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ORIGINAL ARTICLE

A Review on Defluoridation of Ground Water by Various Techniques: A Comparative Study

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ABSTRACT

At present scenario fluoride contamination of groundwater is a worldwide threat to the living world. A number of techniques are being utilized in all over the world to remove fluoride from groundwater. This paper focuses on the defluoridation of groundwater by different methods such as adsorption, ionexchange, coagulation-precipitation, electro-coagulation and types of membrane separation process viz. reverse osmosis, nanofiltration and electro-dialysis. Each one has been depicted with its merits and demerits on the basis of the literature survey and laboratory experiments. From the review presented here, it is cleared that the choice of the technique should be based on the local need and conditions such as contamination source and the groundwater quality. Fluoride contamination has been known as a major issue in many parts of the world including India. Therefore, for a healthy world it is necessary to spread the awareness among the people and defluorinate the contaminated groundwater before consumption.

Keywords: fluoride contamination, groundwater, defluoridation, electro-coagulation.

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INTRODUCTION

Ground water is the most important source of drinking water which is used all over the world. In past years population explosion has been made the ground water polluted. Arsenic, chloride, fluoride, nitrate, sulfate, iron, magnesium, boron and heavy metals are few contaminates that are of great concern if not present within permissible limit (Brindha *et al.*, 2011). It is most important that water which people drink and use for other purpose should be clean water. Due to use of contaminated drinking water human being undergoes from variety of water born diseases. World Health Organization (WHO) reports that 80% of all diseases in human being are caused by water. Industrialization and urbanization seem to main cause of ground water contamination. Contamination of drinking ground water by fluoride is one such example. The intake of the fluoride to the human body is usually done through drinking water. Other than the drinking water, some food items, beverages and tooth pastes are also the source of fluoride consumptions. According to WHO as well as Bureau of Indian Standards (BIS), the permissible limit for fluoride in drinking water is 1.5 mg/l (WHO, 2004 and BIS, 2012). Fluoride is an essential component for mineralization of bones and teeth in low amount (<1mg/l) but high

amount (>1.5mg/l) amount may result in dental and skeleton fluorosis (Battula *et al.,* 2016).

Sources of fluoride in ground water are both natural and anthropogenic. Natural sources include weathering and longtime contact of different kinds of fluoride rich ores, minerals and rocks which are naturally found in earth's crust, such as fluorspar, cryolite, fluoraptite, sellaite, fluorite, phosphorite, theorapatite and topaz (Dargahi *et al.*, 2016 and Singh *et al.*,2001). Fluoride as fluorspar is prominently found in sedimentary rocks. Anthropogenic sources include several human activities such as use of pesticides, mining and brick kilns (Datta *et al.* 1996). In many parts of India, the groundwater is contaminated with the high amount of fluoride rich pollutants. The issue has become more restricted to the areas with very low rainfall. The amount of fluoride found in those areas is far above the permissible limits as per WHO and BIS standards (Sankhla *et al.*, 2018).



Fig. 1: Sources of Fluoride in the environment (Vithanage and Bhattacharya, 2015)

Defluoridation of groundwater is the best way to root out this problem. A number of methods existing so far for this purpose and extensive research have been done on these methods. The literature regarding this is enlisted in this review.

TECHNIQUES USED FOR DEFLUORIDATION OF GROUNDWATER

During the past years, several approaches have been used to remove fluoride from drinking ground water, including adsorption, ion exchange, coagulation and precipitation, electro-coagulation and membrane separation process. Here all the above approaches are presented briefly with their point of merits and demerits.

1. ADSORPTION:

The method involves the passage of water through a contact bed where fluoride is adsorbed on the matrix. Among all the method of defluoridation adsorption is most popular method as it allows the access to a great variety of adsorbent.

