

Chapter 7

New materials for arsenic and fluoride removal

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ABSTRACT

Arsenic and fluoride are two major global water pollutants. They are introduced into the environment via various natural and anthropogenic activities. Several countries in the world are facing the detrimental health and environmental effects of these contaminants. Many technologies have been proposed and implemented for the removal of arsenic and fluoride from water. However, there is an ever-increasing demand for affordable and reliable water purification approaches. All the technologies are primarily based on any one of the following purification techniques: membrane filtration, reverse osmosis, electrodialysis, adsorption, precipitation, coagulation, oxidation, and ion exchange. The contaminant removal efficiency depends on the properties of the material used. Various materials have been tested and are used to deliver arsenic and fluoride-free water. In developing countries, affordability plays an important role in the implementation of water treatment strategies. This chapter gives an overview of simple, efficient, and affordable materials that combined with appropriate technologies, provide promising scalable solutions for the mitigation of arsenic and fluoride in water.

Keywords: arsenic, fluoride, removal technologies, adsorbents, sustainability indices

7.1 INTRODUCTION

Water is vital for the sustenance of life on earth. Though groundwater comprises only 0.6% of the total water resources on earth, it is the major and favoured source of potable water in developing countries' rural and urban areas. In rural India, groundwater caters to 80% of drinking water and 50% of agricultural demand (Meenakshi & Maheshwari, 2006). Several geogenic and anthropogenic activities have contaminated aquatic ecosystems with metals, non-metals, and other

toxic compounds. Arsenic and fluoride are both naturally occurring elements that can be found in the Earth's crust. However, when present in high concentrations in water, they can cause harm to human health. The World Health Organisation (WHO) has categorized both as critical chemicals that can cause large-scale health effects as a result of drinking contaminated water ([World Health Organization, 2017](#)).

7.1.1 Arsenic and fluoride contamination

Groundwater contamination with arsenic and fluoride at hazardous levels has been reported in several countries including Bangladesh, China, India, Mexico, and Argentina ([Mukherjee *et al.*, 2019b](#)). Arsenic and fluoride contamination sources are natural and anthropogenic. Arsenic and fluoride come to be present in ground and surface waters primarily through the dissolution of natural arsenic and fluoride-containing minerals and ores present in rocks and soil ([Maity *et al.*, 2021](#)). Anthropogenic sources of fluorides are attributed to the production of phosphate fertilizers, bricks, tiles, and ceramics. Fluoride is essential in minute quantities for the normal mineralization of bones and the formation of dental enamel.

Arsenic is a Group VA metalloid. Various species of As are arsenic trioxide (AsO_3), arsenite As(III) , arsenate (As(V)), methylated species, arsenobetaine (AB), and arsenocholine (AC). It exists of four oxidation states: arsenite (As^{3+}), arsenate (As^{5+}), arsenic (As^0), and arsenide (As^{3-}). The mobility of arsenic depends on redox potential (Eh), pH, biological activity, and adsorption/desorption from aquifer rocks and sediments. In general, the mobility of As(III) is higher than As(V) in aquifers due to the lower probability of adsorption of neutral As(III) on mineral surfaces ([Hao *et al.*, 2018](#)). The anthropogenic sources of arsenic are mining, ore production and processing, electronic device fabrication, combustion of fossil fuels, synthesis and use of dyes and pigments, pharmaceuticals, and agricultural insecticides and pesticides ([Mukherjee *et al.*, 2019a](#)).

In India, the acceptable limit of arsenic in drinking water is 10 ppb, and in the absence of any alternate source, the limit is 50 ppb ([IS 10500, n.d.](#)). For fluoride, the permissible limit in India is 1 ppm, and in the absence of an alternate source, the limit is 1.5 ppm ([IS 10500, n.d.](#)). Chronic exposure to arsenic can cause various health problems, such as arsenicosis, reproductive disorders, peripheral neuropathy, and it also has a carcinogenic potential ([Mukherjee *et al.*, 2019a](#)). Fluoride can cause discolouration of the teeth (dental fluorosis), weakening of the bones (skeletal fluorosis), defects in knees, hips, bones, and in extreme cases, it may also cause paralysis ([Patel *et al.*, 2019](#)). Hence, restricting the presence of arsenic and fluoride within stipulated limits in water supply is essential.

