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Fluoride removal from Drinking Water

Aster Woldu (M.Sc process engineering)

Adama Science and Technology University; School of Mechanical, Chemical and Materials Engineering, Chemical Engineering Department, Adama, Ethiopia.

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Abstract

The presence of fluoride in drinking water has a profound effect on teeth and bones. WHO recommended, up to a small level (1–1.5 mg/L) strengthens the enamel. Dental and skeletal fluorosis and bone deformities are observed in people after the excessive intake of fluoride. High fluoride concentrations in groundwater, up to more than 30 mg/L, occur widely, in many parts of the world. Ethiopia is listed on the countries, which is reported repeatedly by the effect of Fluoride on the health of the people. This review is aimed at providing highlighted information on effects on the element on health, the technologies used to eliminate it from drinking water.

The fluoride removal technics can broadly divide in two; the membrane and adsorption methods. The membrane techniques use membrane to screening out the fluoride using different ways. It comprises reverse osmosis, Nano filtration, dialysis and electro-dialysis. The adsorption, which is a conventional technique, deals with adsorbents such as: alumina/ aluminum based materials, clays and soils, calcium based minerals, synthetic compounds and carbon based materials. Recently DE metals also attracting an interest in the removal of fluoride as an adsorbent.

Keywords: Fluoride, Treatment technologies, adsorption, DE metals,

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1. Introduction

The relationships between the natural environmental factors and the health of the inhabitants in a terrain have been known for centuries. In this era, all over the world, the pollution of surface and groundwater create a serious risk not only to human beings but also towards aquatic lives due to the presence of emerging contaminants related to industrialization and development. And also, potable water is rare since clean water does not found in nature; water from natural sources contains different dissolved salts in addition to suspended particles. Mainly, the salts and minerals in surface and groundwater originate from the soil and rock with which it is in contact (Gitari, W.M. et al., 2017).

Groundwater is the best available drinking water for rural communities of the developing countries, where there is no pipe-bore water. For example in Ethiopia; according to estimates of the Ethiopian Ministry of Water and Energy, more than 14 million people in Rift Valley region rely on fluoride-contaminated drinking water (Annette Johnson1, et al., 2011). Dependence on groundwater as main drinking-water seems appropriate, because the surface water is susceptible to pollution related to human and animal excretions. Even if; consuming groundwater rather than surface water minimizes the occurrence of waterborne diseases, there might be a number of toxic effects produced related to the water (Annette Johnson1, et al., 2011; Meththika Vithanage and Prosun Bhattacharya, 2015; Gitari, W.M. et al., 2017). Contamination of drinking water by fluoride is one such example. World health organization (WHO) classified fluoride as one of the toxic of human; In addition to arsenic and nitrate, that are creating significant health problems (Meththika Vithanage and Prosun Bhattacharya, 2015).

The concentration for the contamination of fluoride is above 1.5mg/l, but below this value, it is reported its advantage for the development of healthy teeth (WHO, 2011). Consumption of fluoride for length time will result in diseases known as dental and skeletal fluorosis (Meththika Vithanage and Prosun Bhattacharya, 2015; Haldar, D., et al., 2020). The severity of either fluorosis is much dependent on the concentration of fluoride and the period of exposure (Haldar, D., et al., 2020). Therefore; it is essential to decrease the amount of fluoride in drinking water to a recommended level for drinking.

Worldwide Lots of researches are done to resolve the subjects matter including technology development, identification of source and health impact at a particular location related to fluoride. The conventional technologies used for treatment of fluoride are adsorption, ion-exchange, precipitation/coagulation, and membrane processes (Krishna Kumar Yadav, et al., 2019; Vivian Kimambo, et al 2019; Haldar, D., et al., 2020).

Mainly this material is discussed the effect of fluoride on health and its remediation technic for the removal from drinking water.

