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► **To cite this version:**

B. Senthil Rathi, P. Senthil Kumar, Gayathri Rangasamy, Michael Badawi, Tejraj Aminabhavi. Membrane-based removal of fluoride from groundwater. *Chemical Engineering Journal*, 2024, 488, pp.150880. 10.1016/j.cej.2024.150880 . hal-04883820

HAL Id: hal-04883820

<https://hal.univ-lorraine.fr/hal-04883820v1>

Submitted on 13 Jan 2025

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Membrane-based removal of fluoride from groundwater

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Abstract

One of the biggest issues facing the globe today is fluoride poisoning of water, which is a severe risk to the well-being of people since water is essential for consumption from both natural and man-made sources. Fluoride in water can be removed using a variety of methods, although the issue is still not completely solved. Because of its ionic size and reactivity, fluoride can be hard to remove from water-based solutions using traditional electrochemical, precipitation, and adsorption methods. One of the latest innovations that has been shown to be highly successful in bringing fluoride levels down to the required levels is the use of membranes. This review discusses fluoride and its characteristics, sources, impacts on the environment and humans, fluoride measurement techniques, and fluoride applications. The efficacy of different types of membrane separation processes are covered with respect to techniques developed, advantages and drawbacks of defluoridation processes. This review critically examines membrane-based techniques for different defluoridation processes including nanofiltration (NF), reverse osmosis

(RO), membrane distillation (MD), electrodialysis (ED), and forward osmosis (FO). Furthermore, the study addresses developments in ceramic membranes, electrodeionization and membrane capacitive deionization, hybrid membrane techniques, and life cycle assessment (LCA) for membrane-based defluoridation process. The work meets the SDGs goal #6 of clean water supply.

Keywords: Defluoridation; electrodeionization; electrodialysis; LCA; membrane-based treatment; membrane capacitive deionization; membrane distillation; nanofiltration; reverse osmosis

1. Introduction

Fluoride (FL) contamination in aquifers has been identified as one of the globe's most severe issues. FL is one of the World Health Organization's (WHO) water pollutants for use by people, along with nitrate and arsenic, creating widespread health concerns. Elevated FL concentrations (FLC) in groundwater (GW) are found in many places around the globe (Waghmare and Arfin, 2015). FL is abundant in the earth's environment and is often discharged into aquifers through the gradual decomposition of fluorine-containing minerals (Bhatnagar et al., 2011). FL is found mostly as fluorspar, sellaite, fluorapatite, and cryolite. It appears in sedimentary deposits as fluorspar and in igneous stones as cryolite. These FL minerals are extremely difficult to dissolve in water. FL will thus be detected in aquifers only when circumstances encourage their solubility or when high FLC-containing wastewater (WW) is released into aquatic systems from industries (Mohapatra et al., 2009).

FL, as an extremely negatively charged element, is exceptionally attracted to ions that are positively charged calcium ions. Consequently, the effect of FL on mineral-rich tissues, such as bones and teeth, is clinically relevant because these tissues absorb the greatest amount of FL, which accumulates as calc-fluorapatite crystals, due to their high calcium content (Maheshwari, 2006). FL content in water can be measured by following techniques: Electrochemical Methods, Chromatographic Methods, Spectroscopic Methods, sensors, and titration methods (Tolkou et al., 2021). Toothpaste is the most essential FL treatment that is advised for everyone. The necessity for further FL intake is determined by the level of dental activity. The caries-preventive consequences of concentrated FL solutions, gel-like substances, or coatings are not significantly different (Øgaard et al., 1994). The FLC of the liquid phase and cells were the same as in newly prepared specimens after the application of FL gel, and its detrimental impact on pH decrease and electrolyte metabolism was much stronger than before.

FL release and antibacterial activity of ancient glass ionomers might be considerably boosted by applying FL gel to the material (Seppa et al., 1993).

Because of its ionic size and responsiveness, fluoride removal (FLR) from water-based solutions is challenging utilizing traditional electrochemical, precipitation, and adsorption procedures. One of the latest methods shown to be highly successful in dramatically decreasing FL to required standards levels is membrane technology (MT). Membrane systems examined for the FLR process include RO, NF, MD, ED, and FO (Damtie et al., 2019). ED device for quick desalination and purification of synthesized saltwater from groundwater throughout a wide operating electric potential ranging from 5-25 V, rate of flow between 30-70 L/h, and FLC of 10-250 ppm. FLR was 94% high at pH-independent. Yet, FLR decreased with increasing salinity from 94-72%, implying that increasing salinity of groundwater will impair FLR in ED (Amor et al., 2001; Keri et al., 2011; Nataraj et al., 2006; Nataraj et al., 2007).

In MD, commercially available hydrophobic 0.22 mm porous polytetrafluoroethylene and polyvinylidene FL membranes reject over 99% of FL, generating fluxes of up to 39.3 and 26.4 kg/m² h at 60°C, correspondingly (Damtie and Choi, 2017). The RO unit ran at 80% restoration with no scaling inhibitors. Calcium fluoride was nearly four times excessively saturated in concentration. To prevent precipitate formation, conventional flushing was used. To identify the beginning of scaling sufficiently early to begin cleaning, sacrificial components were used on the concentrate. After six months, the first preventative cleaning was performed. After one and a half years, significant fouling with natural organic debris developed. This might be managed with severe alkaline cleanings at pH more than 12 and temperatures greater than 35°C (Sehn et al., 2008).

The effectiveness of NF membranes in removing FL from high FL content water was examined. The membranes employed are conventional thin-film composite membranes that are charged negatively. Pore radius, constant surface electrical potential, and pure water permeability were used to evaluate membrane efficiency. The variables' values were computed by applying the experimental data from 100 to 1000 mg/L Sodium fluoride to the model equation was created (Hu and Dickson, 2006). The efficiency of a three-layer film composite FO membrane's capacity to hold FL ions was investigated. To improve the hydrophilic properties as well as the retention capability of the FO membrane, a cellulose nanocrystal (CNC) layer was coated over the porous substrate. Boric acid was utilized for chemical transformation to increase the dispersion of CNC. The FO tests revealed that the water flow of the three-layer film composite membrane/CNC was enhanced by 46.4% in comparison to the

conventional three-layer film composite membrane. The three-layer film composite / boric acid -CNC could be preserved at 97.0% (Huang et al., 2023).

Electrodeionization (EDI) technique is used to provide FL-free water in a continuous method. The membranes and resins exhibit remarkable electrochemical characteristics as well as mechanical strength. The FLR from three tap water concentrations indicated the importance of water constitution on the efficiency of the EDI unit in terms of the selectivity of FLR and utilization of energy. During EDI, three potentials are used in various sets of tests, with the 12 V/cell pair being more effective with a reduced power usage of 4.6 kwh/kg and 37% current effectiveness (Gahlot et al., 2015). Conducting capacitive deionization (CDI) at a fixed voltage results in WW with varying intensity of ions over time. When compared to a fixed voltage, charging CDI at a fixed current offers various benefits, most notably a consistent and controllable wastewater ion concentration. The viability of FLR extraction from saline ground waters using one-time-pass constant-current CDI in both 0-V and reverse-current desorption regimes was examined, and a mathematical equation was constructed to characterize the particular electro-adsorption of FL and Cl (Tang et al., 2016a).

FL contaminated water for drinking was effectively treated utilizing a hybrid approach known as electrocoagulation-microfiltration, and FLC were successfully reduced to the allowable level of 1.5 ppm. The highest elimination was obtained using operational conditions of 15 A/m², 0.005 m inter-electrode spacing, and 1.95 g/L NaCl concentration (Waghmare and Arfin, 2015). The life cycle assessment (LCA) is an effective technique for analyzing the ecological effect since it considers all loads throughout the process's life cycle. LCA, on the other hand, has been applied to the MT for FLR (Goyal and Mondal, 2022).

Although many strategies for removing fluoride from wastewater have been thoroughly researched, this paper adds new information by thoroughly examining the effectiveness of both conventional and cutting-edge membrane-based approaches. Thus, the review focuses on a comprehensive study of fluoride characteristics, sources, effects, measuring methods, and traditional removal strategies by exploring novel membrane technologies including electrodeionization, membrane capacitive deionization, and ceramic microfiltration. It also does a life cycle study of membrane-based fluoride removal systems and presents a hybrid membrane technology for fluoride removal. This broad technique narrows the gap in the literature and offers useful information for long-term solutions to the problem of fluoride pollution and its removal from water bodies.

2. Fluoride

FL occurs naturally in tiny amounts in water, soil, vegetation, and mammals. When FL is consumed, a portion of it is absorbed by tissues in the body, leading to long-term accumulation in the bones and teeth. A significant reduction in childhood cavities was observed in communities exposed to higher FL levels in drinking water (Harrison, 2005). FL is quite widespread in the Earth's crust and can infiltrate aquifers through naturally occurring mechanisms; soil near the base of a hill is likely to have an increase in FL due to leaching and weathering of fluoride-rich rock (Susheela et al., 1999). Fluoride in water that is consumed has been linked to both positive and negative health consequences (Nagendra Rao, 2003).

2.1. Physical and Chemical properties of fluoride

FL occurs naturally in groundwater (GW), freshwater (FW), and seawater (SW), as well as in rainfall, particularly in metropolitan locations. FL levels in SW typically range from 0.6 to 1.5 ppm. In contrast, the chloride content in seawater is around 19,000 ppm. The low FL content reflects the insoluble nature of alkaline earth FL (Buzalaf et al., 2011). Table 1 represents the physical and chemical properties of FL.

2.2. Sources of fluoride in waste water

Among the most common contaminants in groundwater is fluoride (FL). According to Vithanage and Bhattacharya (2015), fluorosis affects more than 200 million individuals across 25 countries (Tashauoei et al., 2023). FL is naturally present in food, water, soil, and various minerals like fluorite and fluorapatite. FL is an element that occurs in conjunction with other elements to form a compound (Chidambaram et al., 2003). Water, food, industrial exposure, medications, cosmetics, and other items are common ways for FL to enter the surroundings and the human body (Ghosh et al., 2013a). Chemical studies revealed that specimens derived from limestone had low FL concentrations, whereas specimens from marl samples have significant FL contents in soils and rocks. It appears that marl is the most probable source of FL in the research region since marl specimens had a significantly higher FL content and noticeable amounts of chlorite when examined under a transmitted light microscope. Furthermore, marl is interbedded with limestone that has a comparatively high FL concentration. This might be the cause of the FL levels in tests on water not varying spatially. According to the findings, micas and clay minerals are probably sources of FL (Battaleb-Looie et al., 2012). Fig. 1 depicts the sources and pathway of Fluoride in the environment.

The accessibility and solubility of FL minerals, the speed at which water flows, pH, temperature, the level of bicarbonate and Ca ions in water, and many other variables all affect the amount of FL present in the aquifer (Keshavarzi et al., 2010). In comparison with the peak

summer concentration, the highest FL concentration found in the winter was twenty times greater. Beijing had severe wintertime FL pollution as a result of higher coal use for heaters. According to Feng et al. (2003), the primary sources of FL were thought to be coal combustion and soil dust. According to some research, people may absorb FL many times more via food, beverages, dirt, and various other sources than from water (Kahama et al., 1997). The FL content in the different products is shown in Table 2.

2.3. Effects of fluoride

FL exists abundantly in our surroundings, but humans only ingest a minimal quantity of it. Exposure can happen through breathing, eating certain foods, and using FL supplements. Fluoridated water is the most significant determinant for FL presence in the diet (Kanduti et al., 2016). Excessive FL buildup in the surroundings presents major health concerns to individuals, plants, and animals. Fig. 2 gives the acute and chronic impacts of FL on animals, plants, and human beings. In addition to endangering human health, this also has an adverse impact on the growth and development of organisms and the food chain, which in turn disrupts ecological equilibrium. Many recent studies have concentrated on the molecular pathways connected to FL poisoning. These studies have demonstrated that FL may cause damage to mitochondria, stress to the endoplasmic reticulum, oxidative stress, and changes in gene regulation. It can also control intracellular redox homeostasis (Zuo et al., 2018).

The impact of FL on the biological mechanisms that underlie the development of tooth tissues, namely dental enamel, is significant. More stable crystals are produced when FL is included in enamel apatite. Additionally, local fluoride mineral overabundance levels will be higher, which will promote crystal development. Modulating matrix proteins and proteolytic enzymes bind more tightly as a result of these changes in crystal chemistry and shape, including greater ionic and hydrogen interactions. Protein elements in matured tissue are hence better retained and undergo less breakdown. Since the elimination of matrix proteins is required for unimpaired crystal formation, this is probably the cause of permeable fluorotic tissue. More thorough investigations into modifications to the functioning of cells, effects on particular protein organisms, and the nanotechnology of the apatite crystal surfaces will be necessary to address the unanswered questions regarding changes in cells and the precise causes for protein preservation (Robinson et al., 2004). As no other infectious agent or disease in humans is known to generate dentition alterations identical to those caused by FL, it is argued that fluorosis in the mouth provides a sensitive method of documenting prior FL exposure. It is feasible to achieve significant caries reductions without running the danger of fluorosis in the

teeth since the primary cariostatic impact of FL is not caused by its absorption by the enamel during the growth of teeth (Fejerskov et al., 1994).