7.1.2 The current scenario in India

Arsenic and fluoride contamination is a significant problem in certain regions of India. They are the two major contaminants of Indian groundwater in terms of affected areas and the number of people. The worst affected areas are the middle and lower Gangetic planes and some parts of Central and South India. Fluoride contamination, on the other hand, is more widespread, affecting several states. [Table 7.1](#) shows the state-wise list of the number of districts affected by fluoride and arsenic, as reported by the Central Ground Water Board (CGWB) of India ([CGWB, n.d.](#)).

The Indian government and various organizations have been implementing various measures to address the problem. The Jal Jeevan Mission (JJM) is one of the initiatives taken by the government under which community water purification plants (CWPPs) have been installed in arsenic and fluoride-affected regions. However, implementing these measures on a large scale remains a challenge, and more needs to be done to tackle the problem.

Table 7.1 Occurrence of high fluoride and arsenic in groundwater in some states of India.

S. No.	State	Districts Affected by Fluoride	Districts Affected by Arsenic
1	Andhra Pradesh	16	3
2	Assam	2	8
3	Bihar	5	21
4	Chattisgarh	2	1
5	Delhi	7	2
6	Daman & Diu	–	1
7	Gujarat	18	12
8	Haryana	11	8
9	Himachal Pradesh	–	1
10	Jammu & Kashmir	1	3
11	Jharkhand	4	1
12	Karnataka	14	2
13	Kerala	2	–
14	Maharashtra	8	–
15	Madhya Pradesh	13	8
16	Manipur	–	2
17	Odisha	18	1
18	Punjab	9	7
19	Rajasthan	32	1
20	Tamil Nadu	8	–
21	Telangana	–	1
22	Uttar Pradesh	7	13
23	West Bengal	7	8

7.2 MATERIALS FOR ARSENIC AND FLUORIDE REMOVAL

The selection of material(s) for removing fluoride and arsenic using any of the aforementioned purification techniques depends on the removal efficiency, affordability, scalability, and regeneration capacity.

7.2.1 Metal oxides and hydroxides

Metal oxides and hydroxides in the form of nanoparticles or composites are very effective for removing arsenic and fluoride from water due to their affinity. The majority of them are used as adsorbents. However, few of them can be used with other treatment methods.

Iron oxide/hydroxide/oxyhydroxide, such as ferric oxide (Fe_2O_3), ferric hydroxide ($\text{Fe}(\text{OH})_3$), akaganèite ($\beta\text{-FeOOH}$), and ferrihydrites ($\text{Fe}_{10}\text{O}_{14}(\text{OH})_2$) have been used for removing arsenic from water due to their high affinity for arsenic under neutral conditions. These compounds can effectively remove both inorganic and organic forms of the contaminant. Moreover, they are non-toxic, low-cost, and available in significant quantities. The adsorption of arsenic and the effectiveness of the adsorbents depend on arsenic species, pH, and potential. Aluminium oxide and hydroxide, such as

Table 7.2 Metal-oxides and hydroxides-based arsenic and fluoride removal.

