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REVIEW ARTICLE

Cesium removal from radioactive wastewater by adsorption and membrane technology

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HIGHLIGHTS

- Removal of cesium from radioactive wastewater is still a challenging.
- Main approaches used for waste treatment in Fukushima Daiichi accident were reviewed.
- Kurion/SARRY system + desalination system and ALPS were briefly introduced.
- The removal of cesium by adsorption and membrane separation were summarized.

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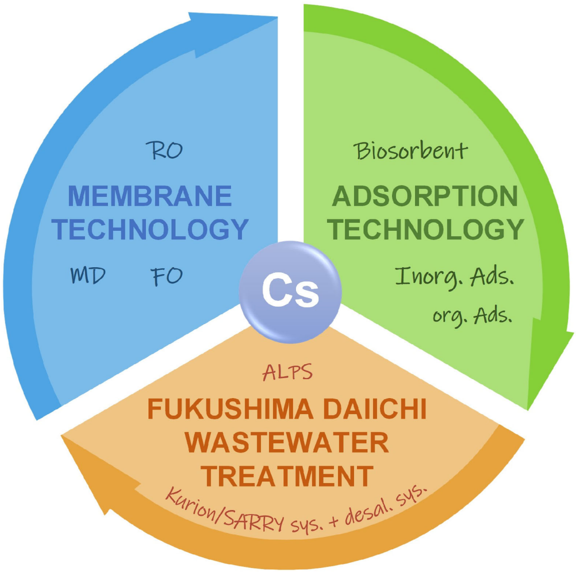
GRAPHIC ABSTRACT



ABSTRACT

Radiocesium is frequently present in radioactive wastewater, while its removal is still a challenge due to its small hydrated radius, high diffusion coefficient, and similar chemical behavior to other alkali metal elements with high background concentrations. This review summarized and analyzed the recent advances in the removal of Cs^+ from aqueous solutions, with a particular focus on adsorption and membrane separation methods. Various inorganic, organic, and biological adsorbents have undergone assessments to determine their efficacy in the removal of cesium ions. Additionally, membrane-based separation techniques, including reverse osmosis, forward osmosis, and membrane distillation, have also shown promise in effectively separating cesium ions from radioactive wastewater. Additionally, this review summarized the main approaches, including Kurion/SARRY system + desalination system and advanced liquid processing system, implemented after the Fukushima Daiichi nuclear power plant accident in Japan to remove radionuclides from contaminated water. Adsorption technology and membrane separation technology play a vital role in treatment of contaminated water.

1 Introduction

The production of radioactive wastewater is an inevitable

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byproduct of various activities related to nuclear operations (Wang et al., 2018). This wastewater typically contains a range of harmful radionuclides, including uranium, plutonium, cesium, and iodine, which emit dangerous forms of ionizing radiation, such as γ rays. Exposure to such radiation can have severe health effects, such as cancer, genetic mutations, and other hazardous consequences for living organisms. Therefore, it is crucial to remove radionuclides from radioactive wastewater to prevent potential damage to public health and the environment.

Radioactive cesium is a critical constituent of radioactive wastewater, and its removal is essential and challenging, due to its ubiquitous presence in radioactive wastewater as a significant fission product of uranium. The most frequently encountered uranium fission products are ^{134}Cs and ^{137}Cs, which were the primary contributors to radioactivity in the Fukushima Daiichi nuclear disaster (Japan) in 2011 (Beyea et al., 2013). These isotopes can cause chemical poisoning and internal radiation exposure if assimilated by organisms, and their soluble ions can migrate considerable distances in the environment. Additionally, ^{137}Cs has a longer half-life of 30.17 years and emits gamma rays, rendering it a primary contributor to later stages of radioactive contamination compared to ^{134}Cs, which has a half-life of 2.06 years. Thus, the removal of cesium ions from solutions, particularly radioactive wastewater, is critical for safeguarding ecosystem health and human well-being.

The distinctive physical and chemical properties of cesium ions pose a significant challenge to their separation from aqueous solutions. Being an alkali metal element, cesium exists only in the + 1 oxidation state and readily forms Cs^+ ions in water. Cesium's large size and low charge make it the most electropositive element and give rise to highly soluble ionic compounds. Additionally, cesium behaves similarly to Na^+ and K^+ and does not form insoluble complexes with common chemicals or precipitate at various pH values. The crystal ionic radii of these three ions follow the sequence $Na^+ < K^+ < Cs^+$, and their sizes are comparable when fully hydrated in the order $Na^+ > Cs^+ > K^+$ (Wang and Zhuang, 2019c). These similarities present a significant challenge to the selective separation of cesium from aqueous solutions, particularly at concentrations as low as ppm or ppb, given the abundant presence of Na^+ and K^+ in such solutions.

