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Review Article

Defluoridation of fluoride contaminated water by adsorption process: A review

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Abstract

Purification of water is crucial today to meet the clean water demand of the expanding population and diminishing water supplies. Because of dangerous chemicals and the ongoing flow of industrial effluent into water bodies without any basic treatment, the quality of both surface and ground water has declined. Waterborne illnesses brought on by ingesting polluted water place a heavy and quantifiable burden on human wellness and have a substantial economic effect on society. The processes like reverse osmosis, ultra-filtration, nanofiltration, and electrodialysis are the conventional methods for fluoride elimination from the water. Due to the disadvantages of the conventional process of defluoridation, these are not suitable for villages and poor communities. Adsorption is a widely used tertiary treatment method because it is simple to use, has good removal performance, and has a variety of on-field uses. Due to its simple manufacturing processes, high stability, significant specific surface area, etc., activated carbon (AC) has become quite popular. This review covers fluoride contamination in groundwater and the application of different types of methods for fluoride removal and also discusses the creation and efficacy of adsorbents produced from various carbonaceous and natural precursors and other materials, for the removal of fluoride, the function of activation procedures, and fluoride-specific modifications.

Keywords: Fluoride; contamination; adsorption; adsorbents; activated carbon

1. Introduction

Water is a necessary component for all creatures including human beings and animals, but various chemicals change the quality of water and make it unsafe for drinking purposes. According to the World Health Organization (WHO), 783 million individuals lack access to basic drinking water utilities, and by 2025, it is anticipated that more than half of the world's population would experience a scarcity of drinkable water (Lacson et al., 2021). There is a global shortage of fresh water as a result of a variety of natural and human activities that may disturb the quality of groundwater as well as surface water. Fluoride is typically found in nature in the combined form of fluorite (CaF_2), and cryolite (Na_3AlF_6), fluorapatite [$(Ca_5(PO_4)3F$]. Fluorite, cryolite, biotite, fluorspar, muscovite, and apatite are among the common minerals that contain fluoride (Bretzler et al., 2015). These minerals contain fluoride ions that come into the groundwater after the dissolution and contaminate the water. These are in constant contact with groundwater, which causes them to dissolve under various chemical circumstances (He et al., 2020). Leaching and enrichment are two geochemical processes that control the movement of fluoride between soil and groundwater, whereas weathering, rock modification, geothermal mixing, and evaporation are responsible for the movement of fluoride between rocks and groundwater (Olaka et al., 2016). Fluoride contents are frequently high in young volcanic rocks generated during the geological age of tertiary and recent times of the earth. Fluorine concentrations in alkalic igneous rocks are typically >100 mg/L, but fluorine concentrations in ultramafic igneous and volcanic rocks are around 1000 mg/L (Kut et al., 2016).

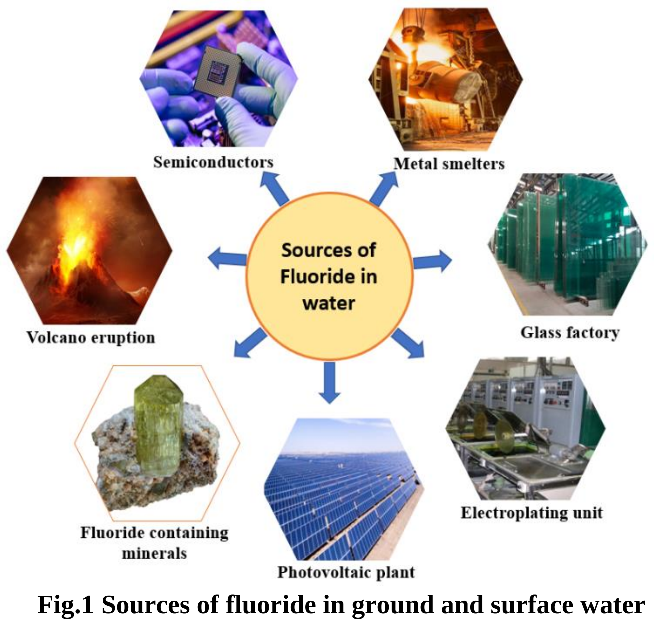

Fig.1 Sources of fluoride in ground and surface water

Wastewater that contains fluoride from electroplating, metalworking, electronics manufacturing, and other industries may pollute water after discharge (Iqbal et al., 2021). Different sources of fluoride in groundwater are presented in Fig. 1. The WHO states that the content of fluoride in drinking water that is more than 1.5 mg/L is unsafe for human health. Water having a high concentration of fluoride is not fit for drinking purposes because it causes dental and fluorosis in human beings. Fluorosis, a set of disorders, is a common occurrence for people who habitually drink water that has excessive amounts of fluoride. Recent studies have revealed that even soft tissues are susceptible to fluorosis, which is referred to as non-skeletal fluorosis (Patil et al., 2018). Long-time consumption of fluoride-contaminated water also causes liver damage, kidney failure, bone deformation, muscular pain, and cancer susceptibility (Keshavarz et al., 2019). Fluorosis is not curable but it can be prevented by eliminating fluoride from drinking water. This review is discussing fluoride contamination in groundwater and its remediation using various techniques. The adsorption method is the main focus with different types of adsorbents for fluoride elimination from the water.