Research has focused on different adsorbent materials that are capable of effectively removing fluoride from ground water such as activated carbon, activated alumina, activated alumina coated silica gel, activated saw dust, activated cocoanut shell carbon, activated fly ash, calcite, groundnut shell, coffee husk, rice husk, magnesia, serpentine, tricalcium, phosphate, bone charcoal, activated soil sorbent, carbion, deflouron-1, defluoron-2 (Kariyanna, 1987, Barbier, 1984, Muthukumaran *et al.*, 1995, Rongshu *et al.*, 1995, Min *et al.*, 1999, Wang, 2001, Nava *et al.*, 2003, Padmavathy *et al.*, 2003, Thergaonkar, 1971), different clays (Ramdani *et al.*, 2010 and Zevenbergen *et al.*, 1996), solid industrial waste

like red mud, spent bleaching earth, spent catalyst (Chaturvedi *et al.*, 1990, Cengeloglu *et al.*2002, Lai and Liu, 1996, Piekos and Paslawaska, 1999, Xu *et al.* 2011) natural and synthetic zeolite and other low cost adsorbent (Onyango *et al.* 2006). The effectiveness of these adsorbents depends on several factors such as temperature, pH, initial concentration of fluoride, contact time, co-existing ions, sorption kinetics, sorbent/ sorbet concentrations, type of adsorbent, size of adsorbent and surface area of adsorbent (Mohapatra *et al.* 2009).

The most commonly used adsorbent for defluoridation of drinking water is activated alumina and activated carbon. The efficiency of activated alumina in defluoridation process gets influenced by hardness and surface loading (Meenakshi and Maheshwari, 2006)

Raul *et al.* (2012) have studied the use of iron oxide-hydroxide nanoparticles for the removal of fluoride from water. The studies revealed that the iron oxide-hydroxide nanoparticles have been found to be a powerful adsorbent for defluoridation of water. The maximum defluoridation capacity of iron oxide-hydroxide is found to be 16.70mgg⁻¹ for fluoride at room temperature and also affected by pH of the medium.

The merits and demerits of adsorption are as follows (Meenakshi and Maheshwari, 2006): **Merits:**

- **1.** Cost effective and simple method.
- **2.** Provide wide range of adsorbent materials.
- **3.** Defluoridation up to 90%, hence highly productive method.
- **4.** Adsorption procedure is worthwhile.
- **5.** Regeneration is conceivable.
- **6.** High quality water is produced.

Demerits:

- **1.** pH sensitive method.
- **2.** Problem of disposal of fluoride rich adsorbent.
- **3.** High amount of Total Dissolved Salts (TDS) causes fouling of the alumina bed.
- **4.** Presence of different anions like sulfate, phosphate or carbonates causes ionic competition.
- **5.** Effectiveness of adsorbent decreases after regeneration process.

2. ION-EXCHANGE PROCESS:

This method involves the removal of fluoride with anion-exchange resin containing quaternary ammonium functional groups. The following reaction occurs during the process-

$$Matrix-NR_3^+Cl^- + F^- \rightarrow Matrix-NR_3^+F^- + Cl^-$$

The chloride ions of the resin are replaced by the fluoride ions due to their higher electro negativity. When all the sites on the resin are occupied, the process stops. Then the resin is washed with dissolved sodium chloride salt containing supersaturated water so that the fluoride ions could be replaced by the new chloride ions leads to the resin recharge to initiate the ion-exchange process again (Meenakshi and Maheshwari, 2006).

Meenakshi *et al.* (2007) have investigated the fluoride removal capacity of chelating resin Indion FR10 (IND) and ceralite IRA 400 (CER). They found chelating resin more selective than an anion-exchange resin for defluoridation.

Sundaram *et al.* (2009) have studied the fluoride sorption using organic-inorganic hybrid type ion exchangers. The ion-exchanger polyacrylamide was modified with $Al(NO_3)_3.9H_2O$, $ZrOCl_2.8 H_2O$ and $Ce(SO_4)_2.4H_2O$. The fluoride removal capacity of Ce-Ex is 2290 mg/kg which is higher than others.

Ku *et al.* (2011) have studied the use of aluminum-loaded Duolite C-467 resin for fluoride removal from aqueous solution. The removal of fluoride was found to relatively constant over entire solution pH.

Chubar *et al.* (2005) have studied the use of a novel ion-exchanger for the removal of fluoride, chloride, bromide and bromated ions.

Ho *et al.* (2004) have studied the ion-exchange property of mesoporous Titaniumoxohydroxide for the removal of fluoride from water. This process is expensive and leads to cause of membrane fouling.

The merits and demerits of ion-exchange process are as follows:

Merits:

- 1. Defluoridation of water occurs up to 90-95%.
- **2.** Taste and colour of water retain.

