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REVIEW ARTICLE

Defluorination Techniques: Past, Present and Future Prospective

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Abstract

Fluoride ions are commonly found in minerals like mica, fluorapatite, muscovite, topaz, biotite, sellaite, cryolite, muscovite, and fluorspar. However, the presence of fluoride in groundwater has become a concern due to its contamination by wastewater from coal thermal power stations, aluminium smelters, electroplating-based industries, and glass and ceramic manufacturing facilities. While low fluoride consumption has some health benefits, excessive intake can lead to serious health issues such as crippling skeletal fluorosis, Alzheimer's syndrome, carcinogenic effects, infertility, and thyroid disorders. To address these chronic health impacts, there has been significant research to find out sustainable and highly efficient methods for fluoride removal. This review paper overviews various defluorination techniques, such as precipitation and coagulation, ion exchange, electrodialysis or reverse osmosis (RO), nano-filtration, adsorption, with their various advantages and drawbacks. The present review aims to provide insight knowledge of importance of fluoride, its toxicity issues and their available removal strategies and their limitations. This will help the researchers in developing cost-effective, environmentally friendly, and convenient techniques for defluorination in different matrices. The paper encourages scientists to work towards achieving reliable and eco-friendly defluorination methods for future application.

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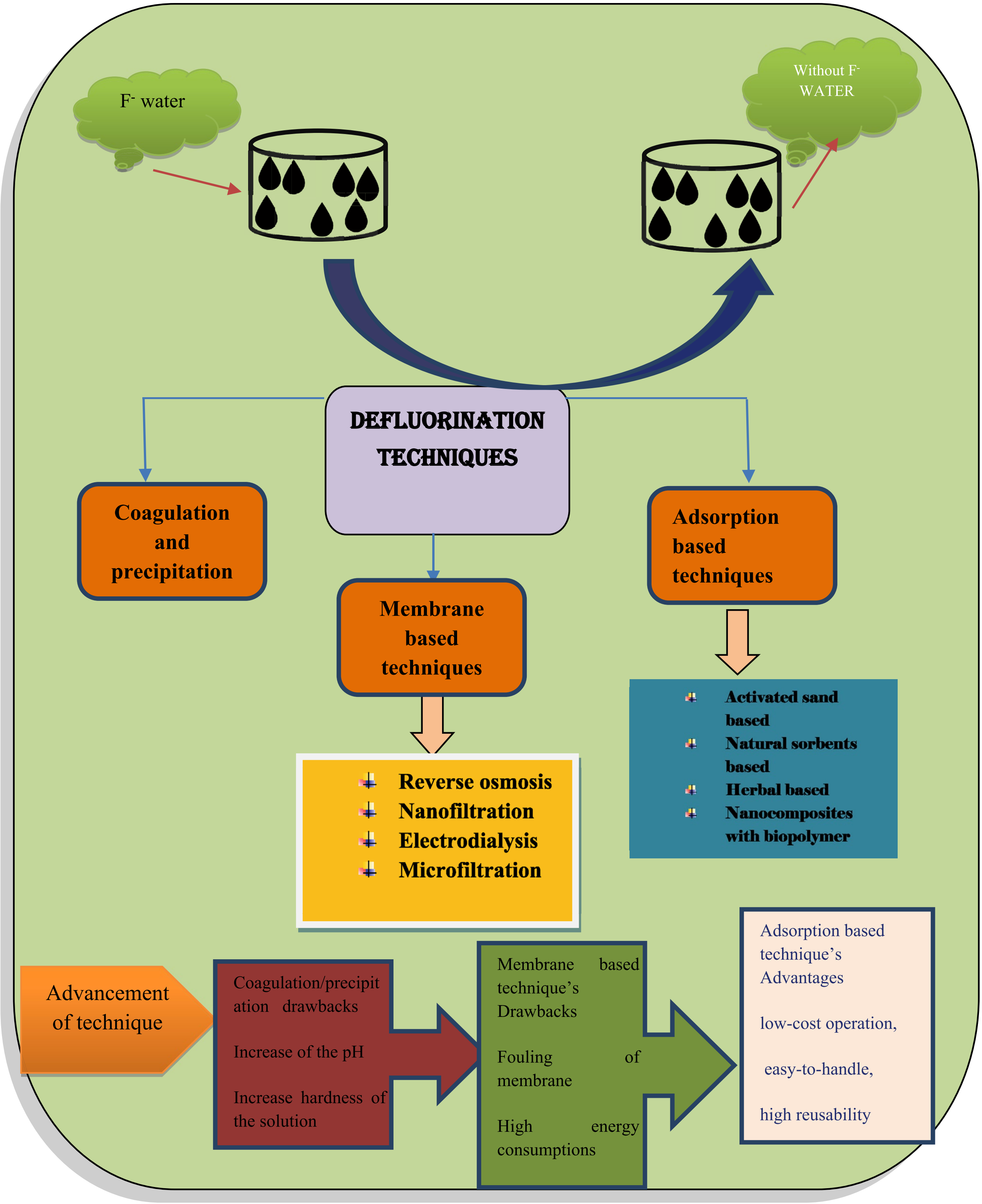
Graphical Abstract



Keywords Fluoride · Defluorination · Membrane based · Adsorption · Future

Highlights

- Illustrate the fluoride and their adverse health issues w.r.t. fluoride concentration.
- Various techniques such as precipitation and coagulation, membrane-based techniques and adsorption-based techniques with their advantage and disadvantage are analyzed in details.
- This review serves as a valuable resource for scientists seeking to develop more effective adsorbents, explore chemical-free regeneration of composites to reduce costs, integrate with advanced monitoring and control devices.

Introduction

Fluoride ions are univalent and negatively charged species that exist in many minerals, such as mica, fluorapatite, muscovite, topaz, biotite, sellaite cryolite, muscovite, and fluorspar. These fluoride minerals showed insolubility in water, but fluoride increased in groundwater by leaching fluoride in conditions that favour their solution [1]. Massive revolutions in the industrial sector and wastewater released from coal thermal power stations, aluminium smelters, electroplating-based industries, and glass and ceramic manufacturing industries contaminate and increase the fluoride concentration [2]. Consumption of fluoride in small quantities showed beneficial effects for the calcification of bones and dental cavities [3]. However, exposure to high fluoride showed high vulnerability to health issues outlined in Table 1 [4, 5].