2. Fluoride in Water

Fluoride contamination of groundwater by natural as well as anthropogenic sources is a major problem of concern since it is imposing a serious threat to human health (WHO, 2011). There are many ores, minerals, and rocks present inside the earth's crust, which are the natural sources of fluoride. In these natural sources; the fluoride occurs mainly as sellaite (MgF_2), fluorspar (CaF_2), cryolite (Na_3AlF_6) and fluorapatite [$3Ca_3(PO_4)_2$ Ca (F Cl_2)] (Abdollah Dargahi et al., 2016; Rohana Chandrajith, et al., 2020). As fluorspar it is present mainly in sedimentary rocks and as cryolite. Weathering and geochemical dissolution of fluoride-bearing of these rocks is the mentioned natural condition for the realizing of fluoride in to the water body. The long contact time of fluoride-bearing ores, minerals, and rocks with groundwater, result in a constant leaching of fluoride ions that is accountable for the high fluoride concentration in groundwater as well as oceanic water (Tesfaye Akafu et al., 2019; Vivian Kimambo, et al, 2019; Rohana Chandrajith, et al., 2020).

The realize of fluoride from industrial activities includes discharge of wastewater from fluorosilicone and fluorocarbon polymer synthesis, fertilizers and pesticides, electroplating operations, coke manufacturing, glass and ceramic manufacturing, electronics manufacturing, steel and aluminum manufacturing, metal etching with hydrofluoric acid, and wood preservatives (Tesfaye Akafu et al., 2019;).

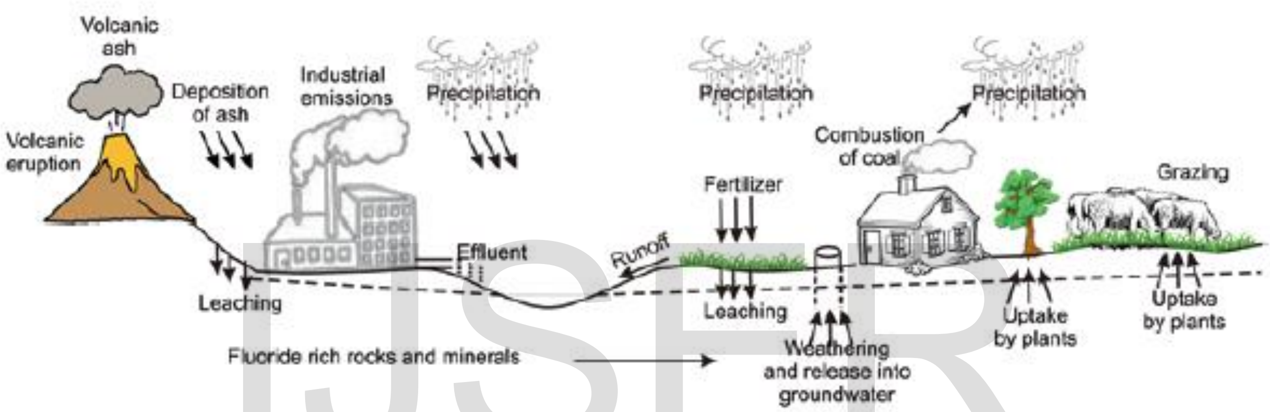

Figure: 1-Diagram showing the fluoride sources in the environment (Meththika.V and Prosun. B, 2015)

Fluoride can enters into the human body through different sources such as water, food, air, medicine, and cosmetics. Among these, drinking water is the most common source which makes fluoride available to human beings (P Senthil Kumar, et al., 2019). Fluoride is known to have both beneficial and detrimental effects on health, depending on the dose and duration of exposure (Tesfaye Akafu et al., 2019 ;). Report by the World Health Organization recommends levels of 0.5 mg/l fluoride in drinking water as beneficial to health but levels in excess this may be harmful. At higher concentrations; one may suffer from dental fluorosis and above 3.0 mg/l skeletal fluorosis (M. Mohapatra, et al., 2009; M. Mupa, et al., 2016*;* Gitari, W.M. et al., 2017; P Senthil Kumar, et al., 2019). See table for detail.