Although it has long been known that the acidogenic dental microbiota inhibits the metabolism of carbohydrates through FL, it hasn't always been understood that oral bacteria differ greatly in how susceptible they are to FL (Hamilton et al., 1990). A significant relationship was found between FL concentration in water that is consumed and a greater likelihood of intellectual disability (IQ <70) and marginal intellect (IQ 70–79). The reference dosage's lower-bound confidence level was 1.85 ppm, while the reference dosage for the dosage-response correlation between IQ <80 and the FL concentration in water for consumption was 2.32 ppm. The baseline concentration for FL was determined to be 0.925 mg/L after accounting for fluorosis of the teeth and other food sources of FL. This result is quite near to the existing national Chinese norm of <1.0 ppm. Drinking water FLC higher than 1.0 ppm in locations where fluorosis is widespread may have a negative impact on children's intellectual development (Xiang et al., 2003).

FL promotes bone mass; however, the strength of the newly created bone may be diminished. FL treatment enhances the fragility of bones and cancellous but reduces cortical density of bone minerals. Because of all these effects, there is a minor tendency towards a reduction in vertebral body fractures but an increase in fractures at locations that have comparable proportions of cancellous and cortical bone or a preponderance of cortical bone. Therefore, our results do not support the clinical effectiveness of the FL—calcium diet for the prevention and treatment of osteoporosis, despite the significant rise in cancellous-bone mass (Riggs et al., 1990).

When exposed to air, soil, and water, FL halide had the greatest detrimental impact on plants of all the halides. In addition to its nutritional value, this element interfered with several physico-biochemical variables in plants, causing negative consequences whether or not there were any obvious signs of damage. Higher levels of FL in vegetation also inhibit plant growth by interfering with many processes related to photosynthesis, respiration, and creation of proteins, metabolism of glucose, and nucleotide synthesis. The two most crucial processes for plants to get and use their fundamental energy are respiration and photosynthesis. FL is quite important in terms of effectiveness on these pathways (Kumar et al., 2017). Elevated FL levels in soil have the potential to severely endanger plant life, destroy the activity of soil microbes, disturb soil the environment, and contaminate soil and water (Shahab et al., 2017).

Animals that exhibit significant symptoms and diseases of fluorosis of the teeth are those who consume contaminated plants and breathe in fluorine (as gases and dusts) regularly

(Kessabi et al., 1984). A disruption to normal teeth, the emergence of medullary lesions, and a lack of hunger that may result in weight loss or reduced milk production are the main signs of the chronic illness that develops, which was most thoroughly investigated in cattle (Suttie et al., 1964). FL has the ability to create oxidative stress that can modify intracellular redox homeostasis, increase protein carbonyl levels and peroxidation of lipids, change the expression of genes, and trigger apoptosis. According to Giri et al. (2016), genes involved in the physiological stress response, enzymes that regulate the cell cycle, interaction between cells, and the transmission of signals are among those that FL modulates.

2.4. Techniques to measure fluoride

FL ion-selective electrode (ISE) potentiometric measurement of FL content in solutions is easy, dependable, and reasonably priced. ISE can measure very low FL-ion levels (down to 10^{-6} mol/dm³) while controlling the level of hydroxide and metal-interfering ions as well as the ion strength of a solution. The direct potentiometer approach may be used for estimating the FL concentration of the samples, and the conventional method of addition can be used in cases when the FL level is very low. The most widely used and practical techniques for determining FL and chloride ions are electroanalytical techniques based on potentiometry with ISE. Because of FL-ISE's excellent selectivity, precision, and low detection limits, it may be used to measure FL levels in water used for drinking. This study's benefits include quick analysis time, no need for sample pretreatment procedures, an easy-to-use measurement method, and somewhat inexpensive equipment costs (Sunitha and Reddy, 2014). Table 3 gives the information about the techniques to measure FL.

While FL may be measured using titrimetric, colorimetric, or ISE methods, ion-chromatography (IC) methods are often not used to estimate FL in soil extract. This is due to the extract including a significant quantity of acetate, which elutes along with FL during isolation and causes a lot of polyvalent anions to build up in the column, which raises noise in the background. A standardised IC technique has been developed to determine FL in soil extracts. IonPac AG4A-SC guard column, IonPac AS4A-SC analytical column, ASRS-I conductivity suppressor, and conductivity detector were utilised in the method's development. A 20 μ L specimen was used. 2.6 minutes after injection, 0.05 M Na₂B₄O₇ was circulated to clean the column. An eluent solution (0.005 M) was utilised at a flow rate of 2.0 mL/min. Following a 1.5-minute washing period, columns were adjusted with 0.005 M Na₂B₄O₇ eluent. The entire procedure took thirteen minutes to complete. After every study, it was discovered that the column needed to be cleaned with concentrated eluent since background conductivity and noise levels rose, most likely as a result of polyvalent anions contaminating the column.

118 distinct soils' water extracts were used to assess the FL concentration (Saha and Kundu, 2003).

IC methods are designed to identify and measure FL in a calcium carbonate matrix. After being dissolved in hydrochloric acid and processed to mostly eliminate the chloride ions, solid specimens are examined using inductive correlation (IC). According to Lefler and Ivey (2011), the 3σ threshold of detection using these techniques is 0.2 mg of FL/kg of calcium carbonate. ISE offers several benefits over the conventional UV/VIS spectroscopy approach, including accuracy, speed, economy, and sensitivity. Potentiometric ISE development is a highly intriguing topic with many potential uses for ion detection in water and other media. Ion species in a trace can be identified using the application of ISE. ISE are appropriate for industrial analysis, control procedures, monitoring the environment, and physiological evaluations. It has been utilised in the food business in recent years to measure a variety of ions, including calcium in dairy products, juice from fruits, and several vegetable varieties (Bratovčić et al., 2009).

Proton activation analysis, enzymatic, and fluorometric analysis are alternative techniques employed for fluoride analysis. Among these, fluorometric analysis requires minimal sample preparation and exhibits sensitivity to even trace amounts of material. Microdiffusion methods, such as the acid-hexamethyldisiloxane (HMDS) diffusion method, are regarded as the most precise means of sample preparation. These techniques facilitate the release of fluoride from organic or inorganic matrices. However, it is imperative for the analysts to exercise caution during sample preparation to prevent contamination, partial matrix release, and volatilization losses. Fluoride analysis often involves working with extremely small sample quantities that necessitates sample modification. Techniques employed include sample transfers using micropipettes, development of microfluoride-selective electrodes, and the adaptation of conventional electrodes for micro- and semi-micro volumes ranging from 0.005 to 5 μ L (Fluorides, 2003).

2.5. Applications of fluoride

FL is primarily used to enhance oral health, and toothpaste with FL is among the most widely used forms of self-applied FL worldwide. FL in toothpaste is quickly absorbed by demineralized enamel and plaque on the teeth, increasing the FL content in saliva. The application of fluoridated toothbrushes and toothpaste is the most effective strategy for preventing cavities. FL can promote the remineralization of enamel and reduce its demineralization. Topical use of FL, including FL toothpaste, FL paint, FL gel, and oral rinse, is available for preventive purposes. Additionally, FL is systematically used and added to dairy

products, salt, and water in several countries. In the medical treatment of early dental caries, FL is also employed. However, it's important to note that excessive FL consumption can lead to adverse reactions, and dental fluorosis can occur due to an excess of FL during tooth growth (Petersen and Ogawa, 2016).

FL is a double-edged sword, and while it offers benefits for oral health, excessive exposure can lead to issues. Dental fluorosis is a condition caused by an overabundance of FLs during tooth development. Therefore, it is crucial to strike a balance in FL usage to reap its benefits without causing harm. For the well-being of individuals, the World Health Organization (WHO) recommends making efforts to provide affordable fluoride toothpaste that can be used in underdeveloped nations. Additionally, when socially and technologically viable, water fluoridation offers significant benefits for public health (Petersen and Lennon, 2004).

3. Traditional methods to remove fluoride

Worldwide recognition of FL pollution in aquatic environments due to human and natural activities has been established. It becomes crucial to develop reliable and efficient technology for removing excess FL from aquatic environments (He et al., 2020; Nataraj et al., 2007; Saravanan et al., 2023). Many processes, including coagulation-precipitation, ion exchange (IE), membrane separation processes (MSP), adsorption (AD), etc., can be used for Fluoride Removal (FLR) from groundwater (GW). The various conventional techniques for removing FL from water are illustrated in Fig. 3. However, MSP and IE are the least common of these techniques due to their significant setup and maintenance costs, while the other two methods are widely used in India (Waghmare and Arfin, 2015).

Defluoridation methods that are extensively utilized include electrochemical, reverse osmosis (RO), electrodialysis (ED), and hybrid procedures that combine AD and dialysis. However, there is a current focus on developing clean, affordable procedures due to financial limitations and strict environmental regulations. Table 4 presents details of various conventional techniques for eliminating FL along with the conditions of their operation.

3.1. Electrodialysis

Electrodialysis (ED) has demonstrated effective performance in removing salt and fluoride from saltwater, but it faces challenges such as operating costs and polarization issues when treating equilibrated freshwater with high fluoride concentration (Sahli et al., 2007). In a study, ED was employed, utilizing an electric field to drive ionized elements from aqueous solutions through ion-exchange membranes (IEM), achieving significant fluoride removal in a relatively short treatment period of 6 minutes in both acidic and neutral media (Belkada et al., 2018).

In another application of ED, wastewater from the graphene industry was treated to predict and enhance operational parameters for defluoridation. The response surface approach was employed, considering factors such as operating voltage, initial fluoride level, and flow velocity. Under optimal conditions, a fluoride removal effectiveness of 99.69% was achieved for the treatment of the graphite industry's secondary wastewater (Wang et al., 2019).

The study also investigated the effects of initial fluoride concentration, the amount of salt, and the presence of organic matter on process efficiency. The results indicated that as the initial fluoride concentration in model solutions decreased, the separation performance improved. The desalination and defluoridation efficiency remained unaffected by the presence of humic acids in fluoride solutions undergoing ED treatment. The computed electrical energy consumption ranged from 0.121 to 0.895 kWh/m³, depending on the composition of the fluoride solution. The absence of monovalent and divalent ions resulted in higher fluoride removal (Majewska-Nowak et al., 2015).

Factors such as the bonding of sulfate and chloride ions with the ions in the membrane and the activity ratio of fluoride in the presence of monovalent and divalent ions were considered. The study found that the inhibitory effect of sulfate ions on fluoride movement was greater than that of chloride ions. Fluoride movement increased with higher feed phase intensity and current density, with chloride ions being superior to sulfate ions in driving ion movement. An actual water sample with 20.6 ppm of fluoride was subjected to ED, and the fluoride content could be reduced to 0.8 ppm, meeting the World Health Organization (WHO) requirement of 1.5 ppm (Ergun et al., 2008).

Studies conducted in saltwater with varying fluoride concentrations and a salt content of 2050 mg/L showed that higher potential led to improved defluoridation performance. The optimal applied potential for defluoridation was found to be 2.0 volts per cell pair, achieving high current efficiency (87.0%) and low power usage (1.2 kWh/kg) (Sharma et al., 2018).

Two approaches were used in ED operations on geothermal fluids. In the first approach, the pH of the geothermal fluids was adjusted to 7–7.5 before ED operation to prevent the deposition of bivalent salts in the concentrated chamber. The second approach involved pretreating geothermal water to precipitate calcium ions. Both approaches were effective in extracting fluoride from geothermal water, but the first approach was considered more cost-effective and technically straightforward due to its less expensive chemical usage (Arda et al., 2009).

One significant drawback, particularly concerning the generation of drinking water, is that uncharged elements such as microbes or organic pollutants remain unremoved during

electrodialysis; only ions are eliminated. Additionally, the relatively high energy consumption associated with electrodialysis, especially when processing solutions with high concentrations of salt, poses another challenge. Similarly, in cases where very low salt concentrations must be achieved in the diluate, the high investment costs become prohibitive due to the low limiting current density, which necessitates a wide membrane area. Consequently, only a certain range of feed water salt content and the required product water quality allow for the cost-effective application of electrodialysis in water desalination (Strathmann, 2010).