People from tropical countries such as Ethiopia, Kenya, Tanzania, Jordon, Northern Thailand, Egypt, Libya, Algeria, Malawi, Mexico, Pakistan, Brazil, Afghanistan, Sri Lanka, Korea, Syria, South Africa, Indonesia, and India showed high fluorosis cases because of hot and humid climate conditions of environment leads to increased intake of fluoridated water [6, 7]. Excessive fluoride consumption also leads to many non-skeletal diseases, i.e. Alzheimer's syndrome, arthritis, brittle bones, cancer, infertility, and osteoporosis in humans [8]. Some studies also revealed that fluoride can mediate the metabolic activities related to carbohydrates, lipids, proteins, vitamins, and minerals and showed a disturbing effect on the synthesis of DNA [9, 10]. Fluoride can also disturb the activities of the number of enzymes that are used in various processes such as oxidative phosphorylation, glycolysis, coagulation, and neurotransmission [10]. A report submitted by CGWB revealed that 95 million people and 6.5 million children suffer. It also highlights that 223 districts in 23 states showed a high impact of severe fluoride contamination [11]. Because of high contamination impacts, the Bureau of Indian Standards specified a standardized limit of 1.5 mg/l in drinking water.

Table 1 Tabular representation of health effects in the presence of various fluoride concentrations

Sr. no.	Fluoride concentration (mg/l)	Health effects
1	Less than 0.5	Retardation of dental cavities
2	0.5–1.5	The beneficial effect for the calcification of bones and dental cavity
3	1.5–4	Dental issues in humans
4	More than 4	Dental fluorosis and skeleton fluorosis
5	More than 10	Crippling skeletal fluorosis, carcinogenic effects

Global scenarios related to fluoride contamination become a big dilemma for researchers that way numerous types of sustainable techniques for the detection [12] as well as removal such as precipitation or coagulation, ion exchange, electro-dialysis or RO, nano-filtration (NF), and adsorption used for the fix-up of fluoride concentration. This paper presents an overview of these sustainable techniques and various adsorbents used to defluorinate water.

Coagulation and Precipitation

The precipitation technique is when the soluble substance precipitates after attaining the supersaturation condition. This technique based on two principles, i.e., charge neutralization and sweep flocculation, which largely depend on the solution's coagulant dosage and pH. A way defluorination using precipitation technique or coagulants reported by many researchers as a convenient method based on the process of direct precipitation by reacting with suitable chemical reagent and co-precipitation in which removal or complex with a macro component from that solution by adsorption, occlusion, or mechanical entrapment as shown in Fig. 1 and this is further categorized into three types:

Direct Precipitation

The cheapest method for the elimination of fluoride was direct precipitation reported by the addition of lime ($Ca(OH)_2$) or more salts of calcium ($CaSO_4$, $CaCl_2$), which resulted in the formation of insoluble CaF_2 which is easily filtered from that solution. A number of researchers used this method for defluorination as the first choice, but their drawbacks such as an increase of the pH of the resultant solution and useful for only high fluoride-contaminated water because after treatment, the resultant solution had a fluoride concentration of about 10–20 mg/l above the permissible limit, which showed the poor settling characteristic of

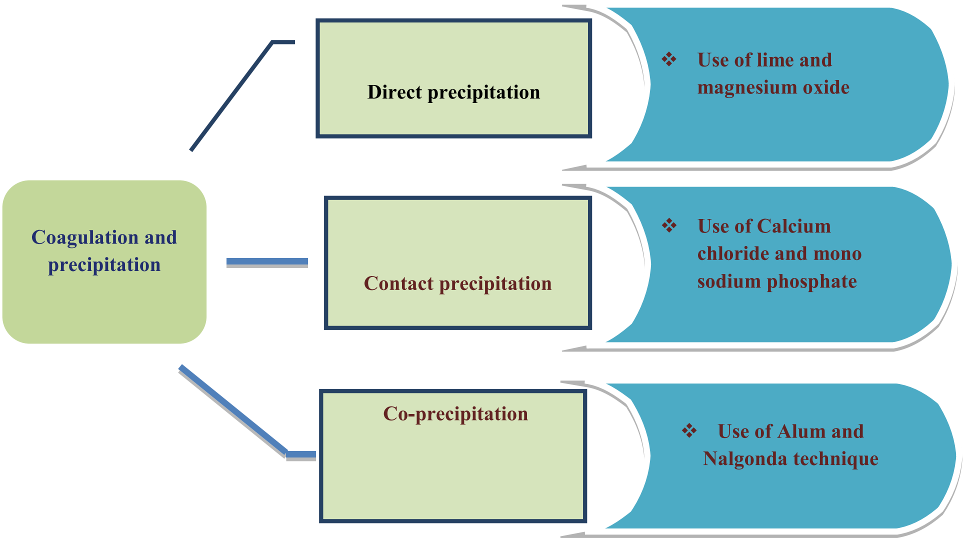


Fig. 1 Graphical representation of various types of precipitation techniques

precipitate, high ionic strength or hardness of treated water [13–18]. Instead of lime, magnesium oxide (MgO_2) was also reported for defluorination from high fluoride-contaminated wastewater [19, 20].

Contact Precipitation

In the case of contact precipitation, calcium chloride and monosodium phosphate were used to treat fluoride, which is based on the fixed bed and column, leading to the removal of fluoride but not the level of the permissible limit. Mekonen et al. [21] reported the precipitation using alum ($Al(OH)_2$) for water contaminated with fluoride of more than 10 mg/l. Still, it showed major drawbacks that give rise to the sulfate and aluminium amount in treated water to undesirable and control over pH in alum coagulation.

Co-precipitation

Co-precipitation technique is a technique in which simultaneous precipitation of the analyte along with the easily precipitated substances. Bulusu, [22] reported a Nalgonda technique that involved the addition of aluminium salt, i.e. aluminium sulfate and aluminium chloride, lime, and bleaching powder, followed by immediate mixing, flocculation, sedimentation, and filtration of flocs. In this technique, lime was 1/20th of the dose of aluminium salt, and 3 mg/l of bleaching powder was used to disinfect treated water. Nath and Dutta [23] studied using crushed limestone in which fluoride solution was acidified using one citric acid or acetic acid can decrease the contamination amount from 10 to 1.5 mg/l. Reardon and Wang [24] reported a two-column based reactor using limestone having removal efficiency and marked that the initial fluoride concentration of 109 mg/l was brought up to low concentration, i.e. 4 mg/l in this technique, the first column showed the precipitation of fluoride and resultant calcite was precipitated and filtered into the second type of column. Coagulation and precipitation techniques are marked as highly efficient and straightforward to operate for fluoride removal. Still, they caused the production of toxic sludge, change in the pH of water, which is not permissible by standards, and low removal efficiency caused unsuitable for field application. To overcome the drawbacks associated with this technique, various researchers work on another alternative method giving better fluoride removal efficiency, such as membrane-based techniques.