The concern on radiocesium in aquatic environments has received increasing attention in recent years. Numerous reviews have been published covering various aspects of cesium ions. For instance, Alby et al. (2018) conducted comprehensive summaries of diverse nanostructured inorganic adsorbents for cesium, and Arnal et al. (2003) reviewed reverse osmosis techniques applied in the context of treating liquid wastes containing ^{137}Cs. An overview of adsorbents based on metal hexacyanoferrates is presented, showcasing remarkable adsorption capacity and selectivity toward cesium ions (Wang et al., 2018). Furthermore, diverse separation methods for cesium ions was summarized and analyzed (Wang and Zhuang, 2019c). Among the array of techniques available, both adsorption and membrane separation technologies have emerged as promising contenders for addressing low-concentration Cs^+-containing solutions. Furthermore, processes centered around reverse osmosis and adsorption techniques have been proven to be of paramount importance in the treatment of effluents from the Fukushima Daiichi site. However, a comprehensive review elaborating on these two pivotal technologies and their practical applications in treating Fukushima's wastewater is conspicuously absent.

Given the significance and complexity of removing cesium ions, this review aimed to provide an overview of recent progress in this field. First, this article highlighted the latest developments in adsorption and membrane separation technologies for cesium removal. Then, this review focused on the removal of radioactive Cs, with particular emphasis on the treatment process for the nuclear wastewater from the Fukushima disaster.

2 Adsorption technology for Cs removal

Adoption of adsorption technology is advantageous due to its high stability, low operational cost, ease of use, minimal secondary pollution, and low energy consumption, making it highly suitable for treating low concentration radioactive wastewater (Zhuang et al., 2022a; 2022b). The adsorbent is a critical component of adsorption technology. Currently, various types of adsorbents have been reported, such as inorganic, biosorbents, and other organic adsorbents, as shown in Fig. 1 and Table 1 (Liu et al., 2014; Wang and Guo, 2020a; 2020b; 2022; 2023). Moreover, Table 2 listed the adsorption capacity of some typical adsorbents. Generally speaking, most types of adsorbents could reach a high cesium adsorption capacity, but in fact only several types have a good adsorption selectivity.

Adsorbents possessing high adsorption capacity and selectivity toward cesium ions are highly desirable. However, as reported by several studies, some adsorbents exhibit poor adsorption capacity for Cs. For instance, Caccin et al. (2013) found that activated carbon made from coconut shells had poor affinity for cesium, with adsorption capacities of less than 1 mg/g. Similarly, Suzuki et al. (2013) did not observe favorable adsorption properties of vermiculite as a cesium adsorbent in artificial seawater. Hexacyanoferrates are recognized as effective adsorbents for cesium ions and play a crucial role in the treatment of Fukushima Daiichi waste effluents (Wang et al., 2018).

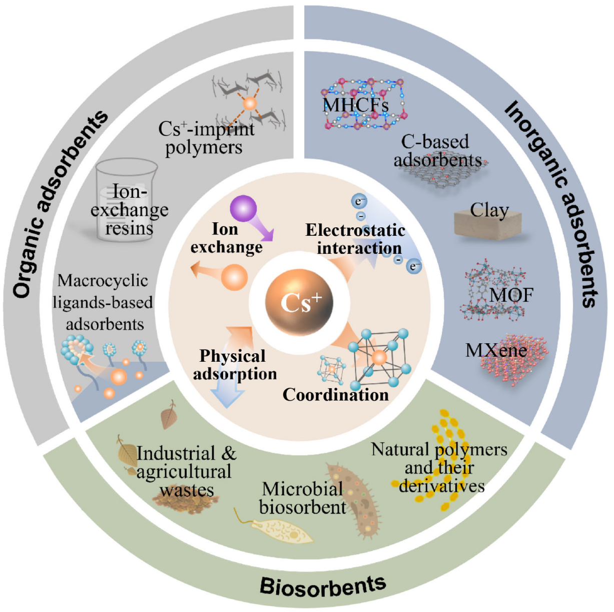

Fig. 1 Key adsorbents and their mechanisms for cesium ions removal.

2.1 Inorganic adsorbents

For cesium ions removal, several inorganic adsorbents have been extensively studied, such as hexacyanoferrates, carbon-based materials, clay minerals, geopolymers, metal organic frameworks (MOFs), and others (Alby et al., 2018). Among these adsorbents, hexacyanoferrate has garnered significant interest because of its high adsorption capacity and selectivity toward cesium ions. Additionally, activated carbon, zeolite, and silicotitanate have also been utilized as effective adsorbents in the actual treatment of the Fukushima Daiichi waste effluents.

