the solutes while water goes through it as in reverse osmosis and nanofiltration. The membrane pores are a great deal less prohibitive than those for nanofiltration, and the solute can be driven through by either the Donnan effect or a connected electric field [26]. Donnan dialysis is otherwise called diffusion dialysis, is similar to ion exchange membrane however unique in relation to electro-membrane process in which the driving force is not an electric current, but rather basically a distinction in chemical potential. Concentration difference is the most obvious driving force for ion transport in Donnan dialysis. A negative ion can be driven out of a feed solution through Donnan dialysis is equipped with anion exchange membrane by utilizing a second alkaline stream. The

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concentration difference of hydroxide ion between the two solutions compels the hydroxide ion to diffuse into feed solution. This makes an oppositely directed electrical field driving an extraction of negative ions from the feed solution [27]. Hichour et al. examined the Donnan dialysis process in a counter current system in which the anion-exchange membrane was loaded with sodium chloride and the feed was 0.001M NaF together with other sodium salts. Fluoride migrated into the receiver as other ions migrated into the feed. This technique was later used to defluoridate solutions made to rectreate high fluoride Africa groundwater (>30mg/l fluoride) and whatever other ions were present the fluoride in the feed could be brought below 1.5 mg/l [27]. Grames et al. used hybrid method of adsorption on aluminium oxide and zirconium oxide and Donnan dialysis with Neosepta-ACS anion exchanger to treat fluoride water of 4mg/l from phosphate mining in Morocco and bring it to desirable level (i.e. 1.5mg/l) for drinking purpose [28]. Durmaz et al. studied the removal of fluoride from diluted solution with Neosepta AHA anion exchanger by Donnan dialysis. Neosepta AHA anion exchanger membrane was used for the removal of fluoride and the flux of fluoride with respect to the concentration, pH, and accompanying anions were obtained between 34 – 136.10-7 (m.mol.cm^{-2}.s^{-1}). The transport efficiencies of the membrane were found to be Neosepta AFN>Neosepta AHA>Polysulfone SB-6407. Alkan et al. used plasma modified and Prinstine and AFX anion exchange membrane by Donnan dialysis for removal of fluoride. The flux values and recovery factors for plasma modified AFX membrane were higher than that of Prinstine membrane which was clarified on the premise of progress of wettability and morphology in the plasma modified membrane [29]. Boubakri et al. studied the fluoride removal from dilute solutions by Donnan dialysis using full factorial design [30].

2.2.4 ELECTRO DIALYSIS

Electro-dialysis is the removal of ionic components from aqueous solutions through ion exchange membranes under the driving force of an electric. Electro-dialysis is like reverse osmosis, except current, rather than pressure, to separate ionic contaminants from water. In any case, electro-dialysis is not suitable for rural because of use of electricity. Adhikary et al. have treated defluoridation of brackish water having fluoride upto 10 ppm with TDS upto 5000 ppm with an energy necessity of < 1 KWh/Kg of salt removed and brought it to reasonable firthest reaches of 600 ppm TDS and 1.5 ppm fluoride [31]. Amor et al. used electrodialysis process to produce drinkable water from defluoridates brackish water containing 3000 ppm of total dissolved solids (TDS) and 3 ppm of fluoride [32]. Annouar et al. studied the fluoride removal of synthetic water and groundwater of the city of Youssoufla