Membrane-Based Techniques

A membrane is a junction that constitutes a selective barrier that controls the transport of solutes between two adjacent phases. The membrane technique showed several advantages, such as transport selectivity, facile nature, easy to integrate with other technologies, and upscaling and downscaling of the membrane [25]. Membrane techniques are classified based on cross section, shape, membrane barrier, and transmembrane gradient or pressure, as shown in Fig. 2 [26]. In the case of passive transport, the driving force is the difference in chemical potential that is raised due to concentration difference, pressure gradient and an electric field. The non-porous membrane is a dense film where permeate transport is dependent on the diffusion model (pressure, electric potential gradient, and concentration), and solid polymeric material characteristics affect its properties, i.e. permeability and selectivity. But in the case of the porous membrane, permeate transport is based on pore size or surface characteristics of the membrane, so separation or permeability depends on the particle size. Instead of the huge application and advantages of the membrane technique, it showed some

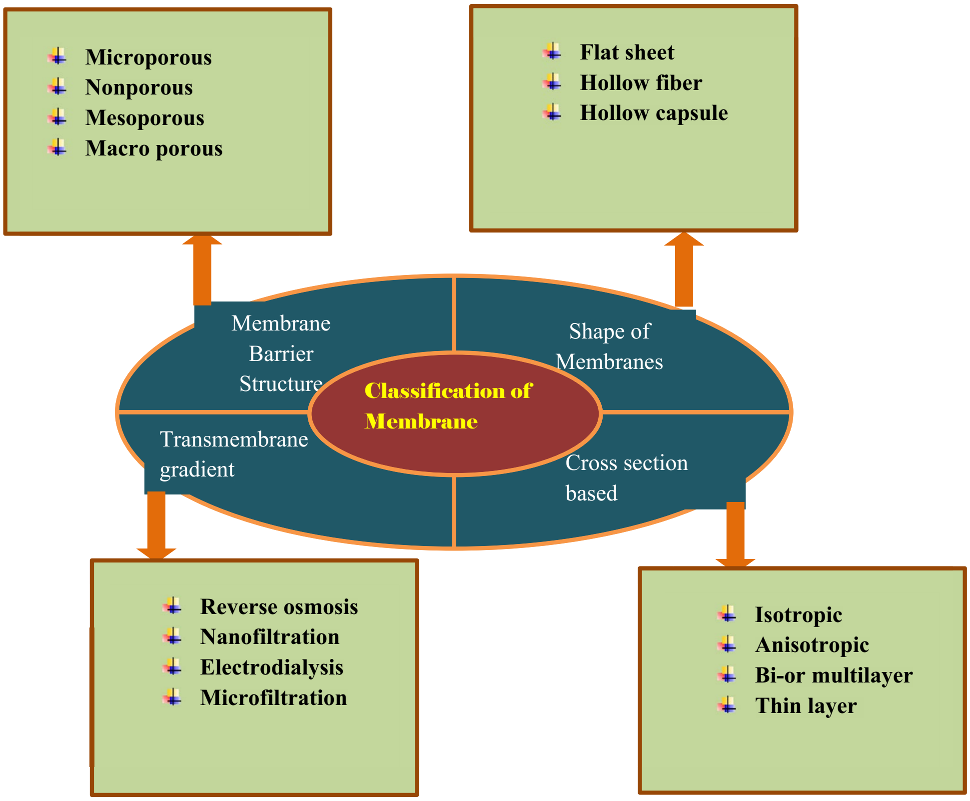


Fig. 2 Depicts a graphical illustration categorizing membranes based on their cross section, transmembrane gradient, shapes, and the nature of their barrier

drawbacks which decreased its efficiency, such as concentration polarization (increased amount of discarded materials on the membrane surface) and membrane fouling (adsorption of undesired colloidal on the membrane surface).

Reverse Osmosis-Based Removal

Reverse osmosis (RO) is a water purification system that shows activities of removal of unwanted or large solute particles under a pressure gradient-driven process through a semi-permeable membrane. Semi-permeable membrane showed the passage of solvent molecules, not solute molecules, through it. The operating pressure for transmembrane passage was studied between 20 and 100 bar for RO modules [27]. Various types of membranes were used for RO modules to purify fluoride, as mentioned in Table 2. These membranes have porous support materials, providing mechanical strength for operation. The industrial application of RO for purification showed the advantages of reusing concentrated stream or processed water and modest energy consumption for the procedure. Shen and Schäfer [28] reported a nano-filtration (NF)/reverse osmosis-based system using six types of membrane BW30, BW 30-LE, NF 90, NF 270, TFC-SR2, TRC-SR3 under the optimized conditions (pressure-10 bar, pH-8.29) for the removal of fluoride and natural organic matter forms the natural Water of Tanzania. The study concluded the removal efficiency of 67–99% with a recovery of 50%. Arora et al. [29] studied the fluoride removal efficiency of thin film polyamide membranes having spiral configuration by taking samples from nearby villages of Gurugram districts. Under the optimized conditions (temperature—30 °C, pH 7.0), the removal efficiency was 82–92.6%. Cohen and Conrad [30] also reported a membrane-based system using NF 100 cellulose acetate and NF 300 TFC membrane to defluorinate Lakeland, California's highly fluoridated natural water (USA). However, this method showed some drawbacks, such as high energy consumption for maintaining pressure gradient for operation, the polarization of the membrane, fouling of membrane (biological and minerals fouling), and post-treatment of water using limestone filters to reduce the water's alkalinity. A significant disadvantage is that the removal efficiency of RO is about 100%, which is why it eliminates all the fluoride, which creates an unpleasant smell and unbalances the minerals' abundance. Kettunen and Keskitalo [31] reported the fluoride and aluminium removal technique from water using a combined module or system of RO and limestone filtration.

Nano-filtration-Based Removal

Nano-filtration (NF) is a multifunctional treatment process with properties between the ultrafiltration and reverse osmosis processes. The developing era of wastewater treatment using the membrane in 1970–1980 considered it a loose RO

Table 2 Provides a summary of various defluorination methods associated with nano-filtration and reverse osmosis technologies