Metal-oxide/Hydroxide/Oxyhydroxide	Uptake Capacity (mg/g)		Reference
	As(III)	As(V)	
Arsenic Removal			
Confined metastable 2-line ferrihydrite	100	100	Anil Kumar <i>et al.</i> (2017)
α -FeOOH nanoparticles	–	76	Ghosh <i>et al.</i> (2012)
Zr-doped β -FeOOH nanoparticles	120	60	Sun <i>et al.</i> (2013)
Magnetic γ -Fe ₂ O ₃ nanoparticles	74.83	105.25	Lin <i>et al.</i> (2012)
Fe/AlO(OH)	–	102–129	Muedi <i>et al.</i> (2021)
Mesoporous iron oxide	136.89	31.82	Bae <i>et al.</i> (2020)
Hydrous cerium oxide (HCO) nanoparticles	170	107	Li <i>et al.</i> (2012)
Iron–zirconium (Fe–Zr) binary oxide	120	46.1	Ren <i>et al.</i> (2011)
Iron hydroxide nanopetalines	91.74	217.76	Wang <i>et al.</i> (2022)
Fluoride Removal			
Fe–Al–Ce trimetal oxide		178	Wu <i>et al.</i> (2007)
Cellulose–Al–Fe nanocomposite (AlOOH and FeOOH in carboxymethyl cellulose matrix)		75.2	Egor <i>et al.</i> (2021)
Boron-doped biochar/Al ₂ O ₃		196.1	Jiang <i>et al.</i> (2019)
Inorganic polymeric coagulant made by etching of aluminium trihydrate (Al ₂ O ₃ ·3H ₂ O) with acid		87.68	Solanki <i>et al.</i> (2020)
Nanoporous biochar-supported magnesium oxide (MgO-BC)		83.05	Wan <i>et al.</i> (2019)

alumina (Al₂O₃) and aluminium hydroxide (Al(OH)₃), have been widely used for removing fluoride from water. These compounds can adsorb fluoride ions from water by forming surface complexes. Table 7.2 lists some metal-oxides/hydroxides/oxyhydroxides used for arsenic and fluoride removal.

7.2.2 Biopolymers and biominerals

Various biopolymers and biominerals have been explored to remove contaminants from water. Reports based on hydroxyapatite, cellulose, alginate, chitosan, and gelatin are described here. Hydroxyapatite (HA; Ca₅(PO₄)₃OH), a member of the calcium phosphate family, is a widely researched adsorbent for treating contaminated water and soil. The surface of HA has tunnels and active sites that aid in improving the adsorption. The adsorption on HA can occur through either ion pair formation (F⁻...OH₂⁺), ion exchange (F⁻ with OH⁻), or H-bonding (F⁻...H...O) in aqueous conditions. These mechanisms result in the formation of mixed fluoridated HA (Ca₅(PO₄)₃(OH,F)) or stable fluorapatite (Ca₅(PO₄)₃F) (Nagaraj *et al.*, 2018). Nayak *et al.* synthesized nanocrystalline HA from egg shells and its fluoride adsorption capacity was 22.3 mg/g (Nayak *et al.*, 2017). He *et al.* developed an HA nanowires-based membrane with a fluoride adsorption capacity of 40.65 mg/g at neutral pH (He *et al.*, 2016b). Nagaraj *et al.* developed a mineral (Al³⁺, La³⁺, and Ce³⁺)-substituted HA nanocomposite material by hydrothermal method with a fluoride removal capacity of 8.36 mg/g (Nagaraj *et al.*, 2018).

Sharma *et al.* used ZnO nanocrystal decorated regenerated micro fibrillated cellulose for As(V) removal with the capacity of 4421 mg/g at neutral pH (Sharma *et al.*, 2019). Sinha *et al.* developed a filter device made of three-dimensional (3D) macroporous alginate/akaganeite nanorod composite (MAAC) scaffolds for arsenic removal with the capacity of 70.5 mg/g for As(III) and 24.4 mg/g for As(V) (Sinha *et al.*, 2018). Guo and Chen fabricated bead cellulose loaded with iron oxyhydroxide for adsorption and removal of arsenic with an adsorption capacity for As (III) and As (V) of 99.6 and

33.2 mg/g at neutral pH. The composite can be regenerated using 2M NaOH (Guo & Chen, 2005). Mukherjee *et al.* reported functionalized microcellulose-reinforced 2-line ferrihydrite composite that showed exceptional As(III) and As(V) adsorption capacities of 143 and 83 mg/g, respectively (Mukherjee *et al.*, 2019c).