When the salinity ranges between 7 and 9 ppt, Electrodialysis (ED) proves to be a more economically viable option compared to reverse osmosis (RO). Specifically, for ED, the product water price stands at 0.384 \$/m³ when dealing with a salinity of 5 ppt, such as groundwater salinity. However, when the feed salinity increases to 35 ppt, as in the case of seawater, the product water cost escalates to 1.532 \$/m³. Furthermore, even with highly salinized water below 100 ppt, ED offers a cost-effective solution for desalination, often costing less than \$5/m³ (Generous et al., 2021).

3.2. Membrane distillation

Membrane distillation (MD) is recognized as an effective water treatment technology, particularly for highly contaminated water sources. In this process, the pressure created by the temperature difference between the permeate and feed of the membrane allows only water vapor to pass through micro-porous hydrophobic membranes. Various factors influence the MD process's rejection and flow, including the properties of the water being fed, operating variables, and membrane characteristics.

The chemical and manufacturing processes of MD membranes play a crucial role in their behavior, impacting permeate water quality and rejection efficiency. Scientists have studied the impact of membrane characteristics on MD efficiency, including the effects of membrane pores and permeability on energy and thermal efficiency, the influence of different membrane variables on water production costs, the impact of membrane shape on distillate performance, and the effect of surface permeability on MD efficiency (Damtie et al., 2018).

In the context of fluoride (FL) removal using MD, studies have shown that a self-made polyvinylidene fluoride membrane exhibited an increased rejection rate for inorganic salt solutes. The distillate water temperature was 20°C, and the feed solution was at 80°C to achieve the optimum permeate flow. Rejection rates in FL and permeate flow were not significantly affected by the feed level. However, the direct use of fresh groundwater as feed led to the precipitation of calcium carbonate, which reduced module performance by contaminating the

membrane's surface and blocking the hollow fiber entrances with an increasing concentration factor (CF). Acidification of the feed mitigated this effect, demonstrating stable permeate flow and distillate quality up to a CF of 5.0. The formation of calcium fluoride deposits on the membrane interface was identified as the primary cause of the progressive drop in membrane module performance when the feed remained concentrated (Damtie et al., 2019).

Several studies have investigated the effectiveness of MD in removing salts, arsenic, FL, and uranium from water with FL concentrations ranging from 1 to 30 ppm. The MD process demonstrated the ability to remove over 99% of these pollutants, producing high-quality permeate suitable for various applications. MD has been considered a feasible method for restoring drinking water from a range of degraded groundwater sources, leveraging low-quality heat sources (Yarlagadda et al., 2011).

Studies have indicated that FL removal effectiveness is enhanced by high feed temperatures, low downstream pressures, low initial feed levels, and high feed flow rates. The temperature and concentration polarizations in the feed boundary layer were identified as having a significant impact on mass movement. Increasing feed flow velocity has been identified as a key strategy to mitigate the effects of temperature and concentration polarization. Under specific conditions, such as a feed temperature of 333 K, a vacuum of 200 mmHg, feed flow rate of 90 l/h, and feed level of 20 ppm, the highest flux achieved was 16.98 kg/m² h, with FL removal ranging from 97.8 to 99.7% (Singh et al., 2013).

The primary drawback of this approach is the conductive heat loss that occurs during the operation, leading to a reduction in pressure and temperature gradient across the membrane. The impact of utilizing waste heat on the profitability of membrane distillation (MD) technology in treating generated water is significant. According to the findings of Tavakkoli et al., (2017), the overall cost of treating generated water with MD amounts to \$5.70/m³ feed. However, when MD is integrated with a waste heat source, the cost decreases dramatically to \$0.74/m³ feed.

3.3. Reverse osmosis/nanofiltration

Reverse osmosis (RO) and nanofiltration (NF) (Fatehizadeh et al., 2020) are the effective techniques for fluoride (FL) removal, exhibiting excellent FL retention compared to traditional methods like adsorption and precipitation (Taheri et al., 2020). Three mechanisms contribute to FL transport across the membrane in NF/RO processes: electrical migration, convection, and diffusion. Generally, FL retention in NF/RO is influenced by electrical and steric factors (Fatehizadeh et al., 2018). Several factors, including feed compositions, membrane properties, and operating conditions, can favor one mechanism over another (Shen and Schäfer, 2015).

Studies have evaluated FL and naturally occurring arsenic removal from groundwater using the RO method, resulting in a significant reduction in FL concentration by almost 60% (Schneiter and Middlebrooks, 1983). FL retention in NF/RO is influenced by charge exclusion, membrane type, and pH, with retention being challenging at neutral and low pH levels. FL retention often exceeds 70% above pH 7 (Richards et al., 2010). Performance metrics of various NF/RO membrane modules, including permeate production, FL retention, and specific energy usage, have been compared. The NF90 membrane demonstrated the highest overall efficiency with an average specific energy consumption of 1.6 kWh/m³ over the course of a solar day. This resulted in the production of 1582 L of pure water for consumption (Shen et al., 2016).

The pH of the solution significantly impacts FL retention, with higher pH leading to greater FL retention due to the increased negative electrical charge on the membrane. The relationship between ion retention and FL concentration varies between RO and NF90 membranes. It is essential to consider steric impact, diffusion, and membrane electric charge when dealing with ionic species to understand rejection processes fully. The preservation of FL processes using NF/RO membranes concerning single-factor effects is not entirely understood, and further research is needed to deepen the understanding of medium composition and the interactions between ions and the membrane. This includes examining different salt combinations and understanding the multi-factor effect on FL retention (Bejaoui et al., 2014).

RO/NF systems are particularly susceptible to early membrane fouling due to their extremely small pore diameters compared to other membrane filtering technologies. This fouling can lead to shortened unit service life, reduced permeate quality, and increased operating expenses if proper pretreatment and process monitoring procedures are not implemented. The NF-RO hybrid system produced about 1.6 times less water (Rs. 99.81/m³) compared to the NF system (Rs. 158.46/m³), and 4 times less water than the RO system used alone (Rs. 400.49/m³). Moreover, by enhancing the capacity of the membrane system and providing additional subsidies for solar systems to consumers, this cost could be further reduced (Garg et al., 2014).

3.4. Forward osmosis

Forward Osmosis (FO) can effectively remove ions from wastewater using the osmotic pressure gradient as its driving force. The key to enhancing FO separation efficiency lies in the use of a high-performing membrane. In an experiment, a mass-to-weight ratio of 0.5 for chitosan and tannic acid, with a co-deposition time of 60 minutes, was found to be optimal. When a thin-film composite with this mass ratio was used to treat wastewater containing 300 ppm of FL, the water flow reached 22.3 LMH after 60 minutes of co-deposition, and FL

rejection was 99.93%. In a FO test on effluent containing FL from a semiconductor manufacturer, the membrane's water flow was 49.3% higher than that of the commercialized Aquaporin Inside FO membrane (Zhang et al., 2024).

The addition of an interlayer improved the membrane's hydrophilic properties and increased its negative charge at the surface, making it easier for water molecules to be transported while retaining FL ions. Three-layer film composite/3-aminopropyl triethoxysilane - cellulose nanocrystal and three-layer film composite/boric acid - cellulose nanocrystal membranes showed higher water fluxes than three-layer film composite/cellulose nanocrystal membranes by 5.9% and 13.0%, respectively, in FO tests. The water flux of the three-layer film composite/cellulose nanocrystal membrane was increased by 46.4% compared to the original three-layer film composite membrane. Three-layer film composite/boric acid - cellulose nanocrystal and three-layer film composite/APTES - cellulose nanocrystal could retain 97.0% and 97.6% of their FL ions, respectively. Molecular modeling of adsorption illustrated that the boric acid - cellulose nanocrystal interlayer was more effective than the cellulose nanocrystal and 3-aminopropyl triethoxysilane - cellulose nanocrystal interlayers in adsorbing water molecules (Huang et al., 2023). This work contributes to the development of a high-performing three-layer film composite-FO membrane for treating FL-containing effluent.

It is crucial to understand that the end result of the forward osmosis (FO) process is a diluted draw solution rather than clean water, unlike most other water purification procedures. To recover the draw solution and provide clean water, FO typically needs to be paired with a second separation stage, usually Reverse Osmosis (RO). Therefore, FO can only be considered genuinely energy-efficient if the draw solution does not require regeneration. There has been considerable misconception regarding FO's power usage because producing clean water often necessitates an additional process. In fact, studies have shown that for saltwater desalination, FO-RO systems consume more power than a standalone RO unit (Mohammadifakhr et al., 2020).

3.5. Adsorption

One critical method for removing fluoride (FL) from water-based solutions is adsorption (AD), where the choice of adsorbent plays a crucial role in the viability of AD methods. The effectiveness of AD methods is generally limited at fluoride concentrations below 2 ppm, and a wide range of materials has been evaluated at fluoride concentrations larger than 2 ppm, with most showing little ability to remove fluoride when concentrations are reduced to less than 2 ppm (Fan et al., 2003). To assess the suitability of readily available activated charcoal for

cleaning up fluoride-contaminated water, batch AD tests were conducted. The impacts of starting fluoride concentration, adsorbent dosage, pH, mixing rate, contact duration, and other crucial AD factors on fluoride removal were investigated and optimized. A sorbent dosage of 2.0 g/100 mL was determined to be ideal, equilibrium was reached in 2 hours, and increased AD was observed at pH 2. Under ideal conditions, a maximum fluoride elimination of 94% was achieved. Langmuir and Freundlich isotherms were plotted, and kinetic constants were calculated (Tembhurkar and Dongre, 2006).

Thermally activated carbon based on *Cynodon dactylon* was studied for its ability to remove fluorocarbons (FL) from aqueous solutions, and the system adhered to both the Langmuir AD isotherm and the Redlich–Peterson isotherm model. The enthalpy change for the AD reaction was determined to be +8.725 kJ/mol, and the entropy change was +0.033 J/mol K. The AD process was endothermic, and under optimal conditions (maintaining 3.0 ppm fluoride concentration and 1.25 g of adsorbent at neutral pH), the best defluoridation occurred at 105 minutes, resulting in an 83.77% success rate (Alagumuthu et al., 2010).

The pH of the water is a significant factor influencing fluoride AD (FL-AD). FL AD typically rises from an acidic to about a neutral pH and then decreases when pH rises. The type and quantity of other ions in processed water, as well as the adsorbent's selectivity for fluoride ions, are additional crucial factors impacting FL-AD, as they can occupy adsorbents' active locations and lower their estimated AD capacity (Habuda-Stanić et al., 2014). When acidic alumina was used to treat natural underground water without adjusting the pH, increased efficiency was observed. The various exchange locations and anion exchange behaviors were attributed to variations in the AD characteristics. More chlorine was replaced with FL in the presence of acidic alumina, and the initial fluoride concentration influenced the AD/release of OH⁻. According to Gon et al. (2012), the majority of exchange sites for basic alumina were supplied by OH⁻, and FL was eliminated via the interaction between FL and OH⁻. Adsorbents such as china clay have been used for fluoride removal (FLR) from wastewater (WW) at varying pH levels, temperatures, and concentrations. For the current system, the AD isotherms and kinetics at various temperatures have been studied. Low concentration, extreme temperatures, and an acidic pH promote fluoride elimination. The highest AD of fluoride in the relevant pH range is caused by the alumina component of china clay (Chaturvedi et al., 1988).

According to the current study, materials like coal and nirmali seeds are ineffective (removing 6 to 8% of the material); in contrast, kaolinite clay removed 18.2% of the material. Under ideal system conditions, the maximum removal capacities were shown by bentonite and charcoal, at 46% and 38%, respectively. According to Simurali et al. (1998), chemical prior

treatment of charfines did not improve fluoride removal (FLR) from groundwater (GW). The thermally processed lateritic soils employed in this investigation had a fluoride elimination capacity ranging from 22 to 47 mmol/kg. For RGS burned at 500°C, the maximum capacity of 47 mmol/kg was achieved. The results show a substantial relationship between the soil's gibbsite concentration and its ability to remove fluoride. This suggests that the active ingredient in lateritic soils that significantly affects the capacity to remove fluoride is debris (Gomoro et al., 2012). Physical adsorption presents several drawbacks, including the costly nature of the adsorbent, challenges in separating it from the dye, and limited surface area. Consequently, significant effort has been directed toward overcoming these limitations and developing highly efficient yet cost-effective adsorbents for dye treatment (Moosavi et al., 2020).

4. Advanced techniques to remove fluoride

To enhance fluoride removal (FLR) in groundwater (GW) treatment, various methods have been implemented, including ion exchange (IE), precipitation by chemicals, membrane separation processes (MSP), the coagulation process, adsorption (AD), and both phyto and bio-remediation. However, many of these techniques have certain drawbacks. For instance, IE is limited by the expense of the resin, the need for waste disposal due to regeneration and subsequent costs, and its lack of sufficient selectivity, making the process inefficient and expensive. Similarly, membrane-based methods face challenges such as high costs for membrane purchase and operation, as well as the need for concentrated disposal, requiring post-treatment of water and rendering the procedure relatively unprofitable.