2. Fluoride removal methods from the water

Several methods have been used over the last two decades including the Nalgonda method which is based on precipitation & coagulation, nanofiltration, ultrafiltration, reverse osmosis, electrodialysis as well adsorption method. The advantages and disadvantages of these methods are discussed below.

2.1 Nalgonda method

This technique works based on flocculation and precipitation principal. In this technique, several steps of operations involve such as quick mixing, flocculation after chemical interaction, sedimentation, sludge separation and disinfection of water (Swarnakar et al., 2007). A coagulant hydrated aluminum salt applies to flocculate the fluoride ions and lime is added to neutralize the alkaline condition of the water. Lastly, bleaching powder adds to disinfect the water.

2.2 Membrane filtration

Membrane treatment is one of the convenient and eco-friendly methods for the treatment of wastewater. It is a physical method without the use of any chemical reagents. In membrane treatment, the membrane used behave as a barrier and restricts the flow of impurities. Several studies have been done on membrane processes like Reverse osmosis, ultra-filtration and electrodialysis for fluoride removal (Platter., 2017).

2.3 Nanofiltration

Due to the great and unique membrane selectivity, nano-filtration appears to be the most effective approach of all membrane techniques for fluoride removal. A study demonstrated that the cross-flow method's composite polyamide NF membrane could successfully remove 98% of the fluoride from contaminated water (Chakrabortty et al., 2013). A membrane is also utilized in this procedure, which is generally a reverse osmosis-like event. While reverse osmosis uses membranes with slightly smaller pores, nanofiltration uses membranes with slightly larger pores. Due to wider pores, water filtering requires little pressure and energy. Due to technical difficulties and the high cost of the membrane, this technology for removing fluoride from wastewater is quite expensive (Diawara et al., 2008).

2.4 Reverse osmosis

Reverse osmosis (RO) is a process in which Fluoride contaminated water is entered via a semi-permeable membrane. This membrane allows water to move through it but restricts the transfer of impurities through it, in this way fluoride ions can be filtered out. If the content of impurity is high then the Nanofiltration method is used, this is a type of RO in which the pressure is kept low (Wan et al., 2021). According to one study, polyamide RO membranes obtained clearance rates of 95%–98% fluoride from water (Gedam et al., 2012).

2.5 Ion exchange

Water is allowed to flow downward via a column that is filled with an ion exchange resin during the ion exchange procedure. Resin gives good efficiency of fluoride removal, however, due to the existence of other anions such as carbonates, phosphates, sulfates, nitrates, etc. the process efficiency is quite low. The filter material needs to be rejuvenated with a mild acid/alkali solution when the resin becomes saturated. The resin is unprofitable and expensive, but it can be easily renewed (Yadav et al., 2018).

2.6 Electrodialysis

Electrodialysis is also a membrane treatment method similar to reverse osmosis but it uses current instead of pressure to remove ionic pollutants. The drawback of electrodialysis is that it works only when there is electricity. However, it is quite expensive, results in significant water loss from brine discharge consumes a lot of energy, has a large capital expenses, and is ineffective in areas where there is no electricity. Pre-filtration and post-pH/alkalinity correction may be necessary (Karunanithi et al., 2019).

2.7 Electro-coagulation

This is an easy and effective method for eliminating the flocculating agent created by the electro-oxidation of a sacrificial anode, which is often constructed of aluminum or iron. In this method direct current is applied to sacrificial electrodes that are dipped in an aqueous solution. This method combines the functions of electrochemistry, coagulation, and hydrodynamics, three fundamentally related processes that work together to remove contaminants (Waghmare et al., 2015).

2.8 Adsorption process

Fluoride removal by adsorption is well known and widely used method due to its cost-effectiveness and easy handling. Since the last decades, a variety of adsorbents has been used for defluoridation of water. The adsorbent may be originated from natural materials (clay minerals, zeolite), plant biomass (biochar, activated carbon), and synthetic materials (ion exchange resins). Different types of adsorbents are originated from the waste biomasses, agricultural residues and plant residues. Biomass based adsorbents have been used for defluoridation of water. But very less adsorbents have been applied for the removal of fluoride from the drinking water. So, there is a need to work on this.

3. Materials used for fluoride removal

3.1 Clay minerals

Basically, clay is a mineral composite that has been finely ground up and is made up of hydrous aluminum silicate and various other minerals and impurities. The most significant phyllosilicate mineral forms are beidellite, montmorillonite, nontronite, saponite and hectorite, collectively known as smectite (Macias-Quiroga et al., 2018). Fluoride from water can be absorbed by both burnt clay and clay powder. If the clay is accessible locally, removing fluoride with clay is fairly inexpensive. Clay has a limited capacity to absorb fluoride, and the process of removing fluoride takes much time as compared to other adsorbents. But, the adsorption capacity of the clay-based adsorbents can be increased by its surface modification using chemical and physical processes.

