Journal for Research in Applied Sciences and Biotechnology

ISSN: 2583-4053
Volume-2 Issue-5 || October 2023 || PP. 132-138

www.jrasb.com

https://doi.org/10.55544/jrasb.2.5.22

Fluoride Removal from Water Using Adsorption Method with Different Compounds: A Comprehensive Review

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www.jrasb.com || Vol. 2 No. 5 (2023): October Issue

Received: 04-11-2023	**Revised:** 08-11-2023	**Accepted:** 11-10-2023

ABSTRACT

This comprehensive review paper explores the diverse range of compounds employed in the adsorption process for the removal of fluoride from water. The escalating levels of fluoride contamination in drinking water sources pose a significant health threat to communities worldwide. Adsorption is a widely acknowledged and effective method for mitigating this concern, involving various compounds such as activated carbon, metal oxides, and biomaterials. It focuses on the mechanisms, adsorption isotherms, kinetics, and factors affecting the efficiency of fluoride removal using these compounds. We discuss the advantages and limitations of each compound, considering their applicability in different environmental conditions and scale of operation. Furthermore, we scrutinize the regeneration and cost-effectiveness of these materials. This review consolidates the existing knowledge on fluoride removal via adsorption techniques, offering valuable insights for researchers, policymakers, and practitioners involved in water purification. The main objective of this paper is to present a comprehensive, and up-to-date assessment of the subject matter.

Keywords- Adsorbent, Environment, Fluoride, Water.

I. INTRODUCTION

The Fluoride range and the cumulative quantity taken gradually, may be advantageous or harmful for human health [1-4]. The dietary factors, body weight, rate of bone formation, activity level, and remodeling all have a significant role in determining when others may react to fluoride exposure [5]. Fluoride may impair fertility and induce dental cavities, when consumed at low amounts. Fluoride promotes the development and maintenance of healthy bone and tooth enamel, when consumed within allowed limit set by WHO, however excessive use of fluoride may cause fluorosis [6-7]. According to the WHO, fluoride levels in drinking water should be between 0.5 and 1.5 mgL^{-1} [8-9]. By removing the hydroxide ions from hydroxyapatite, $Ca_5(PO_4)_3(OH)$, the major mineral component of bones and teeth, fluoride helps to preserve healthy bones and teeth. Fluorapatite, $Ca_5(PO_4)_3F$, is more resistant and durable than hydroxyapatite.

$$\mathbf{Ca_5(PO_4)_3OH + F^- \rightarrow Ca_5(PO_4)_3F + OH^-}$$

Fluorapatite has higher acid tolerance damage and less soluble than hydroxyapatite due to its hardness and toughness; as a result, it aids in the formation of tooth structure and shield it from acids from food residue in the mouth. This is useful, only when levels of fluoride are low. When its concentrations are high, significant quantities of hydroxyapatite are turned into fluorapatite, which might result in the process to extend the hydroxide replacement.

ISSN: 2583-4053
Volume-2 Issue-5 || October 2023 || PP. 132-138
https://doi.org/10.55544/jrasb.2.5.22

$$Ca_5(PO_4)_3F + 9F^- \rightarrow Ca_5F_{10} + 3(PO_4)^{3-}$$

Fluorosis is a condition that develops from this process which causes the bones and enamel to become deep, tough, and more brittle [10-12]. The fluoride exposure at high intake will affect the range and length of its exposure [13-14]. Fluoride levels over 1.5 mgL^{-1} may occur in skeletal fluorosis and dental fluorosis (4–10, 1.5–4 mgL^{-1} respectively). Fluorosis, which is defined by twisting of the stiff joints, bones, and difficulty in walk, occurs at fluoride higher level than 10 mgL^{-1} [15]. In addition to the negative health impacts of high fluoride intake, having too much fluoride in water sources has socioeconomic repercussions.

II. FLUORIDE PRESENCE IN WATER

India, Korea, China, Sri Lanka, Thailand, Yemen, Indonesia, Iraq, Pakistan, and Turkey are among the Asian nations having the highest levels of fluoride in both surface and groundwater. In India, about 66 million individuals are at risk of fluorosis.