Researchers are also investigating biopolymeric carrier mediums along with sorbent material to generate practical filter materials. Scientists have developed biopolymeric beads made of hydroxyapatite-implanted cerium-based metal organic frameworks incorporating alginate and chitosan for fluoride removal. These beads displayed an adsorption capacity of 4.8 mg/g in 20 min (Jeyaseelan & Viswanathan, 2022). Fernando *et al.* investigated a biopolymer-based nanohydroxyapatite (n-HAp) material for the removal of fluoride, arsenic, cadmium, and lead from water. They explored the composites of HA with chitosan, carboxymethyl cellulose, sodium alginate, and gelatin. Their study revealed the HA–chitosan composite as the most versatile sorbent (Fernando *et al.*, 2021).

7.2.3 Biological origin

Recently, researchers have been exploring the application of natural materials to remove various pollutants due to their efficiency, recyclability, and environmental friendliness. Calcium-containing adsorbents are a good choice for fluoride adsorption due to their favourable characteristics, including high ionic exchangeability, availability, adsorption affinity, and capacity to form bonds with various organics of varied sizes (Hashemkhani *et al.*, 2022). Scientists have used oyster shells (Hashemkhani *et al.*, 2022; Kim *et al.*, 2020b) and egg shells (Lee *et al.*, 2021) as a sorbent for fluoride removal from water. Hashemkhani *et al.* (2022) obtained the maximum adsorption capacity of 7.53 mg/g using oyster shell at pH 5.5 and contact time of 85 min (Hashemkhani *et al.*, 2022). Lee *et al.* reported maximum adsorption capacity of 258.28 mg/g using thermally treated egg shells at pH 7 and 25°C (Lee *et al.*, 2021). Fluoride adsorption reduced in the presence of anions in the following order: $\text{HPO}_4^{3-} > \text{HCO}_3^- \gg \text{SO}_4^{2-} > \text{Cl}^-$. According to them, fluoride removal is attributed to the formation of calcium fluorite (CaF_2). Sinha *et al.* (2003) used *Eichhornia crassipes* (water hyacinth) and its activated carbon for defluoridation. H type activated carbon (activated at 600°C) showed better performance than the non-carbonized plant (Sinha *et al.*, 2003). Fox *et al.* studied the removal of arsenic by flocculation–coagulation system using cactus mucilage and ferric (Fe(III)) salt. The system was capable of removing 75–96% arsenic in 30 min, with the majority of the removal occurring in 10–15 min (Fox *et al.*, 2016).

7.2.4 Carbon based materials

Carbon-based materials have been widely used to efficiently remove environmental contaminants owing to their high surface area, surface functional groups, and large number of active sites. Moreover, they can be easily coupled with metal oxides, nanoparticles, and so on, to enhance their adsorption capacities. Some carbon-based composites, such as activated carbon, carbon nanotubes (CNTs), carbon nanowires, graphite, graphene oxide, and carbon nanofibres, along with their uptake capacity for fluoride and/or arsenic, are listed in Table 7.3.

7.2.5 Biochar

One of the relatively new approaches for the decontamination of water is the use of biochar. Biochars are sustainable and renewable carbonaceous compounds synthesized by pre-/post-pyrolysis of biomass derived from feedstock under limited oxygen supply. The micropores of biochar offer high surface area along with several functional groups that are beneficial for the remediation of contaminants. Kumar *et al.* (2019) reported sustainable fluoride remediation using biochars produced by slow pyrolysis of okra stem and black gram straw with sorption capacities of 20 and 16 mg/g. Biochar-based adsorbents for arsenic removal from water have also been researched extensively. Strategies for enhancing the adsorption of arsenic include modifying biochar with metal salts, like FeCl_3 , MnCl_4 , FeSO_4 , and AlCl_3 , and acids (H_2SO_4 , HCl , and HNO_3) (Sharma *et al.*, 2022).

Table 7.3 Carbon-based materials and their uptake capacities.