on chitosan took after by electrodialysis with the help of the CMX-ACS membranes and compares them to brought water inside WHO permissible limit [33]. Sahli et al. also used chitosan and electrodialysis for defluoridation of brackish underground water of Morocco city and by coupling these two procedures; they were treated defluoridated water with 3000 mg/l of total dissolved solids and 3 mg/l fluoride [34]. Kabay et al. studied the removal of fluoride from aqueous solution by electrodialysis under different operating parameters including applied voltage, feed flow rate, fluoride concentration and effects of sulphate and chloride ions. It was observed that the separation performance increases with initial fluoride concentration in feed solution increased and fluoride removal increased as applied potential increased. On the other hand, the performance was not affected by change in feed flow rate. A chloride ion impacts the performance of separation of fluoride while unaffected by sulphate ions [35]. Ergun et al. used electrodialysis with SB-6407 anion exchanger membrane for removal of fluoride from water having 20.6 mg/l of fluoride and decrease it to 0.8 mg/l suitable for drinking purpose in spite of the presence of chloride and sulphate ions [36]. Lahnid et al. have made economical assessment of fluoride removal by electrodialysis. The capital coast of €833,207 with operating cost of €0.154/m^3 was evaluated for an industrial plant with a capacity of 2200 m^3/d water consumption for 50,000 per capita according to Moroccan standards for rural areas [37].

The point of interest and restriction of the electro-dialysis are given below:

Interest
1. Inexpensive pre and post treatment
2. Flexible (seasonal operation)
3. Low chemical request
4. High water recovery

Restriction
1. Only separation of Ionic components
2. Potential formation of H_2 in the electrode rinse
3. Specific power consumption for Pumping
4. Necessity of concentrate treatment

2.3 ION-EXCHANGE PROCESS

Fluoride can be removed from water supplies with a strongly fundamental anion-exchange resin containing quaternary ammonium functional groups. The removal takes place according to the following reaction:

$$Matrix\text{-}NR_3 + Cl^- + F^- \longrightarrow Matrix\text{-}NR_3 + F^- + Cl^-$$

The fluoride ions substitute the chloride ions of the resin. This process proceeds until every one of the sites on the resin are possessed. The resin is then backwashed with water that is supersaturated with dissolved sodium chloride salt. New chloride ions then substitute the

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fluoride ions promping recharge of the resin and beginning the process once more. The driving force for the substitution of chloride ions from the resin is the stronger electronegativity of the fluoride ions. Chikuma et al. studied the removal of fluoride by utilizing anion exchange resin and modified anion exchange resin with lanthanum complex of Alizarian Fluorine Blue in batch and column mode [38]. Chikuma and Nishimura studied the fluoride removal by a chloride loaded anion exchanger, Amberlite IRA-400. The chloride ions held on the surface of this resin were exchanged for fluoride ions in aqueous solution [39]. Castel et al. have likewise studied the removal of fluoride by a two way ion exchange cyclic process [40]. Ho et al. had enhanced the ion exchange capacity by titanium oxohydroxide arranged by using dodecylamine as format. Zirconia and silica have been presented in the mesoporous titanium oxohydroxide to improve the ion exchange capacity because of its smallest particle size with high uniformity among the mesoporous materials prepared. Process is costly and membrane fouling problems was happened [41]. Chubar et al. have studied the removal of different anions like fluoride, chloride, bromide and bromate from instantaneous solutions on a novel ion exchanger taking into account twofold hydrous oxide ($Fe_2O_3.Al_2O_3.x$ H_2O) [42]. Meenakshi et al. have studied the defluoridation capacity of a chelating resin, to be specific Indion FR 10 (IND) and Ceralite IRA 400 (CER). An anion exchange resin was looked at under various equilibrating conditions for the distinguishing proof of selective sorbent. The outcomes demonstrated that chelating resin is more selective than an anion exchange resin for fluoride removal [43]. Sundaram et al. used organic – inorganic type of ion exchangers for fluoride removal. The ion exchanger polyacrylamide was modified with $Ce(SO_4)2•4H_2O$, $Al(NO_3)_3•9H_2O$ and $ZrOCl_2•8H_2O$. Ce-Ex has somewhat higher defluoridation capacity than others (i.e. 2290 mg F-/kg). Likewise it was used for field trials to reduce level of fluoride concentration from 1.96 mg/l to the attractive level using 0.25 g of exchangers for 50ml of samples for 30 min of time of contact [44]. Ku et al. have studied the fluoride removal of water using aluminium loaded Duolite C-467 resin. The removal of fluoride from aqueous solution was found to be relatively steady over entire solution pH [45]. The point of interest and restriction of ion-exchange technique are given below:

Interest
1. High productivity (90-95 % fluoride removal).
2. Retains the superiority of water.