Hexacyanoferrates are effective adsorbents for cesium removal due to their distinctive structure and chemical properties. Hexacyanoferrates consist of metal ions surrounded by cyanide ligands, forming a three-dimensional lattice structure with numerous binding sites for cesium ions to attach to (Wang et al., 2015). The negative surface charge of hexacyanoferrates also facilitates the attraction of positively charged cesium ions through electrostatic interactions. The selectivity of hexacyanoferrates toward cesium is attributed to the size of the cesium ion, which is a good match for the size of the cavities within the hexacyanoferrates lattice structure (3.2 Å) (Escobar et al., 2022). The adsorption capacity of hexacyanoferrates for cesium ions has been reported to vary depending on their composition and preparation conditions. In previous study, it was observed that capacity for Cs$^+$ using hexacyanoferrates can range from 24 to 175 mg/g (Zhuang and Wang, 2019a; Zhuang et al., 2022a), depending on the specific preparation method and

Table 1 Various adsorbents for cesium ions

Types	Adsorbents
Inorganic adsorbents	Hexacyanoferrates: NiHCF, CoHCF, CuHCF, FeHCF etc. Carbon-based materials: activated carbon, carbon nanotubes, graphene oxide, and biochar Clay minerals: bentonite, zeolite, sepiolite, kaolinite, etc. Others: Geopolymers, MXenes, MOFs, titanate nanosheets, ammonium molybdophosphate, etc.
Organic adsorbents	Resins: resorcinol-formaldehyde resin etc. Cs$^+$-imprinted polymers Macrocyclic ligands based adsorbents: crown ethers-based adsorbents and calixarenes-based adsorbent Synthetic polymer modified with polyaniline, polyphosphazene
Biosorbents	Microbes: Bacteria, filamentous fungi, yeast, algae, etc. Industrial and agricultural wastes: brewery's waste biomass etc. Biopolymers: chitosan, alginate, cellulose, etc.

Table 2 Cesium adsorption capacity by some typical adsorbents

Adsorbents	Adsorption capacity	References
Bentonite	37 ug/g–300.35 mg/g	Seliman et al. (2014); Bayülken et al. (2011)
Zeolite	9.055 ug/g–229.6 mg/g	Kim et al. (2013); Dran'kov et al. (2022)
GO	95.46–528 mg/g	Xing et al. (2020); Kaewmee et al. (2017)
CNT	31.23–107.7 mg/g	Yang et al. (2014); Ali et al. (2020)
Activated carbon	0.76–35.426 mg/g	Caccin et al. (2013); Xing et al. (2020)
Alginate	57.1–190.8 mg/g	Mimura et al. (2001); Chang et al. (2022)
Biochar	52.63–458 mg/g	Park et al. (2023); Abu-Khadra et al. (2016)
Microorganisms	16.67–238.1 mg/g	Yin et al. (2017); Ngwenya & Chirwa (2010)
MOF	57.7–432 mg/g	Wang et al. (2022); Wang et al. (2015)
MXene	25.4–315.91 mg/g	Khan et al. (2019); Shahzad et al. (2020)
Hexacyanoferrates	24.9–241 mg/g	Zhuang et al. (2022a); Chen et al. (2015b)

composition of the hexacyanoferrate adsorbent. Additionally, hexacyanoferrates exhibit exceptional resistance against gamma irradiation. All these reasons make hexacyanoferrates an effective adsorbent for cesium removal (Parajuli et al., 2016).

However, the microcrystalline structure of hexacyanoferrates presents a significant limitation for column operation and separation (Chen et al., 2015b). To overcome this limitation, immobilization and magnetic modification have been widely adopted (Vincent et al., 2015). Immobilization involves embedding hexacyanoferrate into various polymers or anchoring its precursors into solids for *in situ* synthesis. In contrast, magnetic modification imparts magnetic properties to hexacyanoferrate, enabling its rapid separation through the application of an external magnetic field. Wang et al. (2018) summarized metal hexacyanoferrates-based adsorbents for cesium removal.

Carbon-based adsorbents, including activated carbon, biochar, graphene oxide and carbon nanotubes, have gained significant attention in the field of wastewater treatment due to their high surface area, functional groups, and cost-effectiveness (Xing et al., 2016; Xu and Wang, 2017; Wang and Wang, 2019; 2022; Qiao et al., 2023; Xi et al., 2023; Zhuang and Wang, 2023a; 2023b). Numerous studies have investigated the adsorption capacity of Cs^+ using various carbon-based adsorbents. Reported adsorption capacities of carbon-based adsorbents have ranged from several tens to several hundred mg/g (Sun et al., 2013; Wang and Zhuang, 2019c). For instance, Palansooriya et al. (2022) demonstrated the effectiveness of gasified biochar produced from different feedstock mixtures and KOH-modified biochar in removing Cs^+ from a solution, with an impressive adsorption capacity of 62.7 mg/g. Furthermore, research studies have shown that graphene oxide exhibits the highest adsorption capacity among carbon-based adsorbents. Kaewmee et al. (2017) reported an impressive adsorption capacity of up to 528 mg/g for Cs^+ using graphene oxide.