Fluoride in drinking water has a significant influence on teeth and bones. Fluoride replace/displace hydroxide ions from hydroxyapatite, $Ca_5(PO_4)_3OH$, which is the essential mineral of teeth, specifically it helps the enamel and bones, to be harder and tougher fluoro-apatite, $Ca_5(PO_4)_3F$. Up to some concentration, this strengthens the enamel. However, if the concentration of fluoroapatite is higher, it become less soluble than hydroxyapatite and that means the conversion of a large amount of the hydroxyapatite into fluoroapatite makes the teeth denser, harder and more brittle and after prolonged exposure, the bones also become denser, harder and more brittle. In the teeth this causes mottling and embrittlement, a condition known as dental fluorosis. With prolonged exposure at higher fluoride concentrations dental fluorosis progresses to skeletal fluorosis (M. Mohapatra, et al., 2009; M. Mupa, et al., *2016;* Tesfaye Akafu et al., 2019; P Senthil Kumar, et al., 2019).

Other health problems associated with drinking water with high fluoride concentration include lowering of Intelligent Quotient (IQ) and long term damage to brain, osteosarcoma (a type of bone cancer), liver, thyroid and kidney plus a host of other health problems (M. Mohapatra, et al., 2009; M. Mupa, et al., *2016;* Q. Duan, et al., 2018)*.* To be amaze and more curious; the accumulation of fluoride over a long period of time can even lead to changes in the DNA structure (P Senthil Kumar, et al., 2019; Krishna Kumar Yadav, et al., 2019).

Table: 1 - Effect of prolonged use of drinking water on human health, related to fluoride content (M. Mupa, et al., *2016*).

F^{-1} concentration, mg/l	Health outcome
<0.5	Dental caries
0.5–1.5	Optimum dental health
1.5–4.0	Dental fluorosis
4.0–10	Dental and skeletal fluorosis
>10.0	Crippling fluorosis

Figure: 2-Dental fluorosis-affected child in Silence (P Senthil Kumar, et al., (2019).

The highest fluoride contamination is reported from numbers of countries such as India, Mexico, Pakistan, Egypt, Ethiopia, Saudi Arabia, Niger, USA, Senegal and China (P Senthil Kumar, et al., 2019; Vivian Kimambo, et al 2019)

3. Conventional Technologies Used for Fluoride Removal

Different conventional technologies are used to remove fluoride form drinking water, Including precipitation/coagulation, membrane-based processes, ion-exchange methods, and adsorption methods (Maurice. S and Hitoki. M, 2006; M. Mohapatra, et al., 2001; Meththika.V and Prosun. B, 2015; Abdollah. D et al., 2016; Lobna Mansouri, et al., 2019: Krishna. K, et al., 2019).

3.1 Coagulation and flocculation: Figure-3 illustrates the, one of conventional chemical technique that is aged for this purpose, reported in late 1975 (P Senthil Kumar, et al., 2019). Three chemicals are applied (lime, alum and bleaching chemical) sequentially to remove fluoride from the contaminated water as $Al^{-}F$ Complex and CaF_2. Nalgonda technique, which is first discovered in India but currently also widely used in Africa, includes number of operational steps, such as rapid flash mixing, chemical interaction, flocculation, sedimentation, filtration, disinfection and sludge thickening and removal. The fluoride ion is adsorbed on flocs of aluminum hydroxide in solution (Kwai Malak Kwai Kut, et al., 2016; P Senthil Kumar, et al., 2019). Alum [hydrated aluminum sulfate $Al_2(SO_4)_3 \cdot xH_2O$] is the most commonly used coagulant for fluoride flocculation. Lime is added as the process is carried out to generate alkaline conditions. Disinfection is achieved using bleaching powder. After thorough stirring, coagulation occurs into flocs which settle at the bottom.

Aqueous fluoride removal by the Nalgonda Technique is not yet fully understood but it is described as co-precipitation of mechanism (Kwai Malak Kwai Kut, et al., 2016; Feleke Zewge, 2016; P Senthil Kumar, et al., 2019).

3.2 Adsorption: Adsorption is considered as attractive technologies for fluoride removal. The advantage of Adsorption basing in its' effectiveness, suitable, easy to design and operate, and essentially it is low cost (Kwai Malak Kwai Kut, et al., 2016).