Restriction
1. Technique is exceptionally costly.
2. pH of treated water is low and contains high concentration of chloride.

3. Interference because of the presence of other anions like sulphate, carbonate, phosphate and alkalinity.
4. Regeneration of resin is a an issue on the grounds that it prompts fluoride rich waste, which must dealt with before last disposal.
5. It requires longer reaction period.

2.4 ELECTRO-COAGULATION (EC) PROCESS

Electrocoagulation is a technique for applying direct current to sacrificial electrodes that are submerged in an aqueous solution [46]. Electro-coagulation is a straightforward and efficient technique to remove the flocculating agent produced by electro-oxidation of a sacrificial anode and generally made of iron or aluminum. In this process, the treatment is performed without including any chemical coagulant or flocculants. In this way, diminishing the amount of sludge which must be disposed. Then again, electrocoagulation is in view of the in situ development of the coagulant as the sacrificial anode corrodes because of an applied current, while the concurrent advancement of hydrogen at the cathode takes into consideration contamination removal by flotation. This technique consolidates three fundamental associated processes, operating synergistically to remove pollutants: electrochemistry, coagulation and hydrodynamics. An examination of the chemical reactions happening in the electrocoagulation process demonstrates that the main reactions occurring at the electrodes (aluminum and iron electrodes) are:

$$Al \longleftrightarrow Al^{3+} + 3e^-$$

$$3H_2O + 3e^- \longleftrightarrow 3/2H_2 + 3OH^-$$
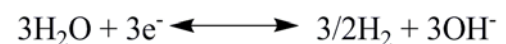
Moreover, Al^{3+} and OH^- ions produced at electrode surfaces react in the bulk wastewater to form aluminum hydroxide:

$$Al^{3+} + 3OH^- \longleftrightarrow Al(OH)_3$$

Also the same chemical reactions occurring in the electrocoagulation process using iron electrodes:

$$Fe(s) \longleftrightarrow Fe^{3+}(aq) + 3e^-$$

$$3H_2O + 3e^- \longleftrightarrow 3/2H_2(g) + 3OH^-(aq)$$

$$Fe^{3+}(aq) + 3OH^- \longleftrightarrow Fe(OH)_3$$

The aluminum and iron hydroxide flocs for the most part go about as adsorbents and/or traps for metal ions. Thusly, they would remove with them from the solution. The core purpose of this investigation was to research of the electrocoagulation process productivity for fluoride removal from aqueous environments with iron and aluminum electrodes and determination of the impacts of

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voltage, pH, initial concentration of fluoride and reaction time on the removal efficiency [47].