Carbon-based adsorbents were used for the removal of Cs^+ from aqueous solution (Xing et al., 2020). Results showed graphene oxide had the highest adsorption capacity toward Cs^+ (95.462 mg/g) in the mono-component system, followed by carboxyl modified carbon nanotubes (44.174 mg/g) and activated carbon (35.426 mg/g), as determined by the Langmuir model. Additionally, carbon materials were used as electrode materials in capacitive deionization (CDI) for the electro-assisted adsorptive removal of Sr^{2+} and Cs^+. Previous study found removal rates of 40.58% and 62.05% for Sr^{2+} and Cs^+, respectively (Liu and Wang, 2021). Moreover, the reverse application of cell voltage could desorb the radionuclides and regenerate the carbon materials.

Clay minerals, including bentonite, zeolite, sepiolite, and kaolinite, are an abundant and cost-effective resource for removing Cs^+ from wastewater (Bayülken et al., 2011). The chemical composition of clay minerals varies depending on the geological conditions, which can significantly affect their adsorption performance. Among different clays, zeolite and bentonite are extensively studied for cesium adsorption (Pandey, 2017). The primary adsorption mechanisms for cesium ions in clay include ion-exchange and electrostatic attraction (Jiménez-Reyes et al., 2021). The enhancement of adsorption capacity in clay minerals can be achieved through acid/base treatment, surfactant modification, and grafting techniques (Wang et al., 2019). Moreover, the incorporation of clay minerals with supplementary materials, including carbon-based substances or metal oxides, has demonstrated improved cesium adsorption capabilities (Novikau et al., 2022).

Geopolymers, classified as an inorganic substance, possess a continuous, three-dimensional amorphous aluminosilicate framework. With their mesoscale porosity, modifiable chemical composition, and chemical durability, they emerge as a highly potential option for the adsorptive elimination of contaminants (Siyal et al., 2018). Additionally, the ability of geopolymers to exchange counter cations (e.g., Na^+ and K^+) with Cs^+ in the framework makes them effective in the adsorption of Cs^+. Recent studies have reported that printed geopolymers can achieve 80.1 mg/g Cs^+ adsorption capacity (Ma et al., 2023), demonstrating their high potential in the field of pollution control and remediation, particularly in the removal of cesium ions from liquid environments.

Novel materials, including MXenes, MOFs, and titanate nanosheets (Ai et al., 2018; Shahzad et al., 2020), have also been utilized for the adsorptive removal of cesium ions. MOFs, in particular, have shown remarkable potential for this application (Sheta et al., 2023). The exceptional adsorption properties of various MOFs for Cs^+ was reported (Naeimi and Faghihian, 2017; Cheng et al., 2018; Guo et al., 2021; Roy and Stoddart, 2021). Aguila et al. (2016) modified a stable MOF, MIL-101, with sulfonic acid groups, resulting in a significant increase in the material's adsorption properties for Cs^+ and Sr^{2+}. At a molar ratio of 4:1 between the adsorbent and ion solution, the removal efficiency for Cs^+ and Sr^{2+} reached 99.9% and 98.92%, respectively. Additionally, Wang et al. (2015) reported on a novel 3D uranyl organic framework material with high radiation resistance, chemical stability over a broad pH range, and a high surface area (924 cm^2/g). This material exhibited an excellent selectivity for cesium removal while maintaining the polycatenated framework structure, with significant umbellate distortions. These studies demonstrate the potential of MOFs and other novel materials for the efficient removal of cesium ions from wastewater.

MXene, a novel material known for its high surface area and abundant surface functional groups, has demonstrated impressive capabilities in the removal of cesium ions through adsorption. Typical MXene, $Ti_3C_2T_x$, has been reported to exhibit a Cs^+ adsorption capacity ranging from 25.4 to 148 mg/g in $Cs^+–H_2O$ solutions (Khan et al., 2019; Jun et al., 2020). The underlying adsorption mechanisms likely involve electrostatic interactions and ion exchanges. However, despite its high adsorption capacity, MXene's selectivity for cesium ions is relatively poor and susceptible to interference from other ions (Jun et al., 2020). To enhance its selectivity, incorporating additional effective adsorbent materials, such as Prussian blue, can prove beneficial (Shahzad et al., 2020). Currently, there is limited reported research on MXene-based adsorbents for cesium ions.

2.2 Organic adsorbents

Extensive research has been conducted on the development of diverse synthetic polymers and composite adsorbents for the purpose of eliminating cesium ions from wastewater (Wang and Zhuang, 2019a; 2019b). These materials include those that have been modified with crown ethers, polyaniline, polyphosphazene, and calixarenes, and have demonstrated high selectivity and capacity for cesium ion removal. In recent years, Cs^+-imprinted polymers have been developed using ion-imprinting strategies, which can further enhance selectivity and adsorption performance. The utilization of these materials has resulted in substantial advancements in the development of efficient and effective approaches for removing cesium ions from contaminated environments.