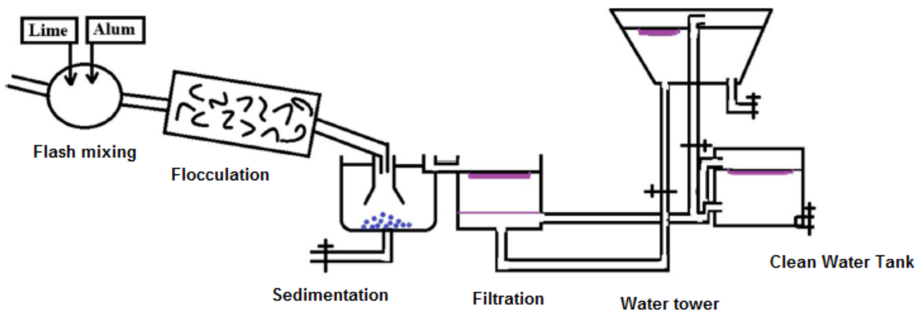

Figure: 3- An example of a conventional Precipitation/coagulation fluoride removal technique (P Senthil Kumar, et al., 2019)

At community and household levels is more preferable because of its low cost and ease of operation, high efficiency, easy accessibility, environmental benignity, and needless of operational skill and electric power to run, and the adsorbents can in principle be reused and recycled. Fluoride adsorption from water is controlled by pH, temperature, co-existing ions, sorption kinetics, sorbent particle size, sorbent/sorbate concentrations and adsorbent surface area (M. Mohapatra, et al., 2009; P Senthil Kumar, et al., 2019). Different carbon based adsorbent like graphite, moringa oleifera, bone chart (Sani et al., 2016; Susan S.A. et al., 2019), chitin, chitosan, and fly ash, zeolites (M. Mohapatra, et al., 2009; P Senthil Kumar, et al., 2019), etc. have been studied and used for the same purpose adsorption (Meththika.V and Prosun. B, 2015; P Senthil Kumar, et al., 2019). Table-2 shows different adsorbent investigated in Ethiopia for the removal of fluoride from water. In different parts of the world; inorganic adsorbent for fluoride removals like titanium-rich bauxite, activated alumina, and manganese oxide-coated alumina have been studied and applied for the past few years (P Senthil Kumar, et al., 2019).

Table: 2- De-fluoridation capability of different materials studied in Ethiopia

Fluoride Con(mg/l)	Material used	Adsorption capacity (mg/g)	References

50	Waste generated during aluminum sulfate manufacturing process	153.8	(Nigussie et al., 2007)
18-73	Red Gullale soil (RGS) fired at 500^{0}C.	0.893	(Gomoro et al., 2012)
-	Mordenite zeolites	0.47	(Gómez- H et al.2013)
20	Hydrous Aluminum (III)- Iron (III)-Manganese (IV) ternary mixed oxide	23.99	(Woyessa et al., 2014)
-	Aluminum hydroxide based	26.2	(Mulugeta et al., 2014
0-200	Nano-hydroxyapatite/ stilbite (nHAST) composite	9.15	(Sani et al., 2016
0-200	Bone Char	1.08	(Sani et al., 2016)
2–29	Iron ore	1.72	(Kebede et al., 2016)