Electrocoagulation includes electrolytic oxidation of a proper anode material. Electro-coagulation reactor is comprised of an electrolytic cell with one anode and one cathode [48]. Yang et al. studied the electrochemical removal of fluoride by delivering aluminium sorbent in a parallel-plate electrochemical reactor. Defluoridation was done by anodic dissolution of aluminium electrodes in a dilute sodium chloride (NaCl) aqueous solution. The NaCl in the solution adequately diminished the power utilization and advanced the sorbent generation by depasivating the aluminum–water electrochemical system. The freshly created Al-sorbent had the capacity diminish fluoride concentration from 16 to 2 mg/l in 2 min. The final concentration was further lessened to 0.1 mg/l by partial neutralization of the mixture to pH 6.3. The sorbent generation and fluoride adsorption was incorporated into a solitary electrochemical reactor. The system had the capacity decrease the fluoride concentration from 16 to 6 mg/l in 2 min of treatment and to around 2 mg/l in 4 min. The effluent from the electrochemical system needs pH alteration to bring the fluoride concentration down to less than 1 mg/l. Feng Shen et al. studied consolidated procedure of electrocoagulation and electro-flotation for defluoridation of industrial water. By this process, the influent fluoride concentration of 15 mg/l, a value after lime precipitation, reduces to 2 mg/l in an effluent when the pH in the coagulation cell is around 6, charge loading is at 4.97 F/m^3 water, and the residence time is 20 min. Indeed, even lower effluent concentration can be accomplished if 50 mg/l of Fe^{3+} or Mg^{2+} are included into the coagulation unit. The anions for the most part decrease the fluoride removal efficiency aside from Cl$^-$ whose corrosion pitting of the electrode can bring about 130% present efficiency. The weakly acidic condition is satisfactory in the treatment, while too high or too low pH can influence the development of the Al(OH)$_3$ flocs [49]. Hu et al. studied a continuous bipolar electrocoagulation – flotation (ECF) system for the treatment of the high fluoride content wastewater following calcium precipitation with addition of addition of an anodic surfactant, sodium dodecyl sulfate (SDS). The dose of SDS in a continuous ECF system was higher than that in the batch system demonstrating the SDS acted not only as frother, but also as collector in the consistent system. The removal of suspended solids (SS) in the consistent system was not as much as that in the batch system in light of the fact that the scum was distributed by the flow of wastewater in the flotation tank [50]. Ghosh et al. examined the treatment of fluoride containing drinking water with concentration 2 to 10 mg/l by electrocoagulation using mono-polar and bipolar electrode connections. It was observed that the removal of fluoride

was preferred for bipolar connection than for mono-polar connection. The final recommendable breaking point of fluoride (1 mg/l) was obtained in 30 min at 625 A m^{-2} using bipolar connection. The operational costs of mono-polar and bipolar connections were 0.38 and 0.62 US $ m^{-3} respectively [51]. Drouiche et al. was completed the treatment of synthetic fluoride containing solutions by electrocoagulation method using aluminium electrodes for the treatment of wastewater from photovoltaic wafer production industry. EC was investigated for applied potential (10-30V), electrolysis time and supporting electrolyte (NaCl) concentration (0–100 mg/l). Our results showed that with increasing applied potential and electrolysis time, the Al^{+3} dosage increases too, and thereby favouring the fluoride ions removal. It was also observed that defluoridation is dependent on the concentration of supporting electrolyte. Un et al. studied the removal of fluoride from synthetic water made by NaF using iron cylindrical reactor as anode and a mechanical stirrer with two blades as cathode. Sodium sulphate (Na$_2$SO$_4$) was added to the aqueous solution to increase conductivity. It was observed that the most effective fluoride removal was occurred at pH is 6.0. The most noteworthy treatment efficiency was acquired for the largest current density and higher concentration of sodium sulphate. The initial fluoride concentration of 5 mg/l was reduced to the 0.70 mg/l with the removal efficiency of 85.9% at 3 mA/cm^2 [52]. Bennajah et al. studied the fluoride removal of synthetic solution using electrocoagulation – electro flotation method in two electrocoagulation cells (i.e. Stirred tank reactor and airlift reactor) of 20 litre capacity with aluminium electrodes. The air lift reactor is advantageous for carrying out the defluoridation removal process based on energy consumption. A variable order kinetic (VOK) derived from the Langmuir-Freundlich equation was created to simulate the kinetics of the defluoridation with EC using bipolar aluminium electrodes in the airlift reactor. The outcomes indicated good agreement between the predictive equation and the experimental data. The external-loop reactor is affirmed as a productive tool to accomplish complete flotation using only electrochemically-generated bubbles without the need for surfactants or compressed air to induce overall liquid circulation. Another favorable position for the external-loop reactor is the instantaneous recovery of the floc, contrasted with the instance of the stirred reactor where the recovery of the floc obtained by the EC needs quite a while or an additional secondary treatment (like filtration or sedimentation) [53]. Khatibikamal et al. studied the fluoride removal from industrial wastewater started from steel industry by electrocoagulation using aluminium electrode. The impact of different operating conditions such as temperature, pH, voltage, hydraulic retention time