With substantial functional groups, ion exchange resins are capable of exchanging with nuclide ions present in wastewater, resulting in the simultaneous removal of multiple nuclides (Fiskum et al., 2020; Wang et al., 2023). This technique finds widespread application in water treatment systems within several nuclear power plants for tasks such as producing primary circuit desalination water, treating secondary circuit cooling water, and dealing with evaporator residuals. In terms of ion exchange resin performance for nuclide removal, El-Naggar et al. (2014) found that the cation exchange resin (Lewatit S100) had an adsorption capacity of 398 mg/g for Cs^+ and 266 mg/g for Co^{2+}. Resorcinol-formaldehyde resin was also utilized to treat actual wastewater collected from the Hanford nuclear power plant, which contained a total Cs^+ concentration of approximately 10 mg/L, along with higher concentrations of other metal ions. The results demonstrated that resorcinol-formaldehyde resin achieved a 99.9% removal efficiency for Cs^+ (Hassan and Adu-Wusu, 2005). These studies show that cation exchange resins are capable of efficiently removing nuclides. However, if the wastewater contains high levels of radioactive or non-radioactive ions, the resin bed may quickly become saturated and ineffective. Additionally, the resin used to treat radioactive wastewater is typically non-regenerable, which necessitates immediate replacement upon failure.

The use of macrocyclic ligands based on crown ethers is widespread in solvent extraction systems for the separation of Cs, due to the matching crystal ionic radius of the Cs ion and the cavity of the crown ether ligand. Strong cation-π interactions have been reported between them (Escobar et al., 2022). To remove radioactive Cs^+, functional materials based on crown ethers have been developed, with different crown ethers incorporated into porous supports using chemical grafting or physical entrapment (Awual et al., 2014; Liu et al., 2019c). However, there are two issues that cannot be ignored. Chemical grafting may disrupt the electronic cloud distribution, which can lead to the failure to capture Cs^+ ions. In addition, loading stability is also a concern for physical entrapment, as small molecule substances may escape from the crosslinked polymer.

Novel organic adsorbents have been synthesized using ion-imprinting strategies to achieve selective separation of Cs^+ ions. Ion-imprinted polymers (IIPs) have specific binding sites that match the size and charge of a particular ion, resulting in high selectivity and efficiency for cesium ion removal (Kusumkar et al., 2021). For example, Shamsipur and Rajabi (2013) synthesized a Cs^+-imprinted polymer using methacrylic acid as the functional monomer, dibenzo-24-crown-8 ether as a selective crown ether, ethyleneglycol dimethacrylate as the crosslinker, and AIBN as the radical initiator. The resulting Cs^+-imprinted polymer demonstrated a high selectivity for Cs^+ over other metal ions and exhibited a maximum adsorption capacity of 50 mg/g. In another study, novel ion-imprinted polymers for Cs(I) ions were synthesized using a surface imprinting technique with a SBA-15 support matrix. The as-prepared adsorbents exhibited a high adsorption capacity toward cesium ions and could be easily regenerated with a dynamic desorption ratio of over 92% using a 2 mol/L HCl solution (Meng et al., 2015). These findings highlight the potential of ion-imprinting strategies for developing efficient and selective adsorbents for cesium ion removal (Kusumkar et al., 2021).

2.3 Biosorbents

The study of biosorbents for the adsorptive removal of cesium ions has encompassed a range of organisms, including yeast, bacteria, filamentous fungi, and algae, as well as products derived from microbes, animals, plants, and industrial and agricultural wastes (Liu et al., 2014). Wang and Chen (2006; 2009; 2014) reviewed the biosorbents and their ability to adsorb heavy metal and radionuclide ions. Given their cost-effectiveness, abundant availability, and environmentally-friendly nature, biological adsorbents exhibit significant promise for practical applications. These radionuclides-loaded biosorbents can undergo additional processing methods, such as high-temperature sintering, impregnation, or vitrification, after adsorption. Subsequently, they can be securely stored in specialized containers to prevent radiation leakage.

Over the years, a series of investigations involving the use of biosorbents to remove Cs^+ have been conducted by our research group. Waste yeast cells (Chen and Wang, 2007a; 2007b; 2008a; 2008b; 2010; Yin et al., 2017; Chen et al., 2020a; 2020b), brown algae (Hu et al., 2020), and natural polymers, including chitosan (Chen and Wang, 2012a; 2012b; 2016; Zhu et al., 2012; 2014; Zhuang et al., 2021; 2022a; 2022b), cellulose (Zhuang and Wang, 2019a; 2019b), and alginate (Yu et al., 2017; Zhuang and Wang, 2018), have been employed as the research foundation by our group. They were chemically or physically modified to enhance their adsorption capacity and selectivity toward nuclide ions like Cs^+. In addition, the preparation and modification of various biosorbents, as well as their adsorption properties toward different pollutants were summarized (Wang and Chen, 2009; Wang and Zhuang, 2017; 2022).

Microbial biosorbents have been proven to be effective in selectively adsorbing radionuclides. Early studies conducted in the 1950s explored the use of biological treatment procedures for cesium removal in water and wastewater (Eden et al., 1954; Morton and Straub, 1956). Subsequent studies found that the microbial slime layer on filter media significantly contributed to cesium elimination. In recent years, advances in biotechnology have increased attention toward using microorganisms for adsorbing radioactive nuclides and other pollutants from wastewater.