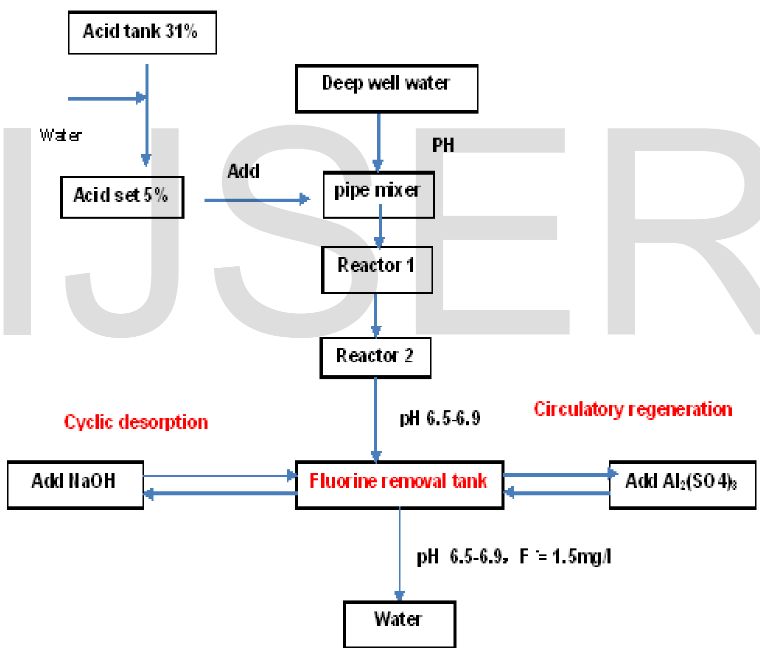

Figure: 4- de-fluorination process **of Hawwasa Industrial Parks** using an activated aluminia adsorbents.

To give an example on the adsorption process, basing activated alum; in Hawwas Industrial park, the process for defluorization of fluorine from 3.8 mg/l to 1.5mg/l is carried out using activated alumina adsorption. To adjust the deep well water pH and to reduce the level of bicarbonate; HCl is mixed with the water and react to generate CO_2. The accustomed water sent to adsorption tank, to adsorb the pollutant present in the water. The regeneration is done by chemical means; NaOH, $Al_2(SO_4)_3$ used for circular desorption and regeneration. See figure-4 for clarification.

3.3 Membrane filtration: Membrane filtration is also the well-known technologies used for this task. Due to its high removal capacities; in the late 1980s, reverse osmosis (RO) is considered as an alternative method to conventional waste treatments (Meththika.V and

Prosun. B, 2015; P Senthil Kumar, et al., 2019). In reverse osmosis, at one side of the semipermeable membrane, the hydraulic pressure is exerted which forces water across the membrane thus leaving the wastes behind (M. Mohapatra, et al., 2009). When porous materials are applied as a membrane the process insure nano and ultra-filtration for inorganic by diffusion, size exclusion, charge repulsion and adsorption (M. Mohapatra, et al., 2009). But in the case of reverse osmosis; nonporous materials are used and the mechanism for transportation is followed by the solution–diffusion model wherein solutes dissolve in dense material and then diffuses through the membrane down by the concentration gradient (M. Mohapatra, et al., 2009). Despite the facts that reverse osmosis can completely remove minerals from water with very low or practically no selectivity for monovalent ions; it has a reported limitation related to high operating pressure, low permeate flux and high energy requirements (M. Mohapatra, et al., 2009). PH and temperature are majorly affecting the removal of fluoride from water. P Senthil Kumar, et al., (2019), reviewed that, the removal of fluoride by reverse osmosis varies from 45 to 90% as the pH of the water is increased from 5.5 to 7.

4.3 Electrodialysis: The other membrane process that separates solutes by transporting the solutes through a membrane rather than using a membrane to hold the solutes while passing water through it is an electro-dialysis (M. Mohapatra, et al., 2009; P Senthil Kumar, et al., 2019). Electrodialysis is a technic used to eliminate ionic components from a solution through ion exchange membranes under the driving force of an electric field (M. Mohapatra, et al., 2009). It is relatively costly and easily influenced by coexisting ions. Recent advancement in the technology show that; the process registered no defects, it is minimized the precipitation risks of the bivalent salts and it consume low energy consumption for operation (M. Mohapatra, et al., 2009; P Senthil Kumar, et al., 2019). Jin et al. (2015) published the possibility of electro-dialysis operation to be carried out in a multiple ways, (with and without a chemical pre-treatment). P Senthil Kumar, et al., (2019) reported that in order to avoid the possibility of bivalent anions transport and accumulation; ACS-CMX membranes are used without pre-treatment, resulted in managing the transport in the following order (.Cl^-> F^-> HCO^-> SO^{2-}).

Since many parts of the world are frightened by fluoride-related health hazards caused by excessive exposure of fluoride in water and the cost of removing the ion with other technologies demand a high cost of processing that is related to power consumption, high cost of chemicals, scale formation and fouling for membrane and health risk related to the use of chemicals for purification; researcher investigated and applied natural materials for the application of Fluoride from water all over the place in the world (Nigussie et al., 2007; Krishna. K, et al., 2019).