S. No.	Carbon Material	Contaminant	Uptake Capacity (mg/g)	Reference
1	Aluminium-impregnated hierarchal web of micro-nano-activated carbon fibres	F ⁻	17	Gupta <i>et al.</i> (2009)
2	La/Mg/Si-activated carbon	F ⁻	220	Kim <i>et al.</i> (2020a)
3	Activated carbon–aluminium oxide composite	F ⁻	14	Iwar <i>et al.</i> (2022)
4	ZrO ₂ embedded in carbon nanowires	As(III) As(V)	28.61 106.57	Luo <i>et al.</i> (2016)
5	Mesoporous graphene oxide-lanthanum fluoride nanocomposite	As(V)	18.52	Lingamdinne <i>et al.</i> (2019)
6	Nano-alumina wrapped carbon microspheres	As(V)	68	Raj <i>et al.</i> (2023)

7.2.6 Metal organic frameworks

The development of metal organic frameworks (MOF), a new class of three-dimensional crystalline inorganic–organic porous hybrid materials with tuneable porosity and exceptional surface area, has made them promising candidates for a variety of applications, including gas storage, catalysis, sensor, drug delivery, ion exchange, and adsorption and removal of hazardous materials. Fe₂Co₁MOF-74 exhibited exceptionally high As(III) and As(V) removal capacities of 266.5 and 292.2 mg/g,

Table 7.4 Non-conventional technologies for arsenic and fluoride removal from water.

S. No.	Material	Removal Technology	Contaminant	Removal Efficiency	Reference
1	Aluminium form of phosphomethylated resin	Electrodeionization	F ⁻	73.2%	Gahlot <i>et al.</i> (2015)
2	Imidazolium-based ionic liquids grafted on thin-film composite forward osmosis membranes	Forward osmosis	As(V)	99.5%	Yang <i>et al.</i> (2019)
3	Hypochlorite oxidation, adsorption on Fe(II) coagulates, and low pressure membrane	Oxidation, adsorption, low-pressure membrane	As(III) and As(V)	>95%	Elcik <i>et al.</i> (2013)
4	Fe anode and air cathode with in situ generation of H ₂ O ₂	Air cathode-assisted iron electrocoagulation (ACAIE)	As(III) and As(V)	~100%	Bandaru <i>et al.</i> (2020)
5	Polyhedral oligomeric silsesquioxane-functionalized graphene oxide/ polyvinylidene difluoride (POSS-rGO/ PVDF) electrospun membranes	Membrane distillation	As(III) and As(V)	99.9%	Leeper <i>et al.</i> (2021)

respectively (Sun *et al.*, 2019). UiO-66 has also shown good arsenic adsorption capacities (Singh *et al.*, 2022). Aluminium fumarate (AlFu) MOF has been reported as a superabsorbent for fluoride with an adsorption capacity of 600 mg/g at 293 K (Karmakar *et al.*, 2016). However, there are not many industrial applications of MOFs for the removal of heavy metals. To address this, the membrane integration of MOFs is being explored by researchers. An adsorption membrane made of zirconium metal-organic frameworks (Zr-MOFs) was prepared for rapid fluoride removal from drinking water. The maximum adsorption capacity was 102.40 mg/g at pH 7.0 (He *et al.*, 2016a). Wang *et al.* reported an iron mesh-based MOF (MIL-100(Fe)) filter with capacity for As(III) and As(V) of 35.2 and 19.2 mg/g, respectively (Wang *et al.*, 2018).

7.2.7 Other technologies

Apart from the conventional methods discussed in this chapter, researchers are exploring several other technologies for the removal of arsenic and fluoride. A few of these methods are listed in Table 7.4.

7.3 EVALUATING SUSTAINABILITY INDICES OF TECHNIQUES

Several factors need to be considered to evaluate the sustainability of any fluoride and arsenic removal technique:

- **Efficiency:** The technique should be strongly able to effectively remove fluoride and arsenic from water.
- **Energy:** The amount of energy needed to operate, including the cost and availability of energy sources, should be economical. The carbon footprint of the process can be reduced by using renewable energy sources, such as solar, wind, and hydropower. Moreover, the amount of contaminant removed per unit of energy consumed must be as high as possible.
- **Cost:** The total expenses of the technique, including initial investment, operation, maintenance, and waste disposal cost, must be considered.
- **Material:** The materials used in water treatment must be affordable, sustainable, and effective in contaminant removal. Its environmental and health impacts must be thoroughly evaluated. The local and global availability of the material also plays an important role in determining the overall cost of the removal method. Moreover, the ability of the material to be regenerated or recycled and its potential for environmental degradation must also be considered.
- **Scalability:** The ability of the technique to operate at the household as well as community level.
- **Environmental impact:** This considers the generation of waste and its method of disposal, emissions during the operation, and the use of natural resources. The life cycle approach can be implemented to evaluate the energy and environmental sustainability of the water treatment method. Figure 7.1 shows the variables for environmental sustainability assessment (ESA) of adsorption and ion-exchange treatment methods. The life cycle inventory (LCI) can be estimated on the basis of the amount of final resources and pollutants for calculating the natural resources sustainability (NRS) and environmental burdens sustainability (EBS), respectively.
- **Social impact:** The technique's effects on the cultural and socio-economic aspects should be considered.
- **Reject/residue management:** Fluoride and arsenic removal plants can generate waste in the form of backwash wastewater, sludge, used adsorbents, or concentrated reject water. These need to be treated and/or stabilized and safely disposed of to prevent contamination of land or water. Various disposal options include landfills, stabilization to reduce toxicity and mobility, mixing with cow dung to promote microbial methylation of arsenic, passive aeration systems, and soil.

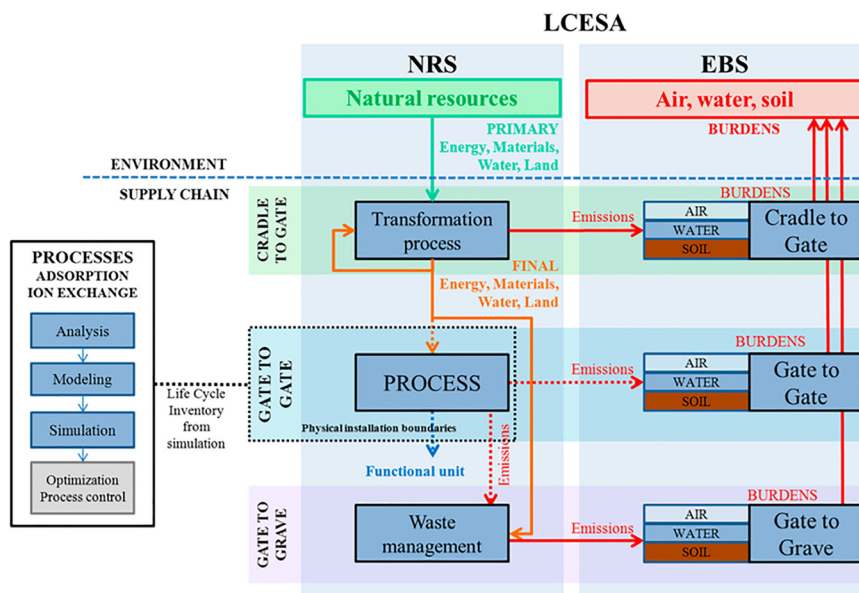


Figure 7.1 Block diagram of the life cycle environmental sustainability assessment (LCESA) for typical adsorption and ion-exchange methods. NRS: natural resources sustainability, EBS: environmental burdens sustainability (Reprinted with permission from Dominguez-Ramos et al. 2014. Copyright 2014 American Chemical Society). (Reprinted with permission from Dominguez-Ramos et al., 2014. Copyright 2014 American Chemical Society).

7.4 CONCLUSION

Arsenic and fluoride-contaminated water have become a global health issue. According to statistics by CGWB, the groundwater of 20 and 19 Indian states is affected by arsenic and fluoride, respectively. It is essential to determine the best management practices to control these contaminants to adhere to water quality standards. This chapter discusses various water treatment techniques and materials for addressing the problem of high concentrations of these toxic contaminants in water. Among the various techniques discussed, adsorption and ion-exchange-based technologies are promising and economical and have found abundant field applications. Extensive research over several decades has resulted in the development of adsorbents with improved removal efficiencies and adsorption capacities. However, all treatment methods produce certain waste materials that must be managed carefully and disposed of properly.

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