Waste yeast cells, specifically *Saccharomyces cerevisiae*, are a typical example of microbial biosorbents that exhibit high efficiency in adsorption and removal of cesium ions. This is due to the abundant functional groups on the cell surface of *Saccharomyces cerevisiae*, which contribute to its ability to adsorb metal ions. Although the adsorption capacity of raw *Saccharomyces cerevisiae* for cesium ions is low, immobilization treatment and irradiation pretreatment can increase its maximum adsorption capacity to 18.778 mg/g. Therefore, utilizing this inexpensive and readily available biological material offers a practical and cost-effective option for cesium ion removal through adsorption.

Waste yeast cells (*Saccharomyces cerevisiae*) are a typical microbial biosorbent with a benign ability to adsorb metal ions. Due to the presence of numerous functional groups on the cell surface and extracellular polymeric substances (EPS), *Saccharomyces cerevisiae* can remove various kinds of metal ions via electrostatic interaction or coordination. It has been demonstrated that *Saccharomyces cerevisiae* can effectively adsorb cesium ions with a maximum adsorption capacity of 10.1 mg/g (Chen and Wang, 2010). However, compared to its adsorption of other heavy metal ions such as Ag^+, there is significant room for improvement in both the adsorption capacity and selectivity for cesium ions. To address this, immobilization treatment and irradiation pretreatment can significantly increase the maximum adsorption capacity of cesium ions by more than 10-fold (Chen and Wang, 2010; Yin et al., 2017). Therefore, the use of waste yeast cells offers a practical and cost-effective option for cesium ion removal through adsorption.

Brown algae exhibit significant promise as biosorbents for removing cesium ions from wastewater. Their unique cell wall components, particularly alginate and fucoidan, contribute to their effectiveness as biosorbents. Alginate, which is a biopolymer composed of β-D-mannuronate and α-L-guluronate residues, contains functional groups like carboxyl and hydroxyl. These functional groups can facilitate metal adsorption through ion exchange and chelation. The efficiency of alginate as a biosorbent may vary based on the composition and sequences of different algal species and tissues. In a previous study, we investigated the biosorption of strontium and cesium ions onto *Undaria pinnatifida*, a marine brown alga, and found that *Undaria pinnatifida* had a maximum adsorption capacity of up to 146.3 mg/g for cesium ions (Hu et al., 2020). This demonstrates the potential of brown algae as a cost-effective and environmentally friendly solution for Cs^+ removal through biosorption.

Natural polymers and their derivatives have also emerged as cost-effective and eco-friendly adsorbents for removing Cs^+. Notably, chitosan holds significant potential due to its abundance in nature and its structure rich in amino and hydroxyl groups. These functional groups enable effective adsorption and removal of nuclide ions through processes such as coordination complexation and electrostatic interaction (Zhuang et al., 2022b). However, most natural polymers present a low adsorption capacity toward cesium ions due to the limitations imposed by the chemical and physical properties of cesium ions. Cesium ions are large ions with a large ionic radius, low charge density, and strong charge shielding effect, which results in weak interaction with natural polymers. To enhance the adsorption capacity of natural polymers for cesium ions, various modification techniques such as changing their molecular structure or preparing composite adsorbent materials are usually adopted to increase their adsorption capacity and selectivity. For example, Yin et al. (2017) coated yeast and magnetic materials with chitosan. The immobilization of microbial cells with a polymer offers several advantages, including easy separation from the reaction medium, the ability to be used continuously or repeatedly, and increased stability of microorganisms.

3 Membrane separation technology for Cs removal

Membrane separation technology has been rapidly developing in the field of radioactive wastewater treatment due to its ability to provide low energy consumption, high decontamination factor, and compatibility with existing systems. The principle behind this technology is to use selective permeable membranes driven by pressure, temperature, and osmotic pressure to selectively separate and concentrate radioactive nuclides in wastewater. As shown in Table 3, different membrane technologies, such as reverse osmosis (pressure-driven), forward osmosis (osmotic pressure-driven), and membrane distillation (temperature difference-driven), can be utilized for this purpose. Despite the advantages of membrane technology, membrane fouling and possible structural changes may occur after radiation exposure. Nonetheless, the rapid development and use of this technology have made commercial membranes more affordable (Chen et al., 2016).

The radiation stability of membranes is a critical factor in their use for treating radioactive wastewater. Researchers have investigated the radiation stability of membrane materials, such as the work of Arnal et al. (2003) on aromatic polyamide composite reverse osmosis (RO) membranes. This study subjected the membranes to gamma radiation (0–2000 cGy) and electron radiation (0–2500 cGy), and found no structural changes. Although low level radioactive wastewater has minimal impact on membrane materials, there have been no significant reports of negative effects on membranes radiation stability in this type of wastewater.