On the other hand, coagulation is considered a cost-effective defluoridation method but requires large dosages, leading to high residual levels, including hazardous aluminum, and generating a significant volume of sludge. Lime-based procedures, while also cost-effective, face issues such as the limited solubility of resultant calcium hydroxide, hindering sufficient FLR from the fluids (Tolkou et al. 2021). In contrast, certain advanced methods, like ceramic microfiltration and electrodeionization membrane capacitive deionization, show great promise among these technologies due to their simplicity of use, affordability, improved selectivity, and energy efficiency. Table 5 provides an overview of the operational status and advanced methods for FLR.

4.1. Ceramic membrane-based microfiltration

Presently, the utilization of combined microfiltration (MF) membrane filters is becoming increasingly popular due to their practicality, process reliability, economic availability, flexibility, relative insensitivity when treating wastewater from different industries, raw water treatment, and lower operating expenses (Hakami et al., 2020). Membranes can save capital

costs, require less maintenance, and demand fewer chemical uses when they are carefully planned, built, and used. Comparing ceramic microfiltration (CMF) membranes to biological and polymer membranes, the former are increasingly losing use in drinking water and wastewater treatment (WWT) sectors owing to their resilience to harsh operating conditions and cleaning procedures (Bottino et al., 2001).

Notwithstanding their numerous benefits, these types of membranes are prone to a clogging process that can negatively impact the smooth and uninterrupted functioning of the plant during all hours of operation. Fouling is a complicated multifactorial phenomenon that is mostly caused by chemical and microbiological agents, such as suspended particles, naturally occurring particulates, bacteria, and heavy metals (Song et al., 2022). In order to avoid the decline of separation efficiency of MF membranes over time, the manufacturing sector currently employs a number of strategies, including chemical-based cleaning protocols, turbulent conditions promoters, and backwashing with air or liquids. These techniques are primarily focused on early detection and treatment (Hua et al., 2007).

The use of CMF membranes in routine WWT is becoming more and more popular. When utilized properly, membranes can provide great productivity for retaining contaminants and offer cheap operating costs compared to other methods, as they don't require chemical additives or water phase shift. Ceramic membranes (CM) are highly resistant to harsh cleaning procedures and operational environments, resulting in longer lifespans and extremely effective filtering performance (Belgada et al., 2021). Dolomite, fly ash, and kaolin were used to create inexpensive CM. It was discovered that the optimum sintering temperature for both fly ash and kaolin-based membranes was 900 °C. A 20% dolomite addition provided all membranes (28–51%) with enough porosity. As the kaolin concentration rose, the pore size shrank while its porousness, power, and durability increased. Depending on the pore properties (mean pore diameter 0.62 µm and porosity 46.3%), the sintered composite with a 75% kaolin content (M75) was found to be the most suitable membrane among the others (Malik et al., 2020).

The main contributing component to irreversible clogging on the CMF was turbidity. According to Sheikhi et al. (2019), there was a quick occurrence of irreversible clogging when the turbidity increased. This may be attributed to tiny particles penetrating the membrane and causing standard pore blockage. Because this results in the most permanent clogging, high turbidity feed water was used for the coagulation study. Poly-aluminum chloride was, therefore, chosen as the ideal coagulant. The removal effectiveness of the polymer and CMF was not significantly different, but the CMF had a decreased propensity for fouling of the membrane and a superior water flow. Furthermore, using a coagulation procedure as a

pretreatment boosted efficiency. As a result, CMF membranes are more effective in treating water with high turbidity (Park et al., 2020).

By changing the proportions of binders like sodium metasilicate, boric acid, and carbonate, and kaolin, five distinct membrane compositions were selected. The permeability of sintered membranes ranged from 21.5% to 34.5%. The size of pores was found to decrease while flexural strength improved in response to increases in binder percentage and decreases in kaolin percentage, correspondingly. In the membrane with increased water flux, the average permeability, pore dimension, and bending strength were 34.5%, 2.28 μm , and 7.1 MPa, correspondingly. The greatest permeability measured was $6.12 \times 10^{-9} \text{ m}/(\text{Pa}\cdot\text{s})$ (Agarwalla and Mohanty, 2022). For fluoride removal (FLR), not much research has been done. Further research on this FLR will be carried out by CMF in the future.

Ceramic membranes are known for their excellent antifouling properties and ease of cleaning. However, their high costs have limited their widespread industrial use. Additionally, the water permeability of a typical ceramic membrane is lower due to its thicker rejection layer compared to polymeric thin film composite polyamide membranes. Despite these challenges, ongoing efforts in cost reduction and performance improvement are expected to enhance the competitiveness of ceramic membranes in the future (Sarp and Hilal, 2018).

4.1.1. Importance of ceramic membrane

Ceramic membranes are superior to most polymer and inorganic membranes in many ways. They have exceptionally high levels of mechanical, thermal, chemical, and physical stability. They don't have to worry about membrane compaction or swelling at high temperatures, pH extremes, or high working pressures of up to 10 bar. Because of this, these membranes are employed in several applications that are not compatible with polymeric or other inorganic membranes. Their lengthy working life and exceptional separation qualities. They are frequently far more advantageous than alternative separation methods in terms of the environment. Ceramic filtering is a gentle, very selective method that doesn't use phase transition. They are proficient at backflushing and steam sterilization. Their strong resistance to abrasion is what makes them unique (Amin et al., 2016).

When considering the cleaning effectiveness, fouling tendency, permeate flux, usable life duration, and environmental effects, ceramic membranes outperform polymeric ones. Due to these advantages, the market for ceramic membranes is expanding, with large-scale installations found in the United States, Singapore, Japan, South Korea, the Netherlands, the United Kingdom, and Switzerland. According to numerous techno-economic studies based on the life cycle of full-scale plants, ceramic membrane plants are more cost-competitive than

polymeric membrane plants. The use of ceramic membranes in innovative (photo)catalytic processes has attracted interest due to their resistance to chemical oxidants and reactive species. The cost of ceramic membranes is expected to continue declining due to rising demand, enabling their widespread use, particularly in developing nations (Muhammad et al., 2009).

4.2. Electrodeionization

Highly pure water has been successfully produced using electrodeionization (EDI), a technique that integrates the benefits of electro-dialysis (ED) and ion exchange (IE) procedures (Arar et al., 2014). EDI has found a variety of new and innovative applications, such as wastewater treatment (WWT), the purification and separation of biological products, and other promising industries. Its capacity to undertake a continuous and thorough deionization (DI) process without renewing chemicals has been a key factor in its success (Rathi and Kumar, 2020). Due to increased interest, there is a need for the improvement of EDI stack building and configuration to achieve greater performance. Additionally, several investigations have been conducted to enhance the understanding of the ion transfer process inside the EDI system (Hakim et al., 2020). Thus far, EDI has proven to be a highly effective and sustainable technique for segregation, concentration, and cleansing. Ongoing development of new materials aims to advance and mature this field of science, which could have significant positive effects on the environment and the global economy (Alvarado and Chen, 2014). The environmentally friendly nature of EDI is likely what makes it commercially viable. According to Wood et al. (2010), it eliminates the wastewater associated with ion exchange resin (IER) regeneration by using electricity instead of the hazardous chemicals often required for IER regeneration. Fig. 4 shows schematic representation of EDI used to remove fluoride from water.

In EDI technique, the unit is constructed for continuous production of fluoride-free water, and investigations are conducted using a laboratory-designed unit. There are two main modes of operation for the EDI process: improved transfer and electro-regeneration. The current effectiveness of the EDI system drops in its second phase due to water dissociation. The processes employed to transform the styrene–divinylbenzene-based interpolymer film into the cation-exchange membrane (CEM) and anion-exchange membrane (AEM) used in this investigation are sulfonation and chloromethylation, respectively. An amphoteric resin, created through radical suspension polymerization, exhibits a 5.7% water uptake and an ion exchange (IE) capacity of 4.57 mequiv/g. Both IER and IEM are characterized using chemical and mechanical characteristics. The IEM and IER exhibit exceptional mechanical stability along with outstanding electrochemical characteristics. The impact of water composition on the selectivity of fluoride removal (FLR) and the energy use of the EDI unit was demonstrated by

the FLR from three different concentrations of tap water sources. In several sets of experiments conducted during EDI, different potentials—9, 12, and 15 V/cell pair—are used. It was discovered that the 12 V/cell pair was the most effective, exhibiting a reduced electrical consumption of 4.6 kWh/kg and a 37% current effectiveness (Gahlot et al., 2015). There hasn't been much research done on FLR. Further study on FLR using EDI will be conducted in the future.

When the hardness of water concentration exceeds 1, the presence of calcium carbonate in water can lead to scaling in the EDI system, rendering it unsuitable for use. In such cases, pretreatment with purification is necessary. The RO membrane facilitates the easy passage of carbon dioxide, which dissociates and raises the water's conductivity. Consequently, any ionic species resulting from the carbon dioxide gas will reduce the exit resistivity of the water produced by the EDI system (Kumar and Rathi, 2024)

4.3. Membrane capacitive deionization

The method of removing ions from wastewater (WW) by membrane capacitive deionization (MCDI) involves creating voltage changes between two porous electrodes set up opposite each other and in front of ion-exchange membranes (IEM). A product stream with a lower concentration of salt is produced as a result of the ions being adsorbed in the electrodes by the applied voltage (Biesheuvel and Van der Wal, 2010). The IEM provide two benefits to the process: first, they prevent co-ions from departing the electrodes, which increases the method's effectiveness in removing salt; secondly, when an opposite voltage is employed during ion discharge, counterions can be more completely flushed from the electrode region, which increases the force that drives for ion elimination in the subsequent cycle. Here, we provide the output of a pilot plant study that examined the elimination of salt in MCDI as a result of flow rate and intake ionic strength (Epshtein et al., 2020). The rate of flow is momentarily zero, and the voltage sign is inverted during the next phase of ion release. This "stop-flow" operation mode produces a narrow and concentrated product stream (Zhao et al., 2013). The schematic illustration of membrane capacitive deionization to extract fluoride (FL) from water is shown in Figure 5.

Based on the same operating principle as capacitive deionization (CDI), MCDI is a version of CDI that incorporates IEM into the electrodes of the CDI cells (Islam et al., 2022). Because the MCDI cell has an anion-exchange membrane (AEM) onto the anode surface and a cation-exchange membrane (CEM) on the cathode surface, co-ions may be successfully prevented from adhering to the electrodes, ensuring that the opposing ions are adsorbed by the two electrodes effectively without the adverse effects of co-ions. As a result, the removal

effectiveness of MCDI is clearly higher than that of CDI (Zhao et al., 2013a). When the electrodes reach a specific capacity for adsorption, the voltage is lowered or even inverted, causing the electrodes to release ions and creating a concentrated salt solution in the spacer channel. The resulting solution is subsequently flushed out, allowing the cycle to begin again. The placement of IEM in front of every porous electrode has the benefit of allowing for ion desorption at reversed voltage while simultaneously preventing co-ions from exiting the electrode area during ion adsorption (Folaranmi et al., 2020). The system's ability to remove salt every cycle is greatly increased by these impacts. A product of desalinated water with a variable salt content is produced using the traditional operating mode of MCDI at a fixed cell voltage (Zhao et al., 2012).

MCDI introduces a number of resistances, including the membrane's internal and interfacial resistances and the resistance of contact among the membrane and carbon electrodes. In recent years, integrated-MCDI has demonstrated great advantages to improve the electrode's counter-ion precision and reduce opposition, which improves the adsorption rate and lowers the use of energy (Wu et al., 2022). This is accomplished through the combination of the "membrane" with the electrode made of carbon without the need for a standalone IEM. The dynamic procedure of removing both FL in CDI cells was found to be correctly described by a simplified 1-D transportation model for dual anions, which was established over a variety of established operating parameters (Kumar et al., 2023). The framework was expanded to explain the elimination of nitrates from saltwater groundwaters and was found to function well, based on its capacity to represent fluoride removal. Therefore, the ion removal characterization approach appears valid, at least in batch investigations, and it may help optimize the architecture and operating conditions to enable optimum elimination of identified ion species even in the face of high background salt concentrations (Tang et al., 2015).