The success behind for the adsorption ability of natural materials is the presence of functional groups such as imidazole, carboxyl, amide, carbonyl, sulfhydryl, phenol, amino and hydroxyl moieties. These functional groups are capable of adsorbing different types of pollutants, from water specially the metal cations (P Senthil Kumar, et al., 2019).

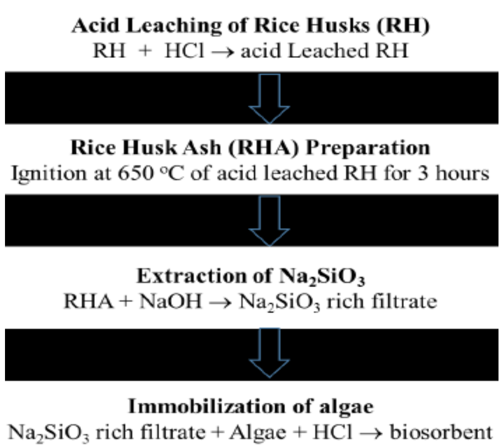

Figure: 5- preparation method for RHA biosorbet (M. Mupa, et al., *2016*)

Basing the technologies in defluoridation involves the basic principle of precipitation, ion exchange, adsorption and other electrochemical methods (Meththika.V and Prosun. B, 2015); many natural inorganic materials like minerals, soils, clays and building materials have been exploited in the removal process (Vinati et al., 2015). Plant materials like tamarind seed, serpentine and tamarind gel, duck weed; royale plants have also been investigated to adsorbed fluoride and proven to be used as an adsorbent for defluoridation (Amit Bhatnagar, et al., 2011; Annette Johnson1, et al., 2011; Feleke Zewge, 2016).

The applications of agricultural waste biomass are also checked for their capability in accumulating fluoride for example egg shell, corn cobs, tea and coffee waste, rice husk and its ash (M. Mupa, et al., *2016*), peanut shells, coir dust, saw dust, dry tree leaves and barks, wheat and rice bran and sea weeds have been studied and reported successfully for this purpose (M. Mupa, et al., *2016;* Krishna Kumar Yadav, et al., 2019; P Senthil Kumar, et al., 2019; Tesfaye Akafu et al., 2019).

3.5 Diatomaceous Earth as Adsorbent: History tells more than 100 years; diatomaceous Earth (DE or Diatomite) filtration technics has been a widely used for fluid purification in different industries. The first well-known use of DE for production of potable water was performed by the U.S. Army during World War II (https://www.waterworld.com). Diatomites (diatomaceous earth) are siliceous sedimentary rocks that basically contain amorphous hydrated or opaline silica ($SiO_2 \cdot nH_2O$) (80–90% silica) with varying amounts of impurities such as clay minerals (2–4% alumina), silica sand, carbonate minerals, iron oxides (0.5–2% iron oxide), and organic matter (Gitari, W.M. et al., 2017). To see it origin; Diatomaceous Earth is the remains of a singled celled aquatic plants called diatoms. These microscopic organisms such as plankton or algae have the distinguished capability to extract silica from water to produce a microporous exoskeleton. When the life cycle of this organism is completed, the organic matter decomposes and the skeletal remains accumulate to form an inorganic sedimentary deposit (https://www.waterworld.com).

The rise in interest for the use of Diatomaceous earth (DE) as an adsorbent in water treatment is due to its unique feature such as low density, high porosity that typically ranged from 10 to 200 μm (80–90% void), its' relatively chemical inertness, high permeability, small particle size ranged from less than 5 to over 100 μm, high surface area, ant its' low thermal conductivity (Gitari, W.M. et al., 2017). In addition; DE cannot undergo degradation that results in water; because it is inorganic (Gitari, W.M. et al., 2017; Tesfaye Akafu et al., 2019). The other advantage of DE is its cheap cost. When it is comparing diatomite with activated carbon is approximately 500 times cheaper than commercial activated carbon (Tesfaye Akafu et al., 2019).