3.1 Reverse osmosis

Membrane filtration is a separation process that operates on the basis of pore size, allowing for the retention of larger particles while smaller ones pass through. Membrane filtration is divided into four categories as shown in Fig. 2. Pore rejection is a critical factor in membrane separation. As per pore rejection theory, cesium ions can not penetrate RO membranes because the hydrated ionic radius of cesium ion is larger than the pore size, while they could penetrate other membranes. For the separation of cesium ions, RO is a promising direct retention method (Chmielewski et al., 2001; Chen et al., 2015a).

Although other membrane filtration methods, such as microfiltration, ultrafiltration, and nanofiltration, cannot directly retain cesium ions, they can still serve two purposes. First, these methods can be utilized as pretreatment stages to decelerate membrane fouling during the RO stage. Second, cesium ions can be absorbed onto large carriers through the use of flocculants, complexing agents, or polymers, and then retained by microfiltration, ultrafiltration, or nanofiltration to facilitate the separation and removal of cesium ions.

3.2 Forward osmosis

Forward osmosis (FO) is a separation process that has gained attention for its potential in removing Cs^+ from aqueous solutions. As shown in Fig. 3, FO is based on the osmotic pressure gradient across a semi-permeable membrane, where a draw solution with a higher osmotic pressure is used to extract water from the feed solution (Wang and Liu, 2021). This results in the concentration of the target solutes in the feed solution, including Cs^+, while leaving behind the majority of the water and other contaminants (Ge et al., 2013).

Compared to other separation processes, FO has several advantages, including low energy consumption, minimal fouling, and the potential for use with a variety of draw solutions. FO demonstrates comparable efficacy to RO in removing radionuclides. Moreover, FO is an energy-efficient process that does not require high-pressure equipment. When treating radioactive wastewater by FO, the proximity of most nuclear power plants to abundant seawater resources allows for the utilization of seawater as a feed solution. We have conducted a series of experiments to investigate the application of FO for Cs^+ removal (Liu et al., 2018; 2019a; 2019b).

FO performance for Cs^+ separation is affected by various factors, including membrane properties, draw/feed solution, and operational parameters (Liu et al., 2019a). The membrane material is a critical factor. Cellulose triacetate (CTA) or polyamide-based thin film composite (TFC) are commonly used as commercial membrane materials. It was found that the CTA membrane had a higher Cs^+ retention level than that of

Table 3 Membrane separation technology used for cesium removal

Membrane separation technology	Active layer of membrane material	Driving force	Separation mechanism of cesium ions	Decontamination factor (DF)
Reverse osmosis	Polyamide	Pressure	Pore rejection: rejection of Cs^+, while allowing the passage of H_2O	$10–10^3$
Forward osmosis	Cellulose triacetate Polyamide	Osmotic pressure	Pore rejection: Rejection of Cs^+, while allowing the passage of H_2O	–
Membrane distillation	Polypropylene, polyethylene, polytetrafluoroethylene, poly (vinylidene fluoride), Ceramic	Temperature difference	Selective retention of non-volatile Cs^+ ions while allowing the passage of volatile water molecules	DF→∞

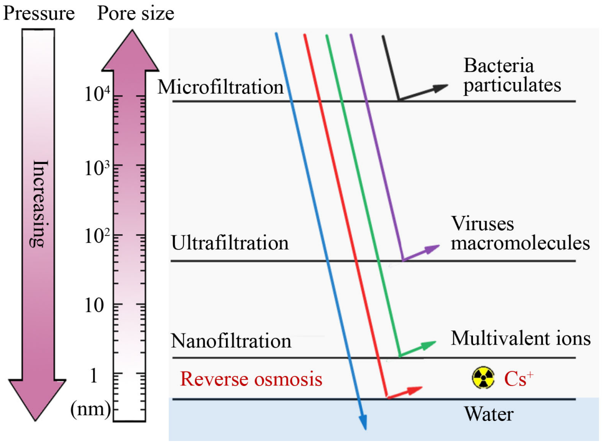

Fig. 2 Diagram of membrane separation technologies (Wang and Zhuang, 2019c).

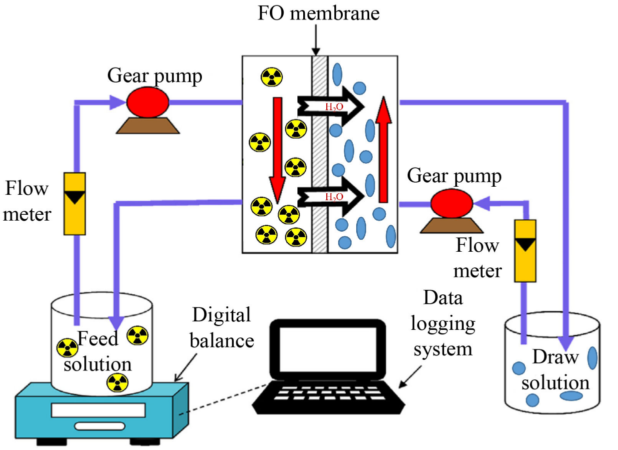

Fig. 3 Diagram of the forward osmosis (FO) system (Liu et al., 2020).