3.2 Zeolite

Zeolites are composed of aluminosilicates having crystallite molecular arrangement. The local electric field and spatial constraint properties in zeolite work during the adsorption phenomenon (Chai et al., 2021). Zeolites are applied as adsorbents for the removal of various pollutants due to nano range structure and adsorptive nature. Zeolite is considered to be porous in nature, with sufficient surface area, and active sites for impregnation that help to improve adsorption efficiency (He et al., 2017).

Table 1. Application of different types of adsorbents for fluoride removal

Adsorbent	Modification	Adsorption Capacity	References
Zeolite	La(III)	20.83 mg/g	(Lai et al. 2018)
Zeolite	Zirconium and pulsed sonication	32.98 mg/g	(Savari et al., 2020)
Clay (Aluminosilicate)	NaOH	2.57 mg/g	(Obijole et al., 2021)
Clay (Montmorillonite)	-	40 mg/g	(Guiza et al., 2021)
Activated Carbon (Leucaena leucocephala)	Nitric acid	1.16 mg/g	(Bifta et al., 2020)
Activated Carbon (magnetic)	Ferric chloride	12.6 mg/g	(Ibrahim et al., 2019)
Mentha Ash	Sodium & Aluminium	87%	(Bhan et al., 2021)
Activated carbon	Mg-Mn-Zr	26.27 mg/g	Mullick and Neogi (2019)
Activated carbon	Zirconium	94.4%	Mullick and Neogi (2018)
Activated carbon	Zirconium (drop-coating)	28.5 mg/g	(Pang et al., 2020)
Carbon nanotubes	Hydropatite composite	11.05 mg/g	(Tang et al., 2018)
Activated carbon	Cerium nitrate	4.6 mg/g	Inaniyan and Raychoudhury (2018)
Coconut husk	N_2 & KOH	6.5 mg/g	(Talat et al., 2018)
Cornhusk	HCl	5.8 mg/g	(Gebrewold et a., 2018)
Rice husk	NaOH	7.9 mg/g	
Spirogyra biomass	-	95%	(Sivarajsekar et al., 2017)
Activated carbon (*Morinda tinctoria*)	Aluminum hydroxide	26.03 mg/g	Amalraj and Pius (2017)
Bentonite	Al-La-Ce	9.87 mg/g	(Nagaraj et al., 2020)
Activated carbon (*Ficus recemosa* leaf)	Zirconium	2.12 mg/g	Bhan and Singh, (2022)

3.3 Biochar

A carbon-rich solid product called biochar (BC) is produced by pyrolyzing biomass in anaerobic or oxygen-restricted conditions. BC has lignocellulosic elements that may efficiently absorb fluoride (Yadav and Jagadevan 2020; Verma and Singh, 2022). Biochar has been used in the treatment of water pollutants and other fields because of its significant specific surface area, rich in porous structure, the abundance of surface functional groups and high mineral content (Tan et al., 2015).

3.3 Activated carbon

Activated carbon come under the category of amorphous carbonaceous content which has large interior surface areas and high porosity. Anthracite and bituminous coals are the main sources of activated carbon until recently, but they can be made from any carbonaceous material. This is the most popular adsorbent for the elimination of pollutants from contaminated water (Saleem et al., 2019). For the generation of activated carbon, any inexpensive substance with a rich content of carbon and a low inorganic concentration can be utilized as a precursor. Various types of low-cost materials have been used for the synthesis of activated carbon such as bamboo, coconut shell, peanut shell, tamarind wood, date stones, peach stones, pea peel, orange peel, pomegranate peel, etc. (Srivastava et al., 2021). Several materials have been prepared as adsorbents and tested for fluoride removal from the water, some adsorbents and their adsorption capacity are given in Table 1.

4. Conclusions and recommendation

This literature discussed the contamination of fluoride in the water bodies and its removal methods. The presence of a very high content of fluoride in drinking water is responsible for serious health issues in human beings. Removal techniques including the Nalgonda technique, membrane filtration, and reverse osmosis are common processes. The filtration techniques like ultra-filtration, nanofiltration, and reverse osmosis are based on membrane filtration, the used membranes are synthetic in nature and also very costly. Adsorption methods are known as cost-effective and easy processes for defluoridation of water. Costs associated with the preparation of adsorbents could be reduced by using natural materials. Making biosorbents from waste biomass by its carbonizing is also a cost-effective method, and contributed to manage waste biomass. The number of adsorbents has been produced and tested only for adsorption efficiency in an aqueous solution. But there is a need to test and apply these adsorbents for groundwater treatment. The adsorbents made of clay, zeolite and activated carbon originating from biomass, are easier to synthesize than chemically created composites. Fluoride-containing water can be purified by the adsorption process by using various types of adsorbents after confirmation of their leachability test.

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Author contributions

Chandra Bhan (Research scholar) has conceptualize, prepared a draft of the article and formal analysis. Jiwan Singh (Supervisor) has reviewed edited and validated manuscript.

Conflicts of Interest

There are no conflicts of interest declared by the authors.

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