Table 1. Fluoride concentration in India

Country	Location	Water Source	Fluoride Concentration (in mgL^{-1})	Reference
India	Andhra Pradesh	Ground water	.11–20.	[16]
	Bihar		.60–8.	[16]
	Delhi		.20–32.50	[17]
	Delhi		.20–32.50	[18]
	Gujarat		.10–40.	[17-18]
	Gujarat		1.58–31.00	[16]
	Haryana		.17- 24.70	[16]
	Hyderabad	Shallow and intermediate ground water	.38-4	[19]
	Jaipur	Ground water	4.50-28.10	[17]
	Jammu and Kashmir		.05-4.21	[16]
	Tamil Nadu	Shallow deep Ground water	1.–3.24	[20]
	Najafgarh	Ground water	.40–100	[16]
	M.P.		.08–4.20	[16]
	Maharashtra		0.11–10.20	[16]
	Kerala	Shallow, intermediate, and deep ground water	.20–5.75	[21]
	Punjab	Ground water	.44–6.	[16]
	Rajasthan		.20–37.	[16]
	Uttar Pradesh	Shallow– deep groundwater	.48-6.70	[18]
	Varanasi	Ground water	.20–2.10	[18]

III. IMPORTANCE OF FLUORIDE REMOVAL

Skeletal fluorosis is a disease, in which the fluoride accumulates in the bones, may be taken on by excessive fluoride ingestion over an extended period. Due to this, seniors may eventually have weak or cracked bones as well as stiffness and pain in the joints. An uncommon osteosarcoma cancer can also cause due to its higher presence in water.

IV. FLUORIDE REMOVAL BY ADSORPTION METHOD

Liming and the ensuing fluorite precipitation are the conventional methods for eliminating fluoride from drinking water [22]. Numerous research has focused on the coagulation processes and ppt involving alum sludge, (III) iron, activated alumina, and Ca [23-26]. Research has also been done on the removal of excessive amounts of F from drinking water utilizing reverse osmosis, ion alteration, and electro dialysis [27-33]. One method of defluoridating water is by the adsorption process. It is commonly used, to yields superior outcomes, and seems to be a more alluring strategy in terms of price, simple manufacture, and convenience [34-35]. The next sections of this research will discuss several common and uncommon adsorbents that tested for their removal.

- ***Calcium-based sorbents for water defluoridation***

Since Ca has a strong affinity for the fluoride anion, several studies have been conducted on its removal utilizing different calcium salts. Turner et al. [36]

ISSN: 2583-4053
Volume-2 Issue-5 || October 2023 || PP. 132-138

https://doi.org/10.55544/jrasb.2.5.22

observed the fluoride removal from high fluoride concentrating solutions ranging from around 3-2100 milligram per liter using batch studies and surface-sensitive methods on fractured limestone (99% pure calcite). The researchers were capable to show that a mixture of surface adsorption and ppt mechanisms oversaw the removal from aqueous systems, which was regulated by the surface area of the calcite. In addition to surface-sensitive techniques like AFM (atomic force microscopy) and X-ray photoelectron spectroscopy, potential measurements were also employed (AFM). The finding shows immediate fluoride adsorption throughout the whole calcite surface, with fluorite precipitating at step edges and bends where the concentration of dissolved Ca^{2+} was highest.

- ***Sorbents with an iron basis for defluoridating H_2O***

Due to iron's higher affinity for fluoride, based on iron compounds have also been thoroughly researched for fluoride elimination. Mostly removed fluoride adsorbents have been evaluated for drinking purpose; although, they were not constant at high pH levels without adjusting the pH. Therefore, refining industrial wastewaters with high amounts of fluoride is a serious issue. Schwert mannite was used to clean up wastewater that was tainted with fluoride because its magnetic qualities and resilience at low pH levels [37].

It has been shown that synthetic siderite is an effective sorbent for the adsorption of arsenic, used by Liu et al. [38] to remove fluoride. Batch studies with the synthesized siderite showed a considerable ability to adsorb F, reaching up to less than two milligrams, with an initial F concentration of 20 mgL^{-1} at 25 degrees Celsius and an adsorbent dose of 5 gL^{-1}. While $(PO_4)^{3-}$ considerably impacted the Fluoride removal from aqueous solution, Chlorine and Nitroxide had little impact on the adsorption of F.

- ***Adsorbents based on alumina and aluminium for defluoridating water***

For many years, scientists have been considering the efficacy of activated alumina as a fluoride adsorbent. Farrah et al. examined the ion interaction of fluoride with amorphous, alumina, or gibbsite across a wide pH and Fluorides concentration (3-8, 0.1–1.0 mM respectively). It was shown that the majority of the amorphous gel dissolves by the formation of AlF complexes at pH values lower than 6 and total Fluoride: Aluminium ratios higher than 2.5, with the F ions distribution being controlled by the equilibrium Fluoride value. Some solid remained in the pH four to seven range at lower F:Al ratios and significantly sorbed F from solution [39].

Moreover, Ku and Chiou investigated how several operational parameters affected alumina's ability to absorb fluoride from aqueous solutions. The range of 5-7 was found to be the ideal operating pH for the elimination of the greatest amount of fluoride (16.3 mg/g). The pH of the solution significantly affected how well fluoride was removed [40].