Sr. no.	Techniques of defluorination	Membrane types	Operative pressure	Source of analysis	Initial fluoride concentration	Removal efficiency	References
1	NF/sorption system	Film tec DOW NF membranes (NF270 and NF90)	1.2 MPa	Ground water	5 mg/dm^3/10 mg/dm^3	67% (NF 90) and 92% (NF 270)	[32]
2	NF/RO system	90 NE 8040 CSM	10 Bar	Ground water	3–0.7 mg/l	–	[33]
3	NF/RO system	70 NE 8040 CSM	10 Bar	Ground water	12.33–2.8 mg/l	–	[33]
4	NF/membrane distillation	Polyester NF membrane	9 Bar	Synthetic water	15.0–1.7 mg/l	89%	[34]
5	NF/RO system	BW30-LE membrane	10 Bar	Natural water	21–0.3 mg/l	89%	[35]
6	NF/RO system	DOW chemical's BW30 and NF270 membranes	9.8 Bar	Synthetic water	50 mg/l	100% and 80%	[36]
7	NF/RO system	DOW chemical, USA NF270/DOW chemical, USA BW30	6.0 Bar	Natural water	56.2 mg/l	51%/99%	[37]
8	NF system	NF membrane (NF90)	3–15 Bar	Synthetic ground water	56.2 mg/l	88%	[38]
9	RO system	Polyamide RO membrane	16 Bar	Synthetic solution	400 mg/l	95%	[39]
10	RO system	XLE RO membranes	6–11 Bar	Surface water mixed with ground water	1.8–0.03 mg/l	98.4%	[40]
11	NF/RO system	TFC BW30 and NF270	15.5 and 4.8 Bar	Natural water	59.7 mg/l	99% and 70%	[41]
12	Hybrid crystallization RO technique	F90 and SW30HR-380 membranes	30 Bar	Industrial waste water	6600 mg/l	95% and 99%	[42]
13	RO system	RO spiral-wound TFC	70 psi	Synthetic solution	10 mg/l	89.81%	[43]
14	NF/RO system	Flat-sheet TFC BW30 and NF270	9.8 Bar	Natural surface water	50 mg/l	95% and 78%	[44]
15	NF/RO system	Flat-sheet TFC BW30 and NF270	9.8 Bar	Natural surface water	50 mg/l	98.9% and 89%	[44]
16	NF/RO system	TFC BW30 and NF270	9.8 bar	Natural surface water	50 mg/l	98.8% and 91%	[44]
17	RO system	Polyamide RO membrane	2.45 MPa	Ground water	1.7 mg/l	60%	[45]
18	NF/RO system	BW30, NF90, and TFC-S	5 Bar	Single contaminant solution	3 mg/l	87%	[46]
19	NF/RO system	UTC-80A, ESPA4, UTC-60, and NF90	5 Bar	Mixed contaminant solution	3 mg/l	80%	[46]
20	RO system	Polyamide RO membrane	30 Bar	Electronic industry waste water	460 mg/l	98%	[47]

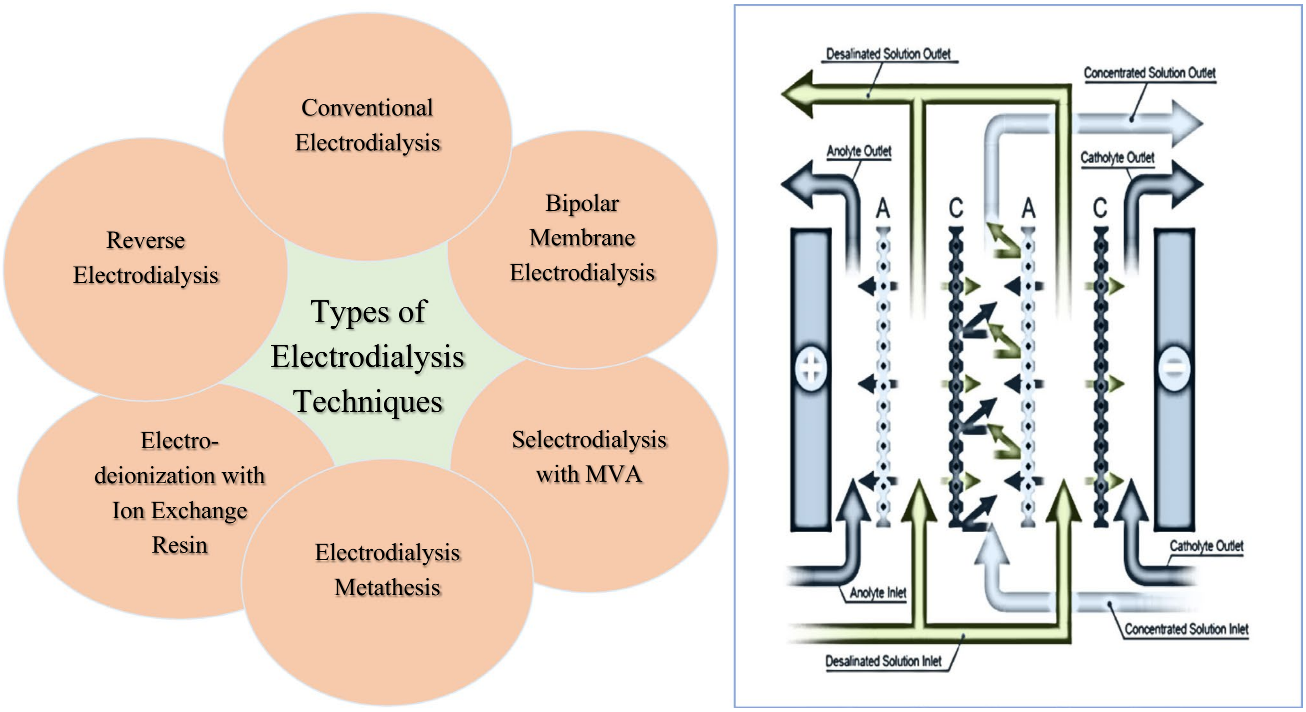
membrane based on its advantages and applications of nano-filtration membranes. Nano-filtration modules having pressure differentials or gradients less than RO modules showed a vast application of nano-filtration in food engineering to Wastewater treatment, the pharmaceutical sector and biotechnology field, and Environmental engineering. The nano-filtration technique showed high flux, high retention of multivalent anion salts, especially ion rejection of about

90–99% for multivalent or 10–90% for monovalent ions, high permeability, high retention of charged particles, low energy consumption for pressure gradient and high susceptibility against membrane fouling against the RO techniques.

The nano-filtration membranes are synthesized in numerous forms, such as spiral wound form, tubular form, capillary, plate and frame form, and hollow fibre formats, from an extensive range of materials, including cellulose derivatives and synthetic polymers, from inorganic materials such as ceramics and from organic/inorganic hybrids materials. NF membranes tend to have the property of a charged surface, with a negative charge at the neutral pH of the solution. This surface charge is essential to the membrane's transportation mechanism and separation properties. The properties like high selectivity or high rejection for ions of nano-filtration and electrodialysis methods showed superior methods for treating fluoride from the water. Simons [48] studied the removal of fluoride and boron using the composite of nano-filtration and diffusion analysis from the ash feed dam water using NF membrane, i.e. NTR 410, Filmtec NF 40, UTC 20 HF, and UTC-60. It was concluded that the membrane's high fluoride efficiency was about 70–80% in the pH range of 3–4.5. Tahaikt et al. [49] reported the treatment of contaminated water with high fluoride contamination using nano-filtration modules in Morocco. Nano-filtration membranes (NF 400 and NF 90) of spiral nature with different porosity 90 and 400 Da were assessed for fluoride removal. It concluded that the NF 400 membrane showed single pass modules and high efficiency for lower initial fluoride concentrated water. Still, in the case of high fluoride concentration, double pass membrane modules were acceptable. Tahaikt et al. [50] reported that a nano-filtration module using three commercialized membranes (NF 270, NF90, TR60) showed a single pass for the lower fluoride concentration of less than 6 ppm but a double-pass with the TR60 and NF 270 for higher than 6 ppm initial fluoride feed. Various types of membranes of the nano-filtration composite with reverse osmosis modules were reported by the researcher and are mentioned or cited in Table 2.