Demerits:

- **1.** Having high cost.
- 2. Preparation of resin is a tedious and difficult process.
- **3.** Pretreatment required maintaining the pH.
- **4.** Presence of other ions like sulfate, carbonate and phosphate can reduce the efficiency of process.
- **5.** Regeneration of resin is an issue as it prompts fluoride rich waste, which has to be treated before disposal.
- **6.** Reaction time is too long.

3. COAGULATION-PRECIPITATION:

Coagulation-precipitation method involves the use of coagulating substance to remove the fluoride from the contaminated water. The addition of chemicals such as lime and alum leads to the precipitation of fluoride as insoluble calcium fluoride. In first step, precipitation done by the addition of lime, then in second step, alum is added resulted in coagulation. The best outcome found between pH ranges of 5.5-7.5 (Potgeiter, 1990). Nalgonda technique which is based on the principle of coagulation- precipitation, involves the use of alum, lime and bleaching powder. This well-known technique introduced by NEERI, includes several steps, such as rapid mixing, chemical interaction, flocculation, sedimentation, filtration, disinfection, sludge thickening and removal (Nawalkhe, 1974 and Technical Digest, 1978).



Fig. 2: Nalgonda technique (Feenstra et al., 2007)

The merits and demerits of coagulation- precipitation process are as follows: **Merits**:

- **1.** Most effective and commonly used technique for the removal of fluoride from groundwater.
- **2.** More practical technique than other fluoride removal techniques.
- **3.** Simple and easy to understand.

Demerits:

- **1.** Large amount of sludge production.
- **2.** Required daily addition of chemicals.
- **3.** Low effectiveness for water having high TDS and hardness(Roy and Dass 2013)
- 4. Increase in residual aluminum in the treated water (Gupta, 1997).
- 5. Required high amount of chemicals.
- **6.** Required skilled persons.
- 7. The amount of sulfate ion increases by the use of alum
- **8.** Because of soluble toxic aluminum fluoride complex, Nalgonda technique is undesirable for the removal of fluoride (Apparao and Kartikeyan ,1986)

4. ELECTRO-COAGULATION (EC):

Electro-coagulation (EC) method involves the use of processed water through electrolysis. This method can remove the contaminants from the water which are difficult to remove by other chemical treatments. In this method an electro-chemical cell is connected metal electrodes and taking power from DC supply. The coagulant is generated at anode which is made up of aluminum metal, where the aluminum hydroxide flocs are produced. The negatively charged fluoride ions precipitated out in the form of sludge. This method proved to be an efficient method for the defluoridation of groundwater.

Battula *et al.* (2016) have studied EC for effective removal of fluoride from groundwater. The studies showed that aluminum electrode is suitable for fluoride removal. The maximum fluoride removal was observed at 30V at neutral pH. The defluoridation increased by increasing the reactive surface and by decreasing the inter electrode distance.

Naim *et al.* (2015) studied the EC by using bipolar aluminum electrodes for the defluoridation of water. Complete defluoridation from 6.44 mg/l analar sodium fluoride was done in about 15 min, at optimum pH with the speed of agitation 300 rpm. The fluoride removal from water by electro-coagulation was more for analar NaF solutions than commercial NaF solutions.

Takdastan *et al.* (2014) studied the use of iron and aluminum electrodes for the removal of fluoride from drinking water by EC. The study revealed that more efficient defluoridation was found at pH 3-6.5 and concentration of fluoride was decreased from 5 ml/l to lower than 1 mg/l. The chemical reaction followed the first order kinetics. The efficiency of fluoride removal was enhanced by increasing the voltage, number of electrodes, reaction time and decreasing the electrode distance.

The merits and demerits of EC are as follows (Mollah et al., 2001)-

Merits:

- **1.** Low sludge is produced that can be easily set.
- 2. The water gained after the treatment is colorless, odorless and safe to drink.
- **3.** Required basic equipments and less support cost.
- **4.** Easy to operate.
- **5.** Removal of very small colloidal particles that has been difficult to remove by traditional coagulation-flocculation method.