- ***Carbon based sorbents for defluoridating water***

A few scientists have also investigated the removal of fluoride using carbon-based sorbents. Bhargava and Killedar experimented with fluoride adsorption over fishbone charcoal in a moving media adsorption device [41]. Utilizing regenerated bone char medium that Kaseva had improved and employed in his studies, drinking water was defluoridated [42]. The maximum fluoride elimination was found to be created by bone char with the smallest grain size (0.5-1.0 mm), and Carbon for two hours was found to be the best regeneration temperature. The findings revealed the maximum fluoride removal and adsorption capacities were, respectively, 70.64% and 0.75 mgg-1. Janardhana et al. also studied in a continuous down flow adsorption mode at constant temperature used zirconium-impregnated activated charcoals to defluoridate drinking water [43].

- ***Industrial wastes used as sorbents in water defluoridation***

As a byproduct of widespread industrial operations, enormous amounts of solid waste are produced. Where one of the advantageous applications of these wastes is their conversion into low-cost sorbents for water and wastewater detoxification. Reducing F from aqua has been tested on several industrial wastes, both with and without treatment. Chaturvedi et al. conducted research on the capability of ash in air, a waste product from thermal power plants, to lower the range of fluoride from water. Fluoride elimination was shown to be beneficial at low concentrations, hot temperatures, and acidic pH levels. Fly ash has a maximal Langmuir sorption capacity for fluoride between 20.0 and 20.3 mg/g [44]. Nigussie et al. exploited the industrial waste leftover produced during the sulphuric acid process to produce aluminum sulphate (alum) from kaolin as the defluoridating medium [45].

- ***Hydrotalcite-like compounds, hydroxyapatite, and apatite were layered as sorbents for defluoridating water.***

The main advantage over other chemical treatment methods (such as precipitation) is that they don't create any chemical sludge and it resume their real complex structure after adsorbing distinct anions, LDHs have also been investigated for their capacity to remove fluoride from an aqueous solution included calcining Mg-Al-CO_3-LDH. Due to the preservation of its inherent structure, LDH that had been calcined at 500 °C had the best ability to remove fluoride, but LDH that had been calcined with a Mg/Al ratio of 2 had an exceptional capability to adsorb anions [46]. The effectiveness of fluoride removal utilizing hydroxyapatite, which was made using a modified chemical wet technique in a highly porous form, was observed by Hammari et al. [47]. To remove fluoride, the efficacy of creating crystalline and porous calcium hydroxyapatite (c-HAp) was assessed. In comparison to the fluoridation rate of p- HAp using a 1 mol/L fluoride solution was 89%. It was found that the

ISSN: 2583-4053
Volume-2 Issue-5 || October 2023 || PP. 132-138
https://doi.org/10.55544/jrasb.2.5.22

sample's higher specific surface area (235 m^2/g) compared to the sample's lower specific surface area (47 m^2/g) encouraged the removal of fluoride ions from aqua solution. The structural and conduction characteristics of porous hydroxyapatites were altered by fluoride adsorption, favoring the stable fluoridated apatite.

- ***Various adsorbents for water defluoridation***

For the adsorption-based removal of arsenate, phosphate, and fluoride ions, a silica gel with lanthanum was created [48]. At equilibrium pH 6, which was seen, the lanthanum ion and silica gel interacted most strongly. From an initial concentration of 0.55 $mmolL^{-1}$, fluoride on the lanthanum-impregnated silica gel was removed with >99.9% efficiency at pH neutral. The adsorption was unaffected by other anions such as Bromine, Chlorine, Iodine, Nitrogen Oxide, and Sulphur di-oxide. As prospective adsorbents for the removal of fluoride from H_2O, several accessible minerals, including seeds of charfines, kaolinite, lignite, and nirmali were also investigated [49].

Nemade et al. also carried out batch adsorption tests to evaluate the efficacy of removing fluoride from brick powder, fly ash, wood charcoal, fish bone charcoal, animal charcoal, etc. Compared to other adsorbents, the fish charcoal bone found a much greater removal of fluoride [50]. Sarkar et al. (2006) also investigated the efficacy of hydroxyapatite, fluorspar, calcite, quartz, and quartz activated by ferric ions as low-cost materials for eliminating fluoride from water [51].

- ***Other metal oxides/hydroxides/oxyhydroxides as sorbents for defluoridating water***

An inorganic cerium-based adsorbent for defluoridation was investigated. The fluoride sorption capacity of the produced CTA was notable. Low pH encouraged the reduction of fluoride, and the adsorption isotherm data matched the Freundlich isotherm. By using the sol-gel process, Xiuru et al. were able to create composite CeO_2-TiO_2/SiO_2 surfaces and assess their capacity to remove fluoride [52]. The surface composite showed a 21.4 mg/g fluoride adsorption capability. Zhu et al. investigated the possibility of silicon dioxide granules modified with magnesia removing fluoride [53]. To remove fluoride from wastewater, Gupta et al. created a micro-nano hierarchical web of carbon nanofibers and activated carbon fibers. When employing the Al-CNF to treat wastewater with a pH range of 5.0 to 8.0, it was discovered that the intake water did not need pre-treatment [54]. The sorbent created by coating granular activated carbon with manganese oxides using a redox technique was used to remove fluoride from aqueous solution [55].