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(HRT), and number of electrodes between anode and cathode plates on removal of fluoride were studied. The outcome demonstrated that the fluoride concentration can be lessened from 4 to 6 mg/l to lower than 0.5 mg/l with the HRT of 5 minutes. They likewise examined the kinetics of fluoride removal, which complies the second-order kinetic model [54]. Vasudevan et al. studied the impact of alternating and direct current in electrocoagulation process on the removal of fluoride from water using an aluminium alloy as anode and cathode. It was observed that the utilization of direct current makes issue of formation of impermeable oxide layer on the cathode and corrosion of anode happened because of oxidation. Which lessened the efficiency of the elctrocoagulation process as the current transfer between the anode and cathode was not productive. This disadvantage was overcome by utilizing alternating current. The removal efficiencies of 93 and 91.5% with energy consumption of 1.883 and 2.541 kWh kL^{-1} was accomplished at a current density of 1.0 A dm^{-2} and pH 7.0 using an aluminum alloy as electrodes using AC and DC, respectively. Langmuir adsorption isother was fitted and second order kinetics was taken after for both alternating and direct current. Temperature studies demonstrated that adsorption was exothermic and spontaneous in nature [55]. Emamjomeh et al. utilized a monopolar batch ECF reactor with aluminium as sacrificial electrode that was submerged in aqueous solution for removal of fluoride. Dissolving aluminum (Al^{3+}) is overwhelming in the acidic condition and aluminium hydroxide has propensity soluble. The defluoridation process was observed to be efficient for a pH ranging from 6 to 8. The residual fluoride may happen in different dissolved forms (F^-, AlF^{2+}, AlF^{4-}) or finely formed to solid (cryolite, $Al(OH)_{3-x}F_x$). The mechanism of the fluoride removal was affirmed to be not just the aggressive adsorption between OH^- and F^- additionally the development of solid cryolite in the last pH range of 5–8 [46]. Bazrafshan et al. utilized aluminium and iron electrodes for removal of fluoride by elecrtrocoagulation method. The outcome uncovered that the most extreme productivity of fluoride removal was acquired in steady electrolysis voltage of 40 V and reaction time of 60 minute [47].Cui et al. studied the executive poly (aniline-co-o-aminophenol) (PAOA) modified carbon felt electrode reactor for fluoride removal in constant mode. This reactor was worked under a more extensive pH range due to coating with a copolymer PAOA ion exchange film. Electrode made up of porous carbon felt gave great electro-conductivity and subsequently acquired upgraded contaminants mass transfer from bulk solution to the electrode surface. Fluoride maintenance expanded as the terminal potential expanded from 0.8 to 1.2 V, and diminished as inlet flow rate and initial fluoride