For the adsorption of fluoride (FL), the related CDI cell exhibits great cycle reliability and specificity. Using a 3-cell CDI stack to remove fluoride from groundwater containing FL, the electrode achieves an excellent elimination rate of 90.5%, resulting in a wastewater level of just 0.32 ppm, much below the WHO's acceptable limit. This three-dimensional reduced graphene oxide-based CDI system, improved by lanthanum-doped hydroxyapatite, shows great promise for purifying high fluoride groundwater (Wang et al., 2022). The economics of CDI suffer from its lower water recovery and longer electrode discharge times, negatively impacting its viability. Additionally, the poor recovery ratio not only increases the cost of brine disposal but also contributes to environmental pollution. Furthermore, capacitive deionization's inability to remove contaminants other than ions, such as bacteria, viruses, and other impurities, presents

another significant disadvantage (Sharan et al. 2021). Fig. 6 shows the limitation of different techniques for the fluoride removal process. Moreover, the SDGs are interconnected and aim to create a more inclusive, equitable, and sustainable world by the year 2030. Governments, businesses, and individuals around the world are encouraged to contribute to the achievement of these goals, fostering collaboration and collective action to address global challenges and improve the well-being of people and the planet.

5. Membrane -based fluoride removal- Life cycle assessment

The concepts of life cycle thinking (LCT) and life cycle assessment (LCA) are becoming increasingly important for facilitating sustainable transitions. The goal of comprehensive coverage of ecological variables, recognition of areas of greatest risk, potential trade-offs, and responsibility shifting within life cycle phases or impact groups are all areas in which Life Cycle Assessment (LCA) is significant. When decision assistance is required in the policy area, these characteristics are equally pertinent (Sala et al., 2021). According to a recent position paper released by the Life Cycle Initiative, LCA methodology may be used to determine the most likely sustainable economic methods and solutions for enhancing the ecological sustainability of society's habits of consumption and manufacturing (Rigamonti and Mancini, 2021).

Although they often don't provide any additional information, LCA regularly acknowledges that time concerns may compromise the accuracy of their conclusion (Koch et al., 2020). These are significant contributors to errors and have an impact on one another, therefore it's critical to understand them well, fill in definitional gaps, systematize temporal problems, demonstrate how they're interdependent, and offer potential remedies (Lueddeckens et al., 2020). It was discovered throughout the study that usually, LCA is used too late in the procedure to make a big difference in the final product. According to Li et al. (2019), there are similar difficulties when combining life cycle costing or modelling of buildings with life cycle assessment (LCA). Although parametric techniques are promising for design development, more verification and regionalization of tools and algorithms are needed before they can be applied across the industry (Nakhate et al., 2020). Without conducting a thorough life-cycle assessment, life-cycle thinking may be implemented through the use of benchmarks, goal values, and other pre-populated data (Roberts et al., 2020).

Using the LCA methodology, decision-makers can make well-informed choices regarding: i) objective and subject matter definition; ii) operational unit and boundaries of the system selection; iii) factors that should be included and their sources of information for stocks; iv)

influence evaluation techniques selection; and v) an efficient method for interpreting information and interaction to decision-makers (Corominas et al., 2020). When comparing the harmful impacts of the MCDI system on the well-being of living things and the surroundings to those of the membrane-based system, it becomes clear that the MCDI method is a cost-effective and ecologically responsible way to reuse wastewater. It is evident from the SimaPro LCA program's conclusions that the membrane system has significant environmental disadvantages. The average difference in damages categories between the two systems is 5641 times in favour of the membrane-based system, indicating that the MCDI system is significantly more environmentally friendly overall. When comparing the two systems that do the same task, it is obvious that MCDI should be chosen based only on the way it protects the environment (Cetinkaya and Bilgili, 2019).

6. Case studies in fluoride removal

Malakootian et al, (2015) was conducted experimental study between May and October of 2013, using both artificial and real water samples obtained from the drinking water of the Koohbanan region in the northwest of Kerman Province, Iran. Bauxite from the Jajarm bauxite mine was utilized in the investigation. The findings revealed that, in the presence of other competing anions in the water, the fluoride (F) concentration could meet the WHO guideline when 15 g of bauxite per liter (g bauxite/L) was added to Koohbanan water with a 90-minute contact time, resulting in a removal efficiency of 77.45% for a 6 mg/L F concentration. This removal efficiency was higher compared to that achieved with the actual Koohbanan water sample, which exhibited a removal efficiency of 53.33% for a 2.46 mg/L F concentration. It was found that the typical defluoridation capacity of Jajarm bauxite was approximately 0.087 mg F/g in Koohbanan water and roughly 0.31 mg F/g in deionized water. This suggests that bauxite can effectively remove fluoride from contaminated water. Moreover, bauxite is readily available in many regions of the world and the defluoridation process using bauxite is simple, cost-effective, and efficient. Therefore, bauxite holds promise as a viable solution for fluoride removal from polluted water sources.

7. Conclusions and future perspectives

The health effects of fluoridated water are significant. Accordingly, the WHO limits the maximum amount of Fluoride Concentration (FLC) that can be present in freshwater to 1.5 ppm. In addition to drinking water, fluoride contamination affects various types of wastewater (WW). Untreated fluoride-enriched WW can have a hazardous effect on aquatic life and environmental health. To eliminate excessive fluoride from water and WW, various techniques have been employed, including adsorption (AD), ion exchange (IE), precipitation by chemicals,

Electrodialysis (ED), the electrocoagulation process, RO and NF. Due to the ease of usage and inexpensive operating costs of the ED process, many companies and organizations have focused on it to eliminate excessive fluoride from water. Even though several evaluations have tried to maximize ED performance for fluoride removal from WW, scaling, membrane fouling, and permselectivity remain issues with ED technology.

Under the real pressure and temperature gradients, the NF membrane may provide water production that is over ten times greater than that of the MD membrane. With single-factor impacts, the preservation of fluoride activities utilizing both NF and RO membranes is not well studied. Future research should be done to better comprehend the impact of medium composition and clarify interactions between ions and the membrane's surface. This will include examining different salt combinations and the multifactor impact that they have on the retention of fluoride. Due to fluoride removal's eco-friendly and reusable qualities, as well as the fact that these materials are accessible in large quantities and at competitive rates, a variety of industrial goods and by-products have been investigated. While those adsorbents had efficient fluoride absorption and high AD capacities, the majority of them were very dependent on pH and often reported difficulties with challenging adsorbent renewal.

Using ceramic membranes (CM) has several benefits, including robustness, extended lifespan, ease of cleaning, longevity, and operating in harsh conditions. Institutions and membrane producers have conducted a great deal of research in the past several years to improve the performance of present CM. In order to enhance product design and ensure long-term, sustainable operation, further study is needed on several topics, including low-cost ceramic materials, the progression of fouling (particularly biofouling), and the effectiveness of cleaning techniques. For many industrial scale applications, CM will probably become an economically viable alternative to polymeric membranes if new production techniques are introduced to lower the investment cost of CM. To ascertain if installing a large-scale ceramic membrane in a possible industrial operation may be a feasible and lucrative endeavour, it is also necessary to construct prediction models that will enable scaling up from small scales to industrial scale applications and to conduct an entire life cycle cost study.

The main goal was to increase the system's hydrodynamics by encouraging agitation in the membrane boundary layer. Less complexity, more dependability, reduced labor and component costs, and smaller footprints are the historical trends in electrodeionization (EDI) systems. The primary operational expenses associated with EDI installations are linked to power consumption, wastewater treatment (WWT), lab control, and EDI modules repair. These costs are closely correlated with the feed water's composition and system output.

Furthermore, it is highly recommended to combine several defluoridation techniques (both membrane processes and traditional procedures) in order to maximise the advantages of each technique. The main problem with integrated-membrane capacitive deionization (MCDI) is the electrode fabrications, which may be split into three types based on the electrode setup: membranes coating, polymer mixing, and modification of the surface. Electrodes covered with membranes have reduced internal resistance and improved contact with carbon electrodes. By exploiting the unique designs of integrated membrane electrodes, MCDI can not only accomplish improved desalination but also offers substantial potentials in anti-organic pollutants and ion-selective elimination like fluoride removal. More research has been conducted on the application of defluoridation procedures for brackish and natural groundwater sources compared to wastewater. For instance, see Aminabhavi et al., US patents on ED membranes used in defluoridation (US Pat 2004/0198849 A1, Oct. 7, 2004; US Pat 7,544.278 B2, Jun. 9, 2009). Further research is necessary to assess how effectively various membrane technologies can handle industrial wastewaters with high fluoridation levels. Their impact on life cycle assessment outcomes is frequently fundamental, thus further research is necessary to have a deeper understanding of this issue and potential remedies.

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Tables

Table 1: Physical and chemical properties of Fluoride

Element Name	Fluorine
Appearance	White or Colourless
Taste	Bitter
Oduor	Odourless
Category	Weak base
Nature	Corrosive
Molecular weight	18.99 g/mol
Symbol	F
Atomic Number	9
Characteristics	Oxidizing agent
Oxidation state	-1
Melting Point	-219.61 °C
Solubility at 25 °C	1.69 mg/L
Chemical Classes	Inorganic
Permissible Exposure Limit	2.5 mg/m ³
Carcinogenic Classes	Group 3

Table 2: Concentration of Fluoride in various food

Different Product	Fluoride concentration	Reference
Beverages	0.93 µg/g	Jackson et al., 2002
Beverages	0.023 µg/g	Cantoral et al., 2019
Black tea	58 mg/kg	Lu et al., 2004
Black tea	1.41 mg/L	Cao et al., 2006
Black tea bags	6.01 mg/L	Cao et al., 2006
Black tea granules	2.44 mg/L	Cao et al., 2006
Bottled drinking waters	0.04 µg/g	Zohoori and Maguire, 2016
Breakfast cereals	1.86 µg/g	Cardoso et al., 2003
Cereals	1.16 µg/g	Cantoral et al., 2019
Chocolate	7.9 µg/g	Buzalaf et al., 2004

Corn	2.7 µg/g	Buzalaf et al., 2004
Dairy products	0.27 µg/g	Cantoral et al., 2019
Drinking water	0.16 to 0.39 ppm	Miya and Jha, 2020
Drinking water bottles	1.2 mg/L	Somasundaram et al., 2015
Eggs	0.023 µg/g	Cantoral et al., 2019
Fast food	1.33 µg/g	Cantoral et al., 2019
Fats and oils	0.62 µg/g	Jackson et al., 2002
Fish	1.48 µg/g	Zohoori and Maguire, 2016
Freshwater	0.3 mg/L	Kanduti et al., 2016
Fruits	0.037 µg/g	Cantoral et al., 2019
Fruits	95 mg/kg	Shu et al., 2003
Grain products	1.36 µg/g	Jackson et al., 2002
Granular black tea	139–223 mg/kg	Cao et al., 2004
Green tea	29 mg/kg	Lu et al., 2004
Green tea	232 mg/kg	Shu et al., 2003
Jasmine tea	76 mg/kg	Lu et al., 2004
Legumes	0.84 µg/g	Cantoral et al., 2019
Meats	3.75 µg/g	Cantoral et al., 2019
Mouthwash	1305 mg/L	Švarc-Gajić et al., 2013
Oat	0.2 µg/g	Buzalaf et al., 2004
Old leaves and twigs	613 mg/kg	Shu et al., 2003
Rice	2.1 µg/g	Buzalaf et al., 2004
Seafood	3.71 µg/g	Cantoral et al., 2019
Seawater	1.5 mg/L	Kanduti et al., 2016
Snacks	0.40 µg/g	Cardoso et al., 2003
Soil	150 to 400 mg/kg	Shu et al., 2003
Soil	320 to 371 mg/kg	Wong et al., 2003
Stick-shaped black tea	96.9 to 148 mg/kg	Cao et al., 2004
Sugars and sweets	0.60 µg/g	Jackson et al., 2002
Sweets	0.87 µg/g	Cantoral et al., 2019
Tea and coffee samples	0.011 to 0.084 ppm	Miya and Jha, 2020
Toothpaste	1000 mg/L	Thakkar et al., 2015

Toothpaste	1479 mg/kg	Švarc-Gajić et al., 2013
Toothpaste	1500 mg/L	Borremans et al., 2008
Toothpaste	0.026 to 0.75 ppm	Miya and Jha, 2020
Toothpastes	1216 mg/L	Sebastian and Siddanna, 2015
Vegetable dishes	0.12 µg/g	Zohoori and Maguire, 2016
Wafer biscuit	0.4 µg/g	Buzalaf et al., 2004

Table 3: Techniques to measure Fluoride

Technique	Sample	Detection limit	Reference
Colorimetric methods	Liquid	1 ppm	Mehra and Lambert, 1973
Combined Ion-selective electrode	Liquid	10 ⁻⁶ mol/L	Bratovic and Odobasic, 2011
Gas Chromatography Method	Liquid	8 to 2000 ppm	Sarathchandraprakash et al., 2014
Gas chromatography/mass spectrometry	Human blood and urine	0.5 ppm	Dhillon et al., 2016
Gas chromatography–Flame ionization detection	Liquid	0.02 µmole/L	Koc et al., 2011
Gas chromatography–Flame ionization detection & electron capture detection	Liquid	0.3 µmole/L	Koc et al., 2011
Gas chromatography–mass spectrometry	Plasma	11 µg/L	Kwon et al., 2014
Gas chromatography–mass spectrometry	Urine	7 µg/L	Kwon et al., 2014
Gas chromatography–mass spectrometry	Liquid	0.003 µmole/L	Koc et al., 2011
Inductively coupled plasma mass spectrometry (ICPMS)	Aerosol	0.1 ng/mL	Dhillon et al., 2016