Demerits:

- **1.** Need of high conductivity of the wastewater suspension.
- 2. Gelatinous hydroxide may be solubilized.
- **3.** Loss of productivity of the EC unit due to the framing of an impermeable oxide film on the cathode.

4. The sacrificial anodes need to be replaced periodically as dissolved into wastewater as an effect of oxidation.

5. MEMBRANE SEPARATION PROCESS:

In membrane separation process, particles are separated on the basis of their molecular size and shape by the use of a semi-permeable membrane which is a thin, porous or non porous polymeric film made up of a gas or a liquid or metal material or ceramic. The membrane must not to be dissolved, break or disintegrate (Seadar and Heneley, 2005). Reverse osmosis, nanofiltration, dialysis and electro-dialysis are the most commonly used membrane separation process.

5.1. Reverse Osmosis (RO):

In this type of membrane separation process, anions are removed through the semipermeable membrane by applying higher pressure on water (Waghmare and Arfin 2015) RO has been proved to be the most effective method in removing fluoride from water as it can remove more than 90% of fluoride regardless of initial fluoride concentration (Ndiaye *et al.*, 2005).

Briao *et al.* 2014) have studied the RO for desalination of water from the Guarani Aquifer System for drinking purpose in southern Brazil. 100% of fluoride, 97% of total dissolved solids (TDS) and 94% of sulphate ions was rejected by RO at 2MPa pressure and 1.61m/s of cross-section flow velocity.

Gedam *et al.* (2012) have studied the RO by using polyamide membrane for the removal of pollutants in groundwater in Chandrapur district. The studies revealed that 95-98% of fluoride was removed from water.

Diawara *et al.* (2011) have utilized the low-pressure RO membrane for removing fluoride and salinity of brackish drinking water in Senegal village. The studies showed that 97-98.9% of fluoride was removed from water.

The merits and demerits of RO are as follows-

Merits:

- **1.** More than 90% of fluoride can be removed.
- **2.** The RO membrane can be recovered again.
- **3.** Most efficient especially for fluoride removal.
- **4.** Other dissolved solid impurities can be removed simultaneously.
- **5.** No disturbance by the presence of other ions.
- 6. Works under wide range of pH.
- 7. Water quality does not deteriorate.
- **8.** Not required too much chemical and labor.
- 9. Water gets treated and purified only in one step.

Demerits:

- **1.** Not ideal for rural settlements.
- 2. Required high setup and maintenance cost (Roy *et al.*,2013)
- 3. Complete demineralization of water (Roy *et al.*,2013).
- **4.** Huge quantity of water as brine is wasted.
- 5. The water becomes acidic and needed to be correct the pH.

5.2. Nano Filtration (NF) Membrane Process:

NF is the newly introduced membrane separation process used for the removal of fluoride from water. This process requires relatively low pressure and removes primarily the larger dissolved solids as compared to RO. Thus, it is more prudent. More over the permeability of NF membrane is greater than RO membrane, thereby making the process more efficient for desalination of brackish water (Lhassani, *et al.*, 2001).

Bejaoui *et al*, (2014) have studied the membrane separation through RO and NF for defluoridation of water and metal packaging industrial effluent and found more than 90% of fluoride retention by both the process.

Pontie *et al.* (2008) have investigated the utilization of NF for defluoridation of water for large scale pilot plants for future.

The merits and demerits of NF are as follows-

Merits:

- **1.** Works under wide pH range.
- **2.** Productivity is high.
- **3.** No need of chemicals.
- **4.** No interference due to the presence of other ions.
- 5. Effective for pesticides, microorganisms, suspended solids and toxins.

Demerits:

- **1.** Comparatively more expensive method.
- 2. Membrane related issues like membrane degradation, fouling or scaling.
- **3.** Demineralized the water and need to be added the essential minerals after the treatment.

CONCLUDING REMARKS

Researchers have adopted different techniques for the purpose of defluoridation of contaminated water so far. On the basis of the review presented here, it has been cleared that each technique used for the defluoridation of contaminated groundwater, has its own merits and demerits. If any particular technique is suitable at any place, it may not fulfill the requirements at another place. Therefore, the available local conditions, resources, costing should be evaluated properly before implementations of a particular method. Groundwater of a particular area should be assessed thoroughly before its utilization for domestic purposes and a suitable method can be selected for its remediation.

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