In water treatment technology; the chemical composition and physical structure of diatomite give it great commercial value for a wide range of applications, such as beer filter aids in beer industry, helps to remove dyes from textile wastewater, removal of different water contaminates in water treatment, and act as sorbet for the sorption of heavy metal ions (Gitari, W.M. et al., 2017). But in Ethiopian; even if it's abundant availability; its application for water treatment and as of fluoride removal from drinking water has not been investigated properly (Tesfaye Akafu et al., 2019)

Research reported that raw diatomaceous earth (DE) has a low fluoride removal potential (Tesfaye Akafu et al., 2019). The highest percent fluoride removal achieved at optimum adsorption conditions is between 23.4% and 25.6% for 8 mg/L fluoride at pH 2, contact time of 30 min, solid liquid ratio of 0.4 g/50 mL, and shaking speed of 200 rpm by A. A. Izuagie, et al. (2016). Means; to improve the fluoride removal efficiency; the DE surface is needed to modified (Gitari, W.M. et al., 2017; Tesfaye Akafu et al., 2019).

The DE surface modified by treating it with aluminum hydroxide was found to be an effective adsorbent for the defluoridation of aqueous solution and natural groundwater taken from Hawwasa and Zeway regions of Ethiopia (Tesfaye Akafu et al., 2019). In this work; at optimal operating conditions (contact time: 180 min, adsorbent dosage: 25 g/L, pH 6.7, at room temperature, and shaking speed: 150 rpm) the maximum percent fluoride removal and adsorption capacity is found 89% and 1.67 mg/g, respectively, for 10 mg/L fluoride-spiked.

Izuagie et al. (2016) reported the more effectiveness of Binary metal Al/Fe oxide-modified sorbent than either Al or Fe oxide-modified DE for fluoride removal. In this work; a substantial amount of fluoride (93.1%) was removed from solution when a sorbent dosage of

0.6 g/100 ml was contacted with 10 mg/l fluoride solution for 50 min at a mixing rate of 200 rpm. The synergy of the two metals in DE to achieve an increased fluoride removal created a motivation for the investigation and assessment of defluoridation capacity of a trimetal oxide-modified DE with a specific interest in the oxides of magnesium (Mg), cerium (Ce) and manganese (Mn). After reviewing the removal effectiveness of the fluoride removal effectiveness of Mg, Ce or Mn oxides individually or in combination with other metal oxides from water; Gitari, W.M. et al., (2017) studied, the optimized synthesis of Mg/Ce/Mn tri-metal oxide modified DE, characterization and Groundwater fluoride removal capacity of the sorbent was evaluated by batch method at various defluoridation conditions at optimized conditions is evaluated.

Form the investigation; Gitari, W.M. et al., (2017) reported sorbent's optimum fluoride uptake capacity is 12.63 mg/g at the initial fluoride concentration of 100 mg/l. At a sorbent dosage of 0.6 g/100 mL (contact time: 60 min, mixing speed of 200 rpm and temperature: 297 K), the fluoride removal is found greater than 93% for solutions containing initial fluoride concentration of 10–60 mg/l. To show the effect of pH on Fluoride removal, analysis is done at initial pH ranges from 4-11 for solutions initial fluoride concentration 9 mg/l with sorbent dosage of 0.6 g/100 ml. The removal efficiency is found to be greater than 91%. It is also indicated that K_2SO_4 is the most suitable for regeneration of spent Mg/Ce/Mn oxide-modified DE when it compared to Na_2CO_3 and NaOH.

4. Conclusion

A brief review on fluoride removal methods from drinking water has been presented. The health effects of the high and the low concentration of fluoride intake is illustrated. The fluoride can be removed in a numbers of technics; the technics can be simply classified membrane and adsorption techniques. Under the membrane methods, the technics employed semi membranes to sieving out the fluoride out are Reverse osmosis, nano and ultra-filtration. But Electro dialysis use membrane to separates solutes by transporting the solutes through a membrane rather than using a membrane to hold the solutes while passing water through it. Adsorption which is a conventional technique deals with adsorbents such as: alumina /aluminium based materials, clays and soils, calcium based minerals, synthetic compounds and carbon based materials.

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