ICPMS	Liquid	0.1 ng/mL	Dhillon et al., 2016
ICPMS	Solid	0.1 ng/mL	Dhillon et al., 2016
Ion selective electrode	Water	10^{-6} to 0.1mol/L	Otal et al., 2022
Ion selective electrode	Water	10^{-5} to 10^{-2} mol/L	Carey and Coleman, 2014
Ion selective electrode with zirconium xylenol orange	Water	0 to 2 ppm	Levin et al., 2016
Ion-selective electrode (Ag/AgCl)	Liquid	0.195 to 200 ppm	Seguí Femenias et al., 2016s
Ion-selective electrodes	Water	0.001 to 20,000 ppm	Dhillon et al., 2016
Spectroscopic method using Eriochrome Black T	Water	0.3 to 5.0 mg/L	Diro et al., 2021

Table 4: Traditional methods to remove fluoride along with its operating condition

Method	Novel material	Analytical Technique to measure fluoride	Optimum Condition	Efficiency	Reference
Adsorption	Thermally activated sepiolite	Ion chromatography	Initial Concentration (IC) -50 mg/L pH - 7 Time (t)- 24 h Adsorbent dose (ADD) - 3.33 g/L	169.95 mg/g	Lee et al., 2020
Nano filtration	Flat sheet membrane	DR/2000 spectrophotometer	Initial Concentration (IC) -4 mg/L Flow rate – 0.4 L/min	83.3%	Yousefi et al., 2016
Adsorption	Spent bleaching earth	HANNA Instrument C200 model	ADD- 10 g/L IC - 5 mg/L Temperature (T) - 25°C pH – 7 Time – 4 h	0.6 mg/g	Malakootian et al., 2011
Adsorption	Granular red mud	Fluoride selective electrode	ADD- 5 g/L IC - 5 mg/L	0.851 mg/g	Tor et al., 2009

			Temperature (T) - 25°C pH – 4.7 Time – 6 h		
Adsorption	Granular ceramic	UV–visible spectrophotometer	IC – 10 mg/L pH – 6.9 t – 72 h T – 20 °C ADD- 20 g/L	0.929 mg/g	Chen et al., 2010
Adsorption	Bauxite	UV/Visible Spectrophotometer	ADD – 2 g/L IC – 5-20 mg/L pH – 6 T – 32 °C t– 120 min	5.16 mg/g	Sujana and Anand, 2011
Adsorption	Iron ore	Spectrophotometer	IC – 10 mg/L pH – 6 ADD – 5 g/L t – 120 min T– 22 °C	86 %	Kebede et al., 2016
Adsorption	Alum-impregnated activated alumina	Fluoride ion selective electrode	ADD – 8 g/L IC – 20 mg/L pH – 6.5 T – 32 °C t – 180 min	99%	Tripathy et al., 2006
Adsorption	Treated hydrated alumina	Fluoride-selective electrode	ADD – 1.6 g/L IC – 20 mg/L pH – 7 t – 60 min T – 23 °C	23.7 mg/g	Shimelis et al., 2006
Adsorption	Untreated hydrated alumina	Fluoride-selective electrode	IC – 20 mg/L pH – 7 ADD – 1.6 g/L t – 60 min T – 23 °C	7 mg/g	Shimelis et al., 2006
Adsorption	Bone char	Fluoride-selective electrode	IC – 50 mg/L pH – 7 ADD – 2 g/L t – 120 min T – 30 °C	7.32 mg/g	Rojas-Mayorga et al., 2013
Adsorption	Original waste mud	Fluoride ion-selective electrode	IC – 80 mg/L pH – 5 ADD – 10 g/L t– 60 min	4.2 mg/g	Kemer et al., 2009

			T – 20 °C		
Adsorption	Activated waste mud	Fluoride ion-selective electrode	IC – 80 mg/L pH – 5 ADD – 10 g/L t – 60 min T – 20 °C	2.8 mg/g	Kemer et al., 2009
Adsorption	Precipitated waste mud	Fluoride ion-selective electrode	IC – 80 mg/L pH – 5 ADD – 10 g/L Time – 60 min T – 20 °C	27.2 mg/g	Kemer et al., 2009
Adsorption column and an electrochemical system	Activated alumina	Fluoride selective electrode	IC – 10 mg/L ADD- 4 g/L pH – 6.2 t – 2 h T – 20 °C	2.2 mg/g	Lounici et al., 1997
Coagulation	Aluminium sulfate	Fluoride ion selective electrode	IC – 10 mg/L pH – 6.5 Coagulant dose – 0.4 g/L t – 60 min T – 60 °C	72.5 %	Dubey et al., 2018
Coagulation	Polyaluminium	Fluoride ion selective electrode	IC – 10 mg/L pH – 6.5 Coagulant dose – 2 g/L t – 60 min T – 60 °C	90.5 %	Dubey et al., 2018
Electrocoagulation	Aluminium electrodes	Ion Meter with a selective electrode	IC - 5-50 mg/L T - 25-55°C pH – 4-8.5	90%	Grich et al., 2019
Electrocoagulation	Aluminium electrodes	Ion Meter with a selective electrode	IC - 10 mg/L T - 25°C pH – 7-12 t – 90 min	68%	Ben Grich et al., 2019
Electrocoagulation	Aluminum plates	Fluoride selective electrode	IC – 500 mg/L pH – 7 t – 20 min T – 20 °C	93%	Aayef et al., 2021
Electrocoagulation	Iron and aluminum electrodes	UV-Visible spectrophotometer	IC – 10 mg/L Coagulant dose – 0.2 mg/L	91.2%	Tashauoei et al., 2023a

			T – 30 °C		
Electrodialysis	11 Cation exchange membrane (CEM) and 10 Anion exchange membrane (AEM)	Ion chromatography	IC – 100 mg/L pH – 8 t – 60 min T – 22 °C Voltage - 15 V Flow rate - 50 L/h	94 %	Aliaskari and Schäfer, 2021
Electrodialysis	CEM and AEM	Spectrophotometer	Current density - 2.34 mA/cm ² IC – 100 mg/L pH – 8 t – 60 min T – 22 °C Voltage – 24.4 V Flow rate - 90 dm ³ /h Energy - 0.895 kWh/m ³	83.4 %	Majewska-Nowak et al., 2015
Electrodialysis	CEM and AEM	Spectrophotometer	Current density - 2.34 mA/cm ² IC – 5 mg/L pH – 8 t – 60 min T – 32 °C Voltage – 24.4 V Flow rate - 90 dm ³ /h Energy - 0.895 kWh/m ³	83 %	Majewska-Nowak et al., 2015
Electrodialysis	CEM and AEM	Fluoride ion-selective electrode	Current density – 7.8 mA/cm ² IC – 5 mg/L pH – 10 t – 140 min T – 32 °C Flow rate - 1.5 L/min Energy - 0.895 kWh/m ³	76.2 %	Banasiak and Schäfer, 2009
Electrodialysis	10 CEM and 10 AEM	-	Current density – 2 A/dm ²	93 %	Amor et al., 1998

			IC – 3 mg/L t– 25 min T – 25 °C Flow rate - 180 L/h Voltage- 15 V		
Electrodialysis	10 CEM and 10 AEM	Ion chromatography	IC – 100 mg/L t– 16 min T – 25 °C Flow rate – 1.6 L/min Voltage- 10 V	98 %	Kabay et al., 2008
Electrodialysis reversal	100 CEM and 100 AEM	Ion chromatography	Current density - 0.3 mA/cm ² IC – 13 mg/L t – 12 h T – 25 °C Flow rate – 1 m ³ /h	90%	Patrocinio et al., 2019
Electrodialysis reversal	100 CEM and 100 AEM	Ion chromatography	Current density – 1.37 mA/cm ² IC – 7.49 mg/L t – 12 h T – 25 °C Flow rate – 1.0 m ³ /h	80%	Patrocinio et al., 2019
Membrane distillation	Polyvinylidene fluoride membrane	Fluoride ion selective electrode	IC – 10 mg/L Velocity - 0.046 m/s T of permeate – 20 °C T of feed – 53 °C	99.90 %	Boubakri et al., 2014
Membrane distillation	Polyvinylidene fluoride membrane	Ion Chromatography	IC – 20 mg/L Flow rate - 1 L/min t – 120 min T of permeate – 20 °C T of feed – 60 °C	99 %	Damtie and Choi, 2017
Membrane distillation	Polytetrafluoroethylene membrane	Ion Chromatography	IC – 5 ppm Flow rate - 0.8 L/min	99 %	Plattner et al., 2017

			Velocity - 0.04 m/s t – 5 h T of permeate – 25 °C T of feed – 55 °C pH – 7		
Nanofiltration and reverse osmosis	Polyamide NF90 4040 membrane	Fluoride ion selective electrode	IC – 2.32 ppm Flow rate - 18,867 L/h t– 4000 s T– 29 °C Maximum pressure- 41 bar	97%	Tahaikt et al., 2021
Nanofiltration and reverse osmosis	Polyamide BW 30LE4040 membrane	Fluoride ion selective electrode	IC – 2.32 ppm Flow rate - 20,737 L/h t– 4000 s T– 29 °C Maximum pressure- 41 bar	99.5%	Tahaikt et al., 2021
Nanofiltration and reverse osmosis	Polyamide TM710 membrane	Fluoride ion selective electrode	IC – 2.32 ppm Flow rate - 22,522 L/h t – 4000 s T– 29 °C Maximum pressure- 41 bar	99.7%	Tahaikt et al., 2021
Nanofiltration and reverse osmosis	TFC-S membrane	Ion Chromatography	IC – 3 ppm Partial pressure $3.8 \cdot 10^{-4}$ atm T– 25 °C pH – 12.5 t – 30 min	29 L/m ² h	Richards et al., 2010
Nanofiltration and reverse osmosis	UTC-60 membrane	Ion Chromatography	IC – 3 ppm Partial pressure $3.8 \cdot 10^{-4}$ atm T– 25 °C pH – 12.5 t – 30 min	164 L/m ² h	Richards et al., 2010
Nanofiltration and reverse osmosis	UTC-80A membrane	Ion Chromatography	IC – 3 ppm Partial pressure $3.8 \cdot 10^{-4}$ atm	10.9 L/m ² h	Richards et al., 2010

			T– 25 °C pH – 12.5 t – 30 min		
Nanofiltration and reverse osmosis	BW30 membrane	Ion Chromatography	IC – 3 ppm Partial pressure 3.8*10 ⁻⁴ atm T– 25 °C pH – 12.5 t – 30 min	17.2 L/m ² h	Richards et al., 2010
Nanofiltration and reverse osmosis	NF90 membrane	Ion Chromatography	IC – 3 ppm Partial pressure 3.8*10 ⁻⁴ atm T– 25 °C pH – 12.5 t – 30 min	54.9 L/m ² h	Richards et al., 2010
Nanofiltration and reverse osmosis	ESPA4 membrane	Ion Chromatography	IC – 3 ppm Partial pressure 3.8*10 ⁻⁴ atm T– 25 °C pH – 12.5 t – 30 min	32.4 L/m ² h	Richards et al., 2010
Precipitation	Hydrate lime	Fluoride-selective electrode	IC – 3200 ppm T– 25 °C t – 120 min pH – 10	98.1%	Ezzeddine et al., 2015
Precipitation and reverse osmosis	Hydrate lime	Fluoride-selective electrode	IC – 3200 ppm T– 25 °C t – 120 min pH – 10 Pressure – 60 psi	99.7%	Ezzeddine et al., 2015
Precipitation and Surface Adsorption	Crushed limestone	Fluoride selective electrode	IC – 3 mg/L ADD- 33.3 g/L pH – 6.38 t – 20 h	96 %	Turner et al., 2005
Precipitation method	Lanthanum chloride	Optical emission spectrometer	IC – 0.03 mol/L pH – 3 t – 6 h T – 60 °C	97.7 %	Xia et al., 2021
Reverse Osmosis	Polyamide membrane	Fluoride-selective electrode	IC – 2.13 ppm T– 40 °C Pressure – 80 psi pH – 8	97.4 %	Gedam et al., 2012