Electrodialysis-Based Removal

The revolution and advancement in techniques for decontamination, the electrodialysis (ED) technique was first commercialized in 1950 for the treatment of brackish water [51, 52]. Electrodialysis is a membrane-based removal technique that shows ionic solute separation using an electric field or direct current and the Donnan effect [53]. In this process, by applying a direct electric potential between the electrodes, the anion showed migration toward the anode through the positive charged membrane or cation towards the cathode through the cation exchange membrane. The membrane was synthesized using synthetic fibres, which caused high mechanical strength and showed coating of ion exchange group on the polymer matrix, as shown in Fig. 3. The membrane is ion-selective, which may be homogenous and heterogeneous in composition, having a thickness of about 0.1–0.6 mm [54]. The electrodialysis technique is a simple process that shows the best hydraulic recovery and lower residue production [55]. However, this technique gained wider attention due to its advantages. It is more cost-effective than other membrane techniques when used against pre-treated (good quality) water, less sensitivity to membrane fouling, high recovery rate, and withstands at high temperatures with the same efficiency. It showed high stability in all domains of pH value. In addition to its vast application [56], ED techniques attract attention towards advanced water treatment such as to reduce inorganic, i.e. bromide [57], radium [58], arsenic [59], nitrate [60]; chromium [61], lead [62], seawater desalination [63], iron and manganese [64], desalination of wells [65], recovery of RO rejects [66] and many more industrials applications [67, 68]. Electrodialysis application for defluorination of different water sources such as groundwater, surface water, or municipal wastewater was quite common [53, 69]. Various types of electrodialysis techniques are classified, such as conventional electrodialysis, bipolar electrodialysis, electrodialysis reversal, electrodialysis with MVA, electrodialysis metathesis, electrodeionisation with ion exchange resin for the wastewater treatment or defluorination of water as shown in Fig. 3a [70, 71]. Banasiak and Schäfer [72] reported a defluorination using the batch electrodialysis technique, which concluded that organic matter enhanced the removal of fluoride and boron from the synthetic solution by forming the boric acid complex. Sahli et al. [73] reported the removal of fluoride from underground brackish water using the combination of adsorption by chitosan and ion exchange membrane (electrodialysis). The technique concluded that the composite of techniques enhanced the removal efficiency by the principle of salinity control using electrodialysis and elimination of fluoride using the adsorption of the chitosan. Lahnid et al. [74] reported the economic evaluation of defluorination of the industrial plant by utilizing the electrodialysis with a capacity of 2200 m^3/day water consumption for 50,000 per Capita. They concluded the overall capital cost was about €833,207 with an operating cost of €0.154/m^3. Arar et al. [75] reported defluorination in the presence of co-ion, which affects the rejection of fluoride from the aqueous solution. In the case of a binary mixture of fluoride and chloride, chloride decreased the removal efficiency of the membrane by 63%. Still, in the same proportion binary mixture of fluoride and sulfate, the removal efficiency for fluoride was about 93%. It concluded the result for a ternary mixture of fluoride, chloride, and sulphate with increasing the operation time showed the enhancement of removal efficiency to 96% after 20 min. Although Electrodialysis showed huge applications



(a) (b)

Fig. 3 **a** Various types of electrodialysis techniques used for water treatment. **b** Basic principle of electrodialysis (Dimagreyl. "selective electrodialysis" web page, June 18, 2012, https://www.deviantart.com/dimagreyl/art/Selective-Electrodialysis-309099410)

for the rejection of fluoride from various matrices as shown in Table 3. It showed many shortcomings such as unable to show a barrier effect against bacterial decontamination, the presence of high chloride, iron, and manganese reduces the electrode lifetime; post-treatment operation; high salt concentration of effluent; pre-treatment operation and polarity reversal in case of electrodialysis reversal shorten the life of electrodes in electrodialysis operation.

The instrument-based defluorination techniques used on routine bases worldwide showed disadvantages such as pre-treatment of effluent, far from the user's hand, high energy consumption, fouling of membranes, high operational cost, and post-treatment of effluent. Many researchers showed a

Table 3 Summarized outlines of various electrodialysis membranes for the defluorination and their rejection rate

Sr. no.	Membrane types	Operational voltage	Source of analysis	Flow rate/temperature	Rejection/recovery (%)	References
1	NEOSEPTA ACS and NEOSEPTA CMX-Sb	06–24 V	Groundwater	100 m^3/h/29 °C	94	[76]
2	AEM and CEM	2 V/cell pair	Brackish water	8 L/H	87	[77]
3	PC-SK or PC-SA and CMX OR AMX	12 V	Tap water	90 L/H	92	[78]
4	AEM and CEM	13.4 V	Secondary effluent	102.5 L/H	99.69	[79]
5	ACS and CMX	5–15 V	Brackish water	180 L/H	80	[80]
6	AFN/CMX and ACS/CMX	10 V	Brackish water	–	63 and 59.09	[80]
7	CMX-SB and AMX-SB	5–25 V	Brackish ground water	30–70 L/H	94	[81]
8	Neosepta® CMX and AMX	5–10.21 V	Synthetic NaF solution	1.6 L/H	–	[82]
9	NEO-SEPTA ACS and AXE01 and NEOSEPTA CMX	–	Ground water	–	50/95	[83]
10	Selenium AMP and photo polymeric MZA	–	Artesian well water	–	97/40	[84]

leading hand toward a more efficient and satisfactory technique, Adsorption for water decontamination, because of low operational cost, high surface area, reusability, and dip to-discard act, advantages over the instrumental-based methods.