V. BIOLOGICAL APPROACHES FOR FLUORIDE REMOVAL

For the treatment or elimination of environmental toxins, biological approaches require the use of living organisms, such as bacteria, plants, or their products. The concept of bioremediation is extensively used in a range of techniques to support the intensive metabolic activity of plants and microorganisms, which helps break down organic contaminants into more basic forms like carbon dioxide and water [56]. They can withstand various contaminants by developing techniques such as bioaccumulation, biotransformation, and biosorption, among others [57].

- ***Phytoremediation***

It alludes to a potential green technology that uses vegetation to purify polluted water, soil, and the air [58]. To deal with the pollutants, it requires a number of processes, including phytovolatilization, phytostabilization, phytostimulation, phytoextraction, and phytodegradation, of which phytostabilization and phytoextraction have previously been described for F remediation.

- ***Phytostabilization***

Plants with the capacity to live in polluted soil act as its mediators. By entangling in the soil matrix and immobilizing F directly, root exudates reduce its solubility. Additionally, via adsorption and precipitation, respectively, inside the root zone, the roots stop the movement of F caused by deflation and erosion [59].

Abdallah et al. (2006) investigated how well grapes were able to balance F buildup in their leaf margins with an equal Ca accumulation and proposed that F may be trapped as CaF_2, which would not interfere with plant metabolism [60]. The inability of F to go through the phloem and reach lower plant organs Plants that can tolerate fluoride may be able to deactivate it more effectively than those that are sensitive [61]. A few possible strategies include switching to metabolic pathways that are less susceptible to F, complexing F with organic chemicals to remove them from enzyme inhibition, reacting with sites of cationic, sequestering vacuoles in Fluoride and translocating it to the surfaces of leaves, etc. By exporting it to the surfaces of their leaves, some plants may reduce internal Fluoride [62].

- ***Phytoextraction (phytosequestration)***

Contaminants are absorbed, relocated, and accumulated in the aerial sections. In the course of extraction, pollutants are first taken up from the nearby environment and then transferred to different harvestable portions (shoots, leaves, etc.) via the roots [63]. This method often uses hyperaccumulators or plants that have a 0.1% or greater pollutant accumulation capacity on a dry mass basis [64-65]. For the bioconcentration factor (the ratio of contaminant concentration in plant roots to soil), the translocation factor (the ratio of contaminant concentration in plant shoots to roots), and the enrichment factor, hyperaccumulators are plant species with values higher than 1.

Nowadays, a number of plants with promising F accumulation potential are known. According to reports, several different plant species can extract F from the soil, but none of them have been used commercially for this purpose. Screening potential hyperaccumulators for

ISSN: 2583-4053
Volume-2 Issue-5 || October 2023 || PP. 132-138

https://doi.org/10.55544/jrasb.2.5.22

higher fluoride and resistivity is now important [66]. Cellular defenses in plants that can withstand, and resist fluoride reduce its harmful effects. Saini et al. (2012) evaluated the accumulation of Fluoride in P. juliflora by organ in relation to this. The species gathered the most F in roots, according to their data [67].

- ***Microbial remediation***

Microbial remediation refers to the process of rejuvenating soil after pollutants and toxins have been removed utilizing bacteria or fungus. The pollutants are used by these microorganisms, who subsequently break them down for fuel and reproduction. Three procedures are used for microbial cleanup:

✓ Natural attenuation that occurs spontaneously thanks to the local soil microorganisms.
✓ Biostimulation involves feeding the soil's microorganisms with nutrients, moisture, and a pH that is right.
✓ Using externally introduced microorganisms for bioaugmentation when naturally existing bacteria are rendered inert due to a high level of pollution.

VI. CONCLUSION

This review highlights the diverse array of adsorption methods employed for fluoride removal from water, utilizing various compounds. The research showcased herein demonstrates the efficacy and versatility of these techniques in mitigating fluoride contamination. The insights provided underscore the importance of continuing to explore and refine adsorption methods as to address the pressing issue of fluoride pollution in water sources.

The future holds significant promise in the realm of fluoride removal. Developing eco-friendly and cost-effective adsorption materials, optimizing existing methods, and exploring emerging nanotechnology-based approaches are key areas for advancement. Additionally, increased attention to scalability and field applications will be vital for widespread impact. Collaborative interdisciplinary research will play a crucial role in delivering practical, sustainable solutions to combat fluoride contamination in water.

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