Vacuum Enhanced Membrane distillation	Polytetrafluoroethylene membrane	Ion Chromatography	IC – 5 ppm Flow rate - 0.8 L/min Velocity - 0.04 m/s t – 5 h T of permeate – 25 °C T of feed – 55 °C pH – 7 Permeate vacuum = 300 mbar	98 %	Plattner et al., 2017
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Table 5: Advanced techniques to remove fluoride along with its operating condition

Method	Novel material	Analytical Technique to measure fluoride	Optimum Condition	Efficiency	Reference
Capacitive Deionization	Graphene Oxide/Hydroxyapatite Composite Electrode	Ion chromatography	IC -1 Mm Energy consumption - 0.18 Wh/mmol Voltage – 1.2 V t – 20 min	96%	Park et al., 2021
Capacitive Deionization	Carbon electrodes	Ion chromatography	IC -2 Mm Voltage – 1.2 V Flow rate - 56 mL/min	0.114 mg/g	Tang et al., 2016
Electrocoagulation and ceramic microfiltration	Polymer cellulose acetate and with polymer-nanoparticle	Fluoride ion electrode	IC -7.89 mg/L Current density - 15 A/m ² pH – 7.9 Time – 20 min	91.76 %	Changmai et al., 2018
Electrocoagulation and ceramic microfiltration	Polymer cellulose acetate and with polymer-nanoparticle	Fluoride ion electrode	IC -4.79 mg/L Current density - 15 A/m ² pH – 7.9 t – 20 min	93 %	Changmai et al., 2018
Electrocoagulation and ceramic microfiltration	Polymer cellulose acetate and with	Fluoride ion electrode	IC -1.78 mg/L Current density - 15 A/m ² pH – 7.9	99.45 %	Changmai et al., 2018

	polymer-nanoparticle		t – 20 min		
Electrocoagulation and ceramic microfiltration	Kaolin	Fluoride ion electrode	IC -10 mg/L Current density - 625 A/m ² t– 35 min Pressure – 200 kPa	91 %	Ghosh et al., 2013
Membrane Capacitive Deionization	Acrylic plate, anode electrode, CEM	Ion chromatography	IC -1 Mm Volt - 1.2 V Flow rate - 30 mL/min t - 10 min pH – 10	21.61%	Pan et al., 2018
Membrane Capacitive Deionization	Activated carbon electrodes, AEM and CEM	Fluoride ion-selective electrode	IC - 20 Mm T– 20 °C pH – 7 t – 1800 sec	77.9%	Pang et al., 2022
Membrane Capacitive Deionization	Porous carbon electrodes AEM and CEM	Fluoride ion-selective electrode	IC – 7.71 ppm Energy consumed - 0.06 kWh/m ³ Flow rate – 0.3 L/min Voltage – 1.39 V Current – 6.75 A	99%	Uzun And Debik, 2021
Membrane Capacitive Deionization	Wood apple shell activated carbon electrode	Ion chromatography	IC – 10 ppm Voltage – 1.2 V pH – 7.2	89.23%	Gaikwad and Balomajumder, 2018
Membrane Capacitive Deionization	Wood apple shell activated carbon electrode	Ion chromatography	IC – 100 ppm Voltage – 1.2 V pH – 7.2	31.49 %	Gaikwad and Balomajumder, 2018

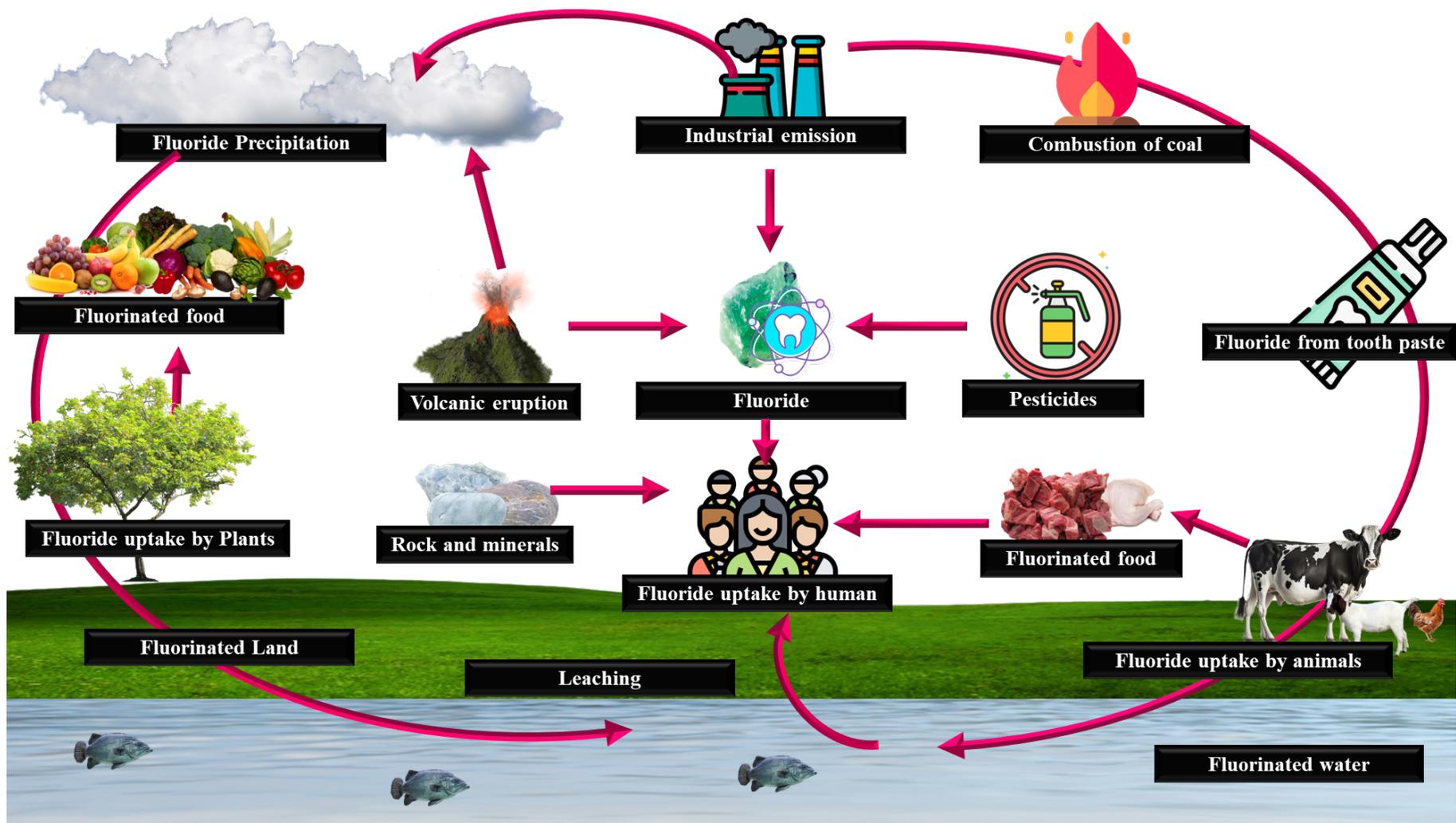


Fig. 1. Sources and pathway of Fluoride in the environment

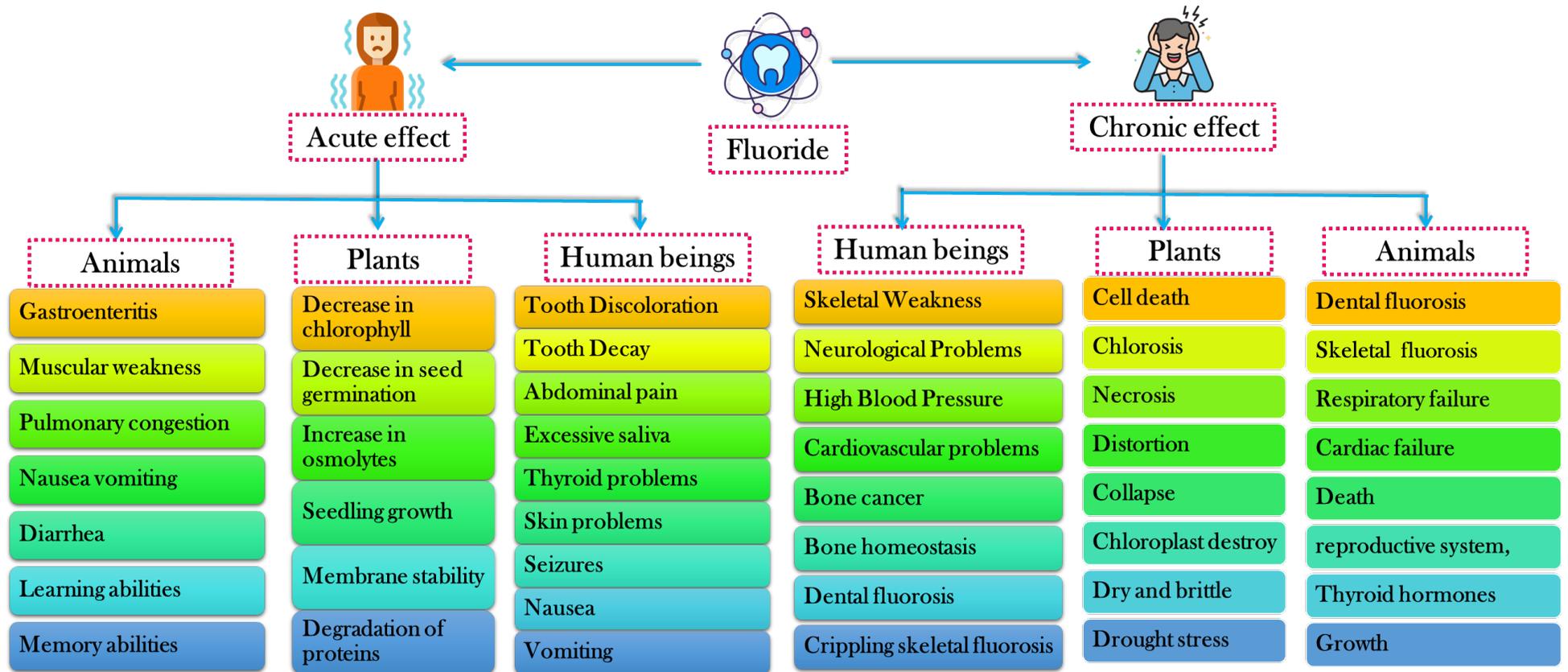


Fig. 2. Acute and chronic effects of Fluoride on animals, plants and human beings