Adsorption-Based Removal

Adsorption is a surface phenomenon in which molecules or ions bind to the surface of adsorbate due to physical forces or chemical forces rather than penetration to the volume. The adsorption technique may be physical adsorption, which showed less activation energy for the adhesion of ions and fast removal of ions from the adsorbent. However, due to weak binding efficiency, chemisorptions overcame physisorption due to high stability and binding efficiency for the respective ions. But in the case of adsorption, both forces are simultaneously not isolated. That is why low-cost operation, easy-to-handle, and high reusability showed that adsorption is highly efficient for worldwide decontamination applications in core research areas. A wide range of adsorbents such as modified or natural zeolite, kanuma mud, modified sand, calcium-coated sand, chitosan, activated charcoal, activated rice husk, nano-adsorbents, and magnesium coated biochar, magnetic biochar, non-magnetic biochar, and minerals-based sorbents was used for the removal of fluoride from wastewater since long time as shown in Fig. 4.

Activated Sand-Based Adsorption

Many researchers reported sand as a highly efficient adsorbent for removing fluoride from water due to its high surface area, pore volume, low operational cost, and reusability of sand adsorbent. Togarepi et al. [85] studied the defluorination efficiency of thermally activated sand by calcinations for 3 h at a high temperature around 550 °C. The activated sand was modified with the iron oxide (10% Fe_2O_3) in excess of NaOH solution and dried in the oven overnight before the defluorination of water. The activated sand showed 90% fluoride removal efficiency, followed by Freundlich isotherm with an adsorption capacity of 10.3 mg/g at optimized

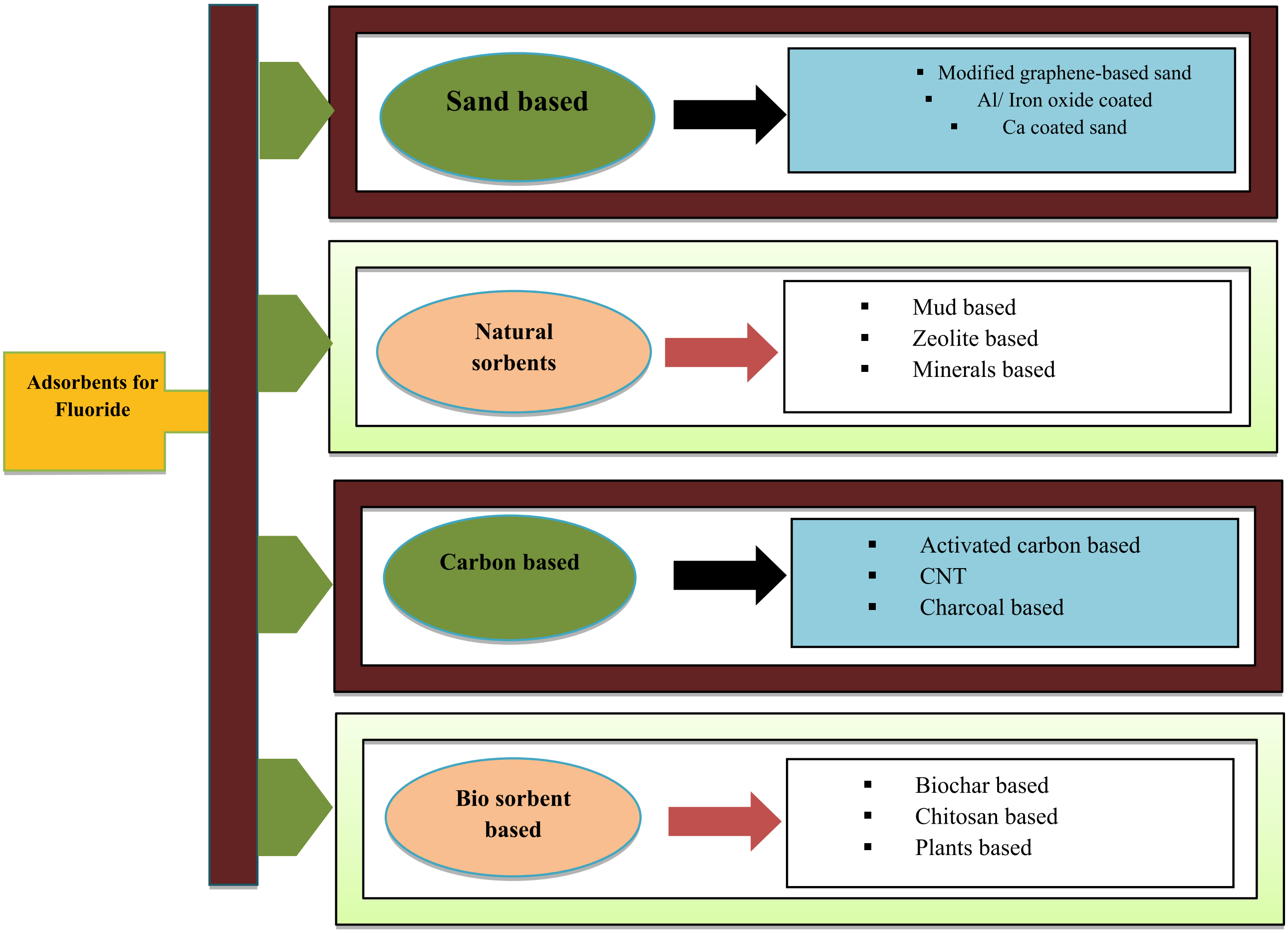


Fig. 4 Illustrates a graphical depiction showcasing various types of adsorbents for defluorination

conditions. Gogoi et al. [86] studied the locally available sand of Kanai Ghats, which has 90% silica in composition. The sand was coated with iron oxide (Fe $(NO_3)_3$ by repeated pyrolysis treatment under temperatures of 110 °C and 600 °C. The modified sand reported 89% removal efficiency against the uncoated sand, which showed only 7% removal efficiency, followed by Langmuir isotherm and pseudo-second order of the kinetic model. They examined a Langmuir adsorption capacity of 2.04 mg/g at the optimized conditions. Liyanage et al. [87] studied the defluorination efficiency of chemically modified sand with multiple coating of graphite oxide by repeated pyrolysis at 120 °C temperature. The reported modified sand is a dual-functionality cord for fluoride and turbidity removal. Based on the Hill adsorption model, multiple layers of graphite oxide on the sand showed 70% efficiency in the case of optimized pH (6.30).

Natural Sorbent-Based Adsorption

To overcome the traditional techniques, natural sorbents gained enormous attention from researchers to decontaminate wastewater. Natural sorbents take over conventional methods due to low cost, high sorption capacities, high porosity with mechanical solid stability, and heterogeneity of surface with molecular sieving. Various types of natural sorbents, such as clay minerals [88], natural zeolites [89], sedimentary rocks [90], and limestone [23] were used for the defluorination of water. Gómez-hortigüela et al. [91] reported natural zeolites from Ethiopia without further treatment. The different zeolitic structure was evaluated, and concluded that analcites and mordenite zeolite showed the maximum sorption efficiency for fluoride. These reported zeolites also showed the regeneration treatment with 1 mM NaOH and the recovered efficiency of 87%.