concentration expanded. Optimal removal was seen at around 1.2 V, and the total amount of fluoride removal reached 10.5 mg/g at pH 7.2 with an initial fluoride concentration of 10 mg/L. The reactor was utilized upto seven cycles with back to back recovery [56]. Mumtaz et al. studied the performance of electrolytic defluoridation of synthetic solution of sodium fluoride by nonstop process with aluminium electrodes using direct current. The optimal defluoridation was happened at conditions of optimal pH of 6.5, optimal current of 3A and flow rate of 550 ml/min for residual fluoride concentration inside of as far as possible [57]. Naim et al. used batch electrocoagulation-floatation procedure for removal of fluoride from analar (AR) and commercial grade sodium fluoride solution using bipolar and monopolar ECF. Complete removal of fluoride was acquired after 5 min for AR grade (5mg/l) and for commercial grade (4.93 mg/l) solution without including any added substances. Monolayer configuration had more powerful removal than the bipolar one [58]. Babu et al. studied the defluoridation of distilled and ground water by using electrocoagulation – floatation method with mild steel electrodes. The maximum fluoride removal efficiency of distilled water was 84.9% and 79.4% in batch and constant mode respectively for the highest applied voltage of 25 V and pH of 6.42 in batch system. Removal efficiency from ground water was 79.6% and 28.7% in batch and consistent mode respectively [59]. Andey et al. studied performance the solar based electrolytic defluoridation plants by using aluminium plate electrodes with direct current. It was observed that electrolytic defluoridation create the treated water with fluoride under 1 mg/l and 90 - 99 % decrease in bacterial load from the raw water with the fluoride in the range of 2 to 5 mg/l and total coliform and fecal coliform counts in the range of 120 to 630 CFU/100 ml and 70 to 100 CFU/100 ml respectively in raw water, Reduction in hardness and nitrate is likewise seen in treated water. The repeating expanse for the treatment worked out for electrolytic defluoridation showing plant is $ 0.4 / m^3 of treated water which is significantly more not exactly the treatment cost by some other defluoridation system accessible [60]. Tastaban et al. studied the fluoride removal of synthetic water through electrocoagulation by utilizing aluminium electrodes at acidic pH range. It was found that fluoride removal was more efficient for the pH range between 3 – 6.5 and fluoride concentration was get decreases from 5 mg/l to lower than 1 mg/l. First order kinetic model was fit for this study. Takdastan et al. studied defluoridation of drinking water by Electrocoagulation (EC) procedure utilizing iron and aluminum electrodes. The fluoride removal of 97.86% was accomplished by aluminum electrodes at optimum condition (pH of 7.5, applied voltage of 20V and contact time of 40 min) and discovered more than defluoridation

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by utilizing iron electrodes. The fluoride removal efficiency was expanded by expansion of voltage, number of electrodes, reaction time as well as decrease of space between electrodes. The energy consumption of aluminum electrodes was not as much as iron electrodes [61]. Sandoval et al., 2014 studied the fluoride removal from synthetic drinking water (10 mg/l F$^-$ in 0.5 g/l Na$_2$SO$_4$ and 1.5 mg/l ClO$^-$ at pH 7.7 and conductivity 410 μScm^{-1}) by electrocoagulation with aluminum as sacrificial anode in a consistent filter press reactor combined with a flocculator and clarifier (Sludge settler). The treated water with fluoride under 1.5 mg/l was observed by electrocoagulation at optimum conditions (current density = 5-7 mA/cm^2 and linear flow velocity = 0.91-1.82 cm/s) gave aluminum doses between 19.28-52.67 mg/l. The fluoride removal from 10 mg/l to 1 mg/l was obtained along with minimum energy consumption of 0.37 kWh/m^3 at current density of 5 mAcm^{-2} and mean linear flow velocity of 1.8 cms^{-1} [62]. Naim et al., 2015 studied the defluoridation of water by electrocoagulation (EC) technique by using bipolar aluminum electrodes. The complete fluoride removal from 6.44 mg/l analar sodium fluoride solution was accomplished inside of 15 min. The optimum minimum weight of NaCl for complete defluoridation from fluoride solution of 10 mg/l was observed to be 0.5g. The optimum pH and speed of agitation was 6 and 300 rpm. The defluoridation of water by EC was more for analar NaF solutions than commercial NaF solutions [63].

The point of interest and restriction of electrocoagulation are depicted as takes after [48]:

Interest

1. EC obliges basic equipment, simple to handle and less support cost.
2. EC treated water is consumable, colourless and odourless.
3. EC produces low sludge that is promptly settable and simple to de-water since it essentially content metallic oxides or hydroxides.
4. EC produces more steady and effectively separated by filtration.