TFC membrane (Liu et al., 2018). Moreover, the Cs^+ retention process in the CTA membrane was minimally affected by the presence of B ions (Liu et al., 2018). Moreover, the support layer of the membrane significantly impacts water flux, and the use of a polyester screen support layer is beneficial in reducing concentration polarization, resulting in higher water flux. Therefore, the cellulose triacetate with embedded polyester screen support membrane shows promise in terms of both high water flux and Cs^+ retention.

Membrane orientation is another crucial factor affecting FO performance. The orientation of the active layer, whether facing the feed solution (AL-FS) or draw solution (AL-DS), affects the water treatment process. The membrane orientation is also influenced by the support layer. CTA with a cast nonwoven support membrane in the AL-FS mode exhibited higher Cs(I) retention than in the AL-DS mode. Conversely, CTA with embedded polyester screen support membrane and TFC with embedded polyester screen support in the AL-DS mode showed higher Cs(I) retention than in the AL-FS mode (Liu et al., 2018).

3.3 Membrane distillation

Membrane distillation (MD) uses a hydrophobic

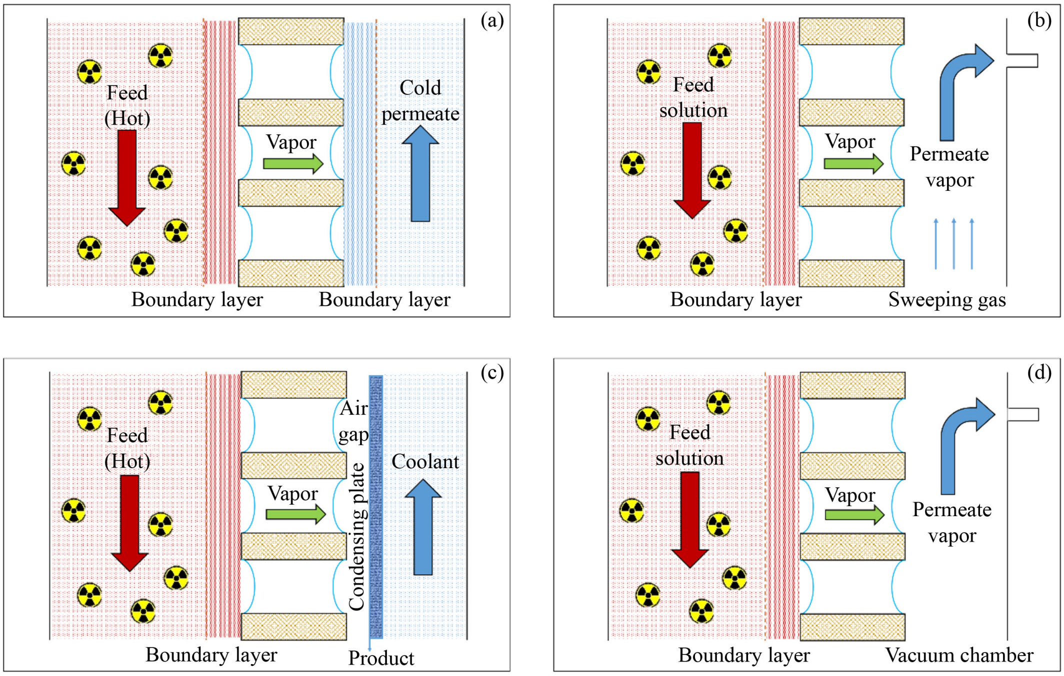

Fig. 4 Diagram of four types of membrane distillation. (a) Direct contact membrane distillation; (b) air gap membrane distillation; (c) sweeping gas membrane distillation; (d) vacuum membrane distillation (Wang and Zhuang, 2019c).

membrane to separate two phases — a hot feed solution and a cold distillate stream (Fig. 4). The separation driving force is the vapor pressure difference across membrane, which allows the transfer of volatile compounds such as water vapor from the hot feed to the cold distillate stream. This technology has been increasingly explored for the removal of radioactive contaminants, including cesium ions.

MD presents numerous advantages in contrast to traditional membrane filtration technologies. The inherent hydrophobicity of the membrane prevents fouling and facilitates the treatment of high-salinity solutions. Additionally, MD can function at lower temperatures and pressures compared to other membrane technologies, thereby reducing energy consumption. Finally, MD exhibits the potential to concentrate the radioactive waste, leading to a smaller volume of waste to be treated (Khayet, 2011).

Some studies have explored the utilization of MD for the separation of cesium ions from various feed solutions, including radioactive wastewater and seawater (Zakrzewska-Trznadel et al., 1999; 2001). These studies have demonstrated that MD can achieve high cesium removal efficiencies of over 99%, making it a promising technology for the treatment of radioactive waste.

A series of work in the field of nuclide separation using MD has also been conducted by us. We have found that direct contact membrane distillation could reject nuclides almost completely (Liu and Wang, 2