In some cases, natural zeolite sorption efficiency was enhanced by the chemical modification of zeolite surface, such as natural Stilbite zeolites modified with the $FeCl_3$, which was examined by Sun et al. [92]. The sorption efficiency was enhanced due to electrostatic attraction between the fluoride and iron ions present on the surface of the zeolite. This showed the pseudo-first-order of the kinetic model followed by Langmuir adsorption isotherm with an adsorption capacity of 2.31 mg/g. Regeneration ability was evaluated using the 1 mol/l of HCL solution as an eluent. Based on the oppositely charged binding sites on the adsorbent surface enhanced the adsorption efficiency of fluoride by the electrostatic attraction. Dessalegne et al. [93] examined the fluoride sorption efficiency of commercialised zeolite and natural Stilbite zeolite which was supported with aluminium hydroxide in different-2 ratio as AO-Z (2:1, 6:1). It showed good adsorption efficiency for fluoride followed by Langmuir model of adsorption with maximum adsorption capacity of 12.12 mg/g (STI-AO; 2:1) and 7.26 mg/g (STI-AO; 6:1). Velazquez-peña et al. [94] examined the defluorination efficiency of different natural zeolitic structure, i.e. Clinoptilolite, Mordenite, Chabazite supported with Fe–Zr and followed the optimisation for the maximum adsorption capacity. The Fe–Zr-modified mordenite and chabazite showed the highest adsorption capacity 3.15 mg/g or 2.6 mg/g, followed by a pseudo-second-order kinetics model in slightly acidic conditions. Alagumuthu [95] reported the removal efficiency of montmorillonite clay for fluoride from the water. Adsorption of fluoride under the optimised condition (natural pH, time = 50 min, particle size 75 µm) followed both the Langmuir and Freundlich adsorption isotherm in different range of temperatures (30–60 °C) with maximum adsorption capacity of 1.910 mg/g. The maximum adsorption capacity was reported on the high reaction temperature, showing the sorption as an endothermic reaction. Meenakshi et al. [96] reported the defluorination efficiency of kaolinite clay mineral, which was mechanically activated and increment of surface area from 15.11 to 32.43 m^2/g. However, the chemical activation of South African Clay using 1% Na_2CO_3 and dilute HCL was also reported for fluoride removal by Coetzee et al. [97]. It examined that clay having the gibbsite and Al_2O_3 showed a high potential for fluoride adsorption.

Herbal-Based Techniques

Herbal-based techniques in which plant waste biomass and plant parts such as granulated roots, stem, extract and leaves were used for utilization in various research fields. The demand for eco-friendly, cheaper, locally accessible solid adsorbents for water treatment has increased in the last few years. Plant-based adsorbents (green adsorbents) come out on top because of their vital properties such as eco-friendly, readily available, recyclable dry biomass, high efficacy with no hazardous effluent after treatment and easy to operate without instrumentation. Adeniyi and Ighalo [98] reviewed various research based on removing heavy metals, chemical pollutants and organic dyes using the plant leaves from the aqueous solution. By taking along this concept, the defluorination efficiency of the citrus liminum (lemon) leaf was examined by Tomar et al. [99]. The defluorination efficiency was reported as much as 70% for the 2 mg/l fluoride concentration under the optimized condition. The optimized conditions were evaluated for high efficiency in an acidic nature with an agitation time of 145 min by taking 10 g of adsorbent dose. Ganapaty Alagumuthu et al. [100] reported a defluorination efficiency of 83.77% using activated carbon prepared by *Cynodon dactylon* biomass under optimized conditions. The regeneration or reusability was also examined as 67.4% by using 2% NaOH, which showed it is a highly cost-effective biosorbent for defluorination. Due to the leading foot towards biosorbent, many more were also reported for defluorination, such as devdaru leaf powder [101], sandalwood leaf powder [102], jamun leaf ash [103], neem leaf powder [104], *Azolla filiculoids* [105] and *Ficus benghalensis* leaf powder [106] along their removal efficiency outlined in Table 4.

Nanocomposite-Based Adsorption with Bio-polymer

A significant population boom leads to a lack of sustainable approaches for water defluorination. Hence, it leads to health-related issues or is an invisible disaster for the people. Therefore, the intervention of nanotechnology or carbon-based adsorbent, i.e. biochar [107], graphene [108], or modified biochar was likely used by the researcher for the defluorination approaches as reported in the TABLE. Nanocomposites are composites having one phase that has nanoscale physiology such as nanotube, Nano fullerene and nanoparticle. Still, chemical treatment or instrument involvement was used to prepare adsorbents, and the need for sustainable approaches was marked as a priority for the researchers for defluorination. Therefore, biopolymer-based techniques showing massive diversification, highly versatile, biodegradable, and renewable nature characteristics take over other water purification methods. Biopolymers are polymeric biomolecules synthesized by living organisms or natural sources. Natural biopolymers such as cellulose, alginate, chitin, and chitosan have been adopted for water purification [109]. Aranaz et al. [110] reported the application of chitosan and composite for sustainable water treatment. Voisin et al. [111] reported nanocellulose as a potential biopolymer for water purification. As a way, many researchers reported chitosan or nanocellulose and their composites for removing fluoride from the water and wastewater, as shown in Table 5.

Table 4 Summarizing different bio-sorbents and their effectiveness for the removal of fluoride

Bio sorbents	Defluorination efficiency (%)	References
Cynodon dactylon-activated carbon	83.77	[100]
Citrus leaf powder	70	[99]
Devdaru leaf powder	77	[101]
Sandalwood leaf powder	75	[102]
Jamun leaf ash	77.8	[103]
Neem leaf powder	80	[104]
Ficus benghalensis	92.2	[106]
Azolla filiculoids	98	[105]
Epipremnum aureum (money plant)	88	[12]

Environment and Economics Aspects

Various methods for removing fluoride from water have been reviewed, considering their effectiveness and environmental impact. However, advancements in these techniques have led to concerns about their negative effects on the environment. For instance, precipitation and coagulation methods generate concentrated sludge or precipitates, increase water hardness, and pose challenges for proper disposal, which can harm the environment. In response to these issues, membrane-based techniques were introduced, which partially address the problem of mineral imbalance but still generate concentrated brine sludges. To address these environmental concerns, researchers have turned to an eco-friendly and more sustainable defluorination technique: adsorption. This method not only mitigates environmental issues but also offers cost-effective advantages. The economic analysis of these techniques is crucial in determining their feasibility. Precipitation and coagulation approaches incur high costs due to chemicals, infrastructure, and post-treatment expenses, which can exceed \$0.38 per cubic meter [138]. This financial burden has driven researchers to seek cost-effective alternatives. While membrane-based approaches offer high efficiency, their operating costs, approximately \$2880 per year, are significantly higher than other methods [139]. This is why adsorption-based approaches have gained popularity, as they are cost-effective, economically viable, and easy to operate without the need for expensive equipment.