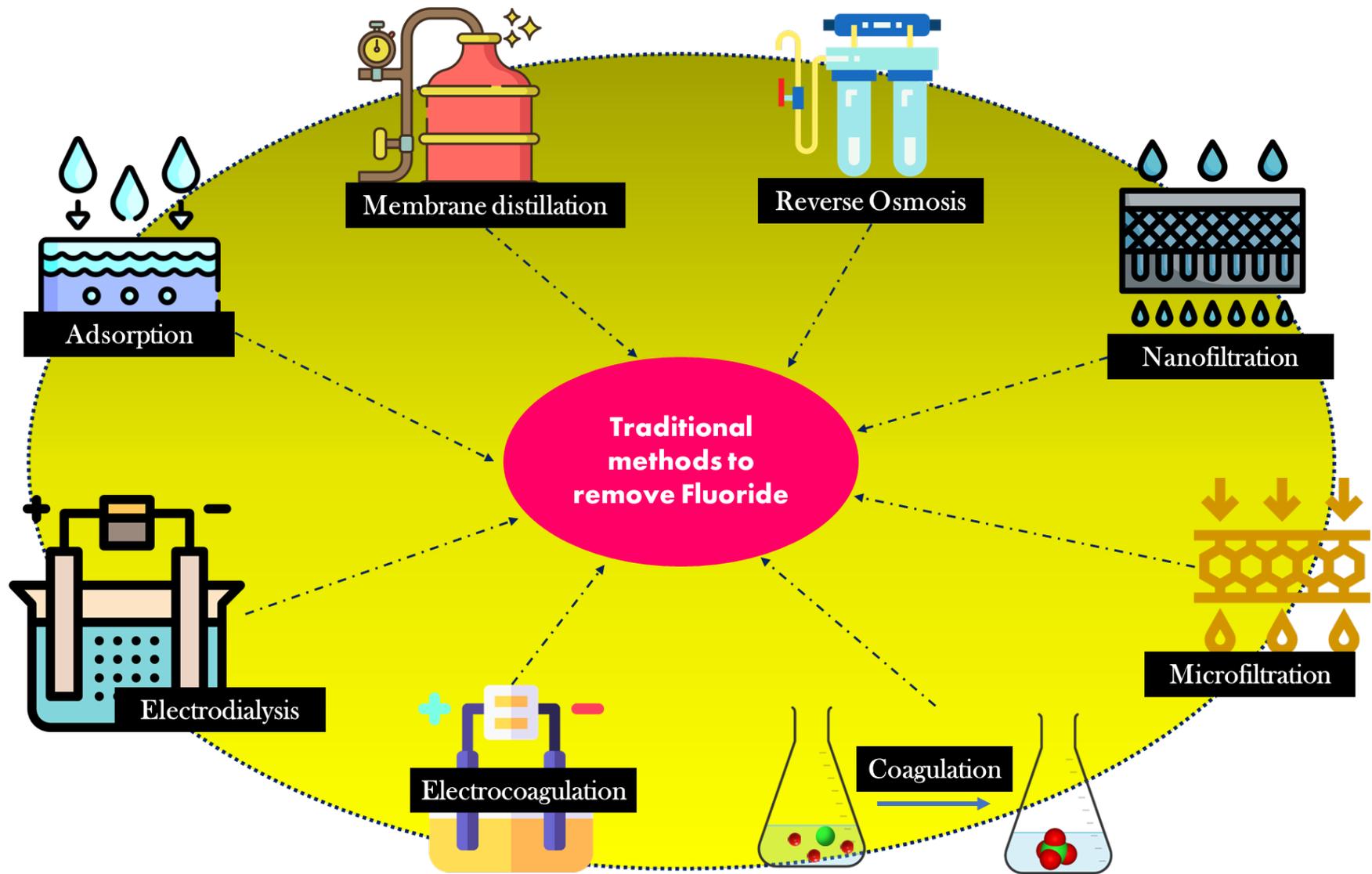


Fig. 3. Various traditional methods to remove fluoride from water

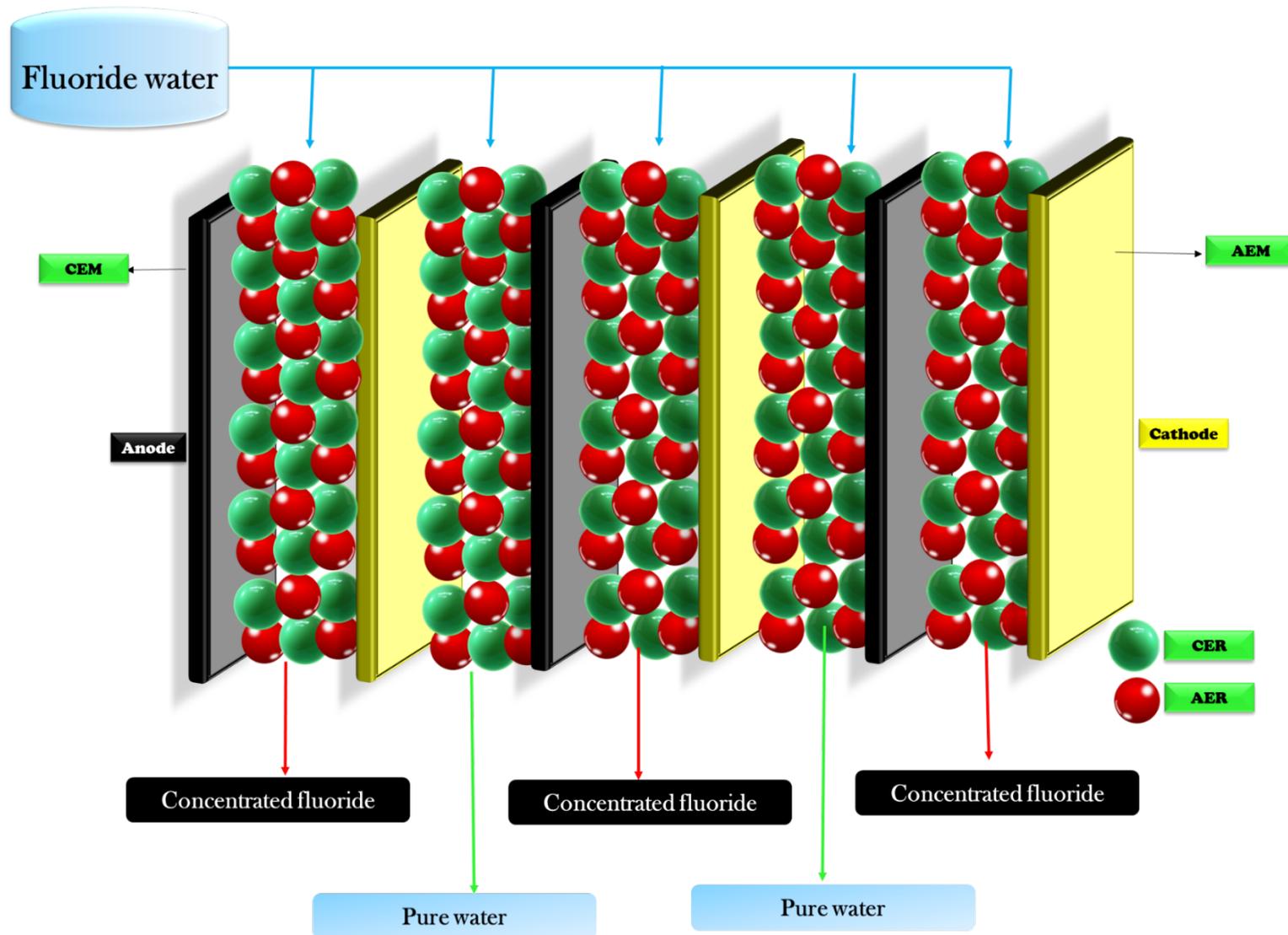


Fig. 4. Schematic representation of electrodeionization to remove fluoride from water

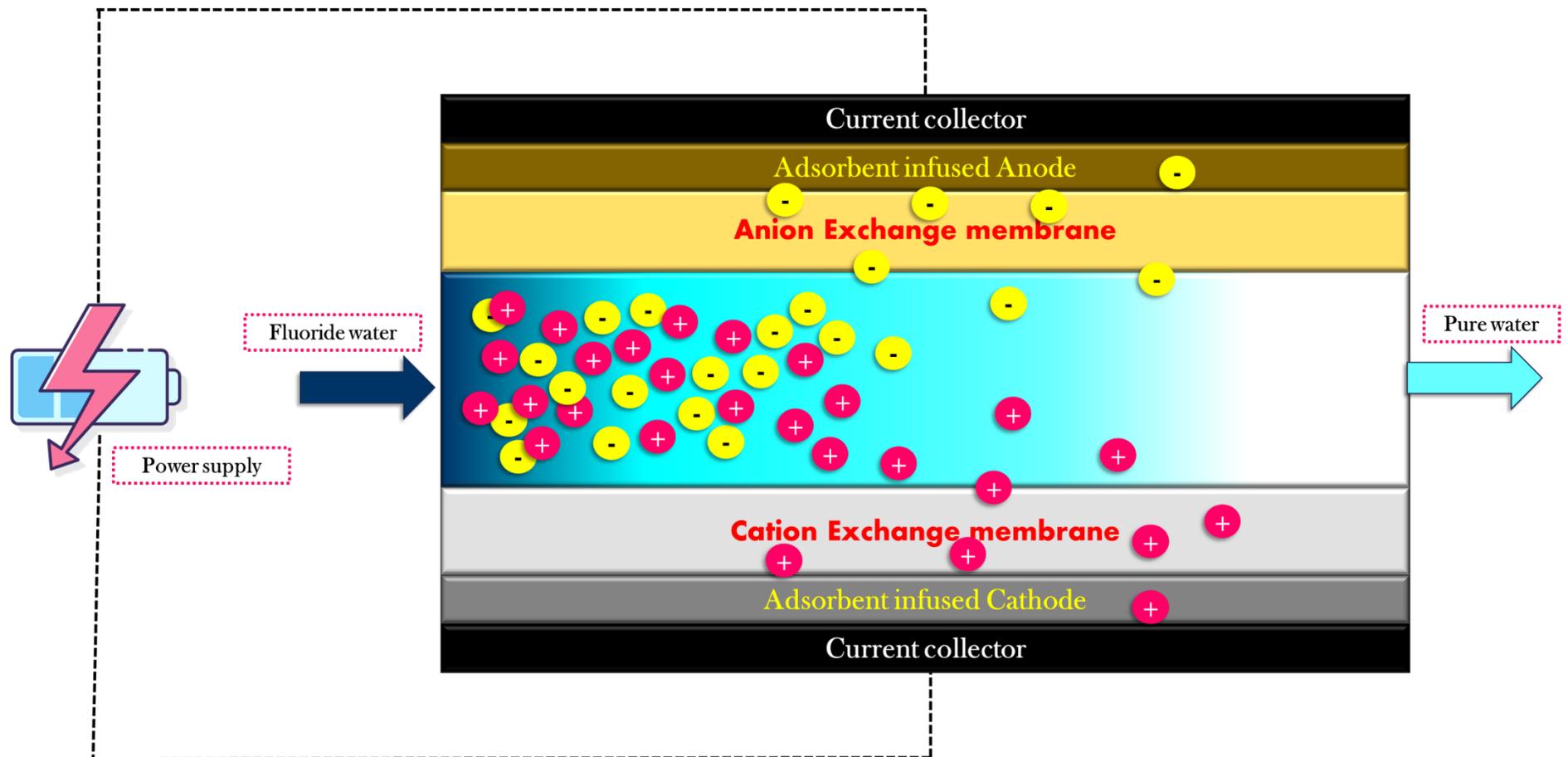


Fig. 5. Schematic representation of Membrane capacitive deionization to remove fluoride from water

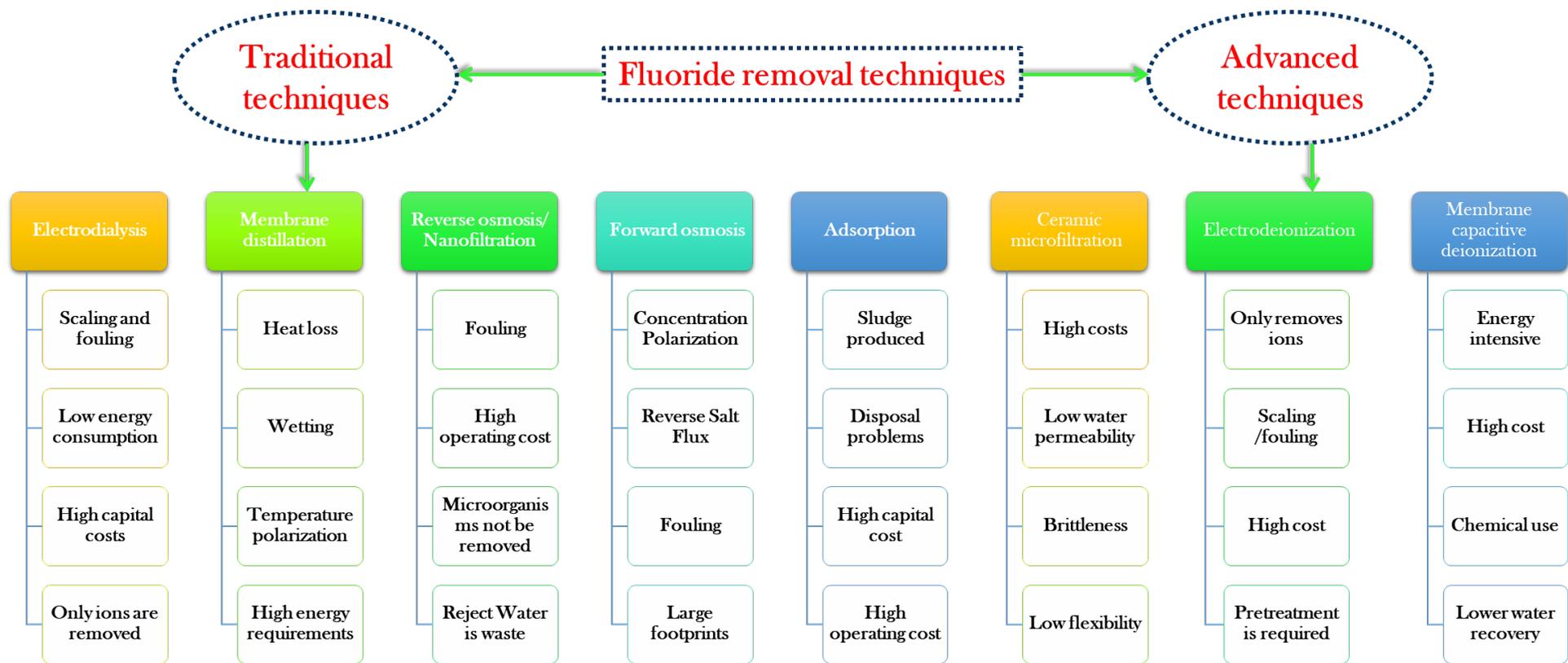


Fig. 6. Limitation of different techniques for fluoride removal process