Restriction

1. The 'sacrificial electrodes' are dissolved into wastewater streams as an after effect of oxidation, and should be consistently supplanted.
2. The utilization of electricity may be lavish in numerous spots.
3. An impermeable oxide film may be framed on the cathode prompting loss of productivity of the EC unit.
4. High conductivity of the wastewater suspension is needed.
5. Gelatinous hydroxide may tend to solubilize now and again.

2.5 ADSORPTION

Adsorption is the bond of molecules species from bulk solution for a surface of of a solid by physical or chemical forces. Adsorption procedures include the water's entry through a contact bed where fluoride is removed by ion exchange or surface chemical reaction with the solid bed matrix. As contrast with different procedures of defluoridation, adsorption method is prominent because of its straightforwardness and also accessibility of extensive variety of adsorbents. Adsorption onto solid surface is straightforward, flexible and suitable procedure for treating drinking water systems, particularly for small groups. Adsorption technique is efficient and can remove ions over an extensive variety of pH to a lower leftover concentration than precipitation [64,65]. A few adsorbent materials have been attempted in the past to check their possibilities and techno-economic feasibility as defluoridating specialists. Activated alumina, activated carbon, activated alumina coated silica gel, calcite, activated saw dust, activated coconut shell powder, activated fly ash, groundnut shell, coffee husk, rice husk, magnesia, serpentine, tri-calcium phosphate, bone charcoal, activated soil sorbent, defluoron-1, defluoron-2 and so on various adsorbent materials reported in literature [66-74]. The most regularly utilized adsorbents are activated alumina and activated carbon. The fluoride removing efficiency of activated alumina gets influenced by hardness, pH and surface loading (the ratio of total fluoride concentration to activated alumina dosage). The adsorption procedure can remove fluoride up to 90% and the treatment is exceptionally practical. Regeneration is needed after at regular intervals of 4–5 months and viability of adsorbent for fluoride removal reduces after every regeneration cycle. Mckee and Johnston investigated the utilization of powdered activated carbon for fluoride removal and accomplished noble outcomes [75]. The procedure is pH dependent with noble results just at pH 3.0 or less. Hence, the utilization of this material is costly because of need of pH alteration. Activated alumina method for defluoridation is being prolifereated in a few villages by the voluntary organizations funded by UNICEF or different agencies to give safe drinking water. Sarita Sansthan, Udaypur, Rajasthan is spreading the method with the viable help of UNICEF by giving a bucket (approximately 20 L capacity) fitted with a microfilter at the bottom containing 5 kg of activated alumina. The point of interest and restriction of adsorption are given below:

Interest

1. Ease of operation.
2. Adsorption procedure is worthwhile
3. High productivity for fluoride removal and can remove up to 90% fluoride.

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4. Produce high quality water.
5. Regeneration is conceivable.

Restriction

1. Disposal of depleted adsorbents and concentrated regenerated makes issue.
2. Interference because of the vicinity of different anions may bring about competition for active sites on adsorbent.
3. Drop in removal effectiveness after regeneration step.
4. Highly pH subordinate.
5. High concentration of total dissolved salts (TDS) can brings about fouling of the alumina bed.

3. Conclusions and future perspective

This review has endeavored to cover an extensive variety of procedures which have been utilized so far for the removal of fluoride from the drinking water and industrial wastewater. The traditional system of removing fluoride from drinking water is liming and the attending precipitation of fluoride. Then again, the weaknesses of the vast majority of these strategies are high operational and upkeep costs, auxiliary contamination, for example, generation of toxic sludge and so on and intricated process included in the treatment. Distinctive tecnnologies systems, utilized for defluoridation, were discriminatingly examined. It was reasoned that coagulation strategies have by and large been discovered compelling in defluoridation, however they are unsuccessful in conveying fluoride to fancied concentration levels. The quest for option and appropriate fluoride removal methods thusly still stays of curiosity.

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