Conclusion and Future Perspectives

This review paper extensively covers a wide range of techniques for removing fluoride from various sources. The defluorination process begins with instrumental-based methods and culminates with biopolymer adsorption. The paper also discusses several emerging methods that have enhanced fluoride removal efficiency, including nano-filtration, electrodialysis, reverse osmosis, and nanocomposites. However, instrumental-based methods have many drawbacks that hinder their commercial viability, making adsorption-based techniques more favourable due to their simplicity and sustainability. This review serves as a valuable resource for scientists seeking to develop more effective adsorbents, explore chemical-free regeneration of composites to reduce costs, integrate with advanced monitoring and control devices, and improve scalability and adaptability.

Table 5 Summary of fluoride adsorption capacities of adsorbents and their respective composites

Sr. no.	Adsorbent	Adsorption capacity	Isotherm model	Kinetic model	pH	References
1	Aluminum impregnated chitosan	84% removal	Freundlich isotherm	–	6.5	[112]
2	Iron impregnated chitosan	20.75 mg/g	Freundlich isotherm	Pseudo-***second order	–	[113]
3	Aluminum chitosan iron hydro gel	31.16 mg/g	–	Pseudo-second order	5.0	[114]
4	La(III) incorporated carboxylate-chitosan	4711 mg/kg	Freundlich isotherm	Pseudo-second order	3–12	[112]
5	Carboxylate-chitosan beads	11.11 mg/g	Both Freundlich and Langmuir isotherm	Pseudo-second order	7.0	[140]
6	Glutaraldehyde/chitosan beads	7.32 mg/g	Both Freundlich and Langmuir isotherm	Pseudo-second order	7.0	[140]
7	Zeolite/chitosan composite	10.5 mg/g	Langmuir isotherm	Pseudo-first model	3.0	[115]
8	Mesoporous alumina/chitosan composite	8.624 mg/g	Langmuir isotherm	Pseudo-second order	3.0–9.0	[116]
9	Chitosan cryogel cross linked ethylene glycol diglycidyl ether	280 mg/g	Both Freundlich and Langmuir isotherm	–	6.0	[117]
10	Nano CaO/Fe(OH)$_3$ chitosan composite	41.36% fluoride removal	Both Freundlich and Langmuir isotherm	Pseudo-second order	7.10	[118]
11	Aluminum oxide modified CNT	9.6	–	Pseudo-second order model	6.0–9.0	[119]
12	Carbon and CNT Composite	–	Langmuir isotherm	Pseudo-second order model	–	[120]
13	Hydroxyapatite modified CNT	11.05	Freundlich model	Pseudo-second order model	6.0	[121]
14	Chitosan/CNT composite	975.4	Freundlich model	–	3.0	[122]
15	Iron–zirconium-modified zeolite		Freundlich model	Pseudo-second order model	4.6–6.4	[94]
16	Iron modified stilbite zeolite	2.31 mg/g	Langmuir model	Pseudo-first-order model	6.94	[92]
17	Montmorillonite clay	1.910 mg/g	Both Freundlich and Langmuir isotherm	–	2.0	[95]
18	Bauxite clay	–	–	–	5.0–6.0	[88]
19	Al, Fe, or Ca-modified silty clay	–	–	–	6.0	[123]
20	Aluminum hydroxide-modified stilbite zeolite	12.12 mg/g or 7.26 mg/g	Langmuir isotherm model	–	5.0–8.0	[93]
21	Natural zeolite (stilbite, analcime, mordenite)	0.47 mg/g	–	–	5.0–8.0	[91]
22	Kaolinites	0.810 mg/g or 0.7822 mg/g	Both Freundlich and Langmuir isotherm	–	2.0	[96]
23	MgO impregnated Biochar	83.05 mg/g	Langmuir isotherm of adsorption	Pseudo-second order model	8.0	[107]
24	Watermelon Rind Biochar	9.5 mg/g	Freundlich isotherm	Pseudo-second order model	1.0	[124]
25	MCSBC/CSBC	4.11 mg/g/6.42 mg/g	Langmuir isotherm	Pseudo-first order model	2.0	[125]
26	Cocus Nucifera shell biochar	82.45% removal	–	–	6.5	[126]
27	FeOOH/ FeOOH + Ac/GO	17.65 mg/g/19.82 mg/g	Both Freundlich and Langmuir isotherm	Pseudo-second order model	2–10	[127]

Table 5 (continued)

Sr. no.	Adsorbent	Adsorption capacity	Isotherm model	Kinetic model	pH	References
28	Aluminium sulfate @ graphene hydro gel	33.4 mg/g	Langmuir isotherm model	Pseudo-first-order model	7.2	[128]
29	Zr incorporated graphene-coated sand	6.12 mg/g	Both Freundlich and Langmuir isotherm	Pseudo-second order model	4.0	[129]
30	Zr–graphene hybrid adsorbent	44.14 mg/g	Langmuir isotherm	–	–	[108]
31	Zr-CTS/GO membrane adsorbent	29.06 mg/g	Freundlich isotherm	Pseudo-first and second order model	3–11	[113]
32	Iron–aluminum oxide nanoparticle/GO	64.72 mg/g	Langmuir isotherm	Pseudo-second order model	3–9	[130]
33	Protonated chitosan beads	1664 mg/kg	Both Freundlich and Langmuir isotherm	Pseudo-second order model and interparticle diffusion model	–	[131, 132]
34	Al-impregnated Chitosan	1.73 mg/g	Both Freundlich and Langmuir isotherm	Pseudo-first order of kinetics	6.5	[133]
35	Magnetic nanocomposite chitosan	15.38 mg/g	Langmuir isotherm	Pseudo-second order model	5.0	[134]
36	Cellulose nanofiber-PANI-templated ferrihydrite (CNPFH) nanocomposite	50.8 mg/g	–	–	3–4	[135]
37	Nanocellulose–PVP matrix	11.363 mg/g	Langmuir and Temkin isotherm	Pseudo-second order model	–	[136]
38	PDMAEMA-grafted cellulose fibres	8.59 mg/g	Both Freundlich and Langmuir isotherm		3–10	[137]

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Author Contributions RK created significant contribution to whole conception, data accumulation and drafting of the review article. SN donated significantly to the work' understanding design and draft revision and approved the compiled version of the article. All authors have read out and approved the manuscript.

Data Availability It is a review paper. There is no experimental data access.

Declarations

Conflict of interest The authors declare that there are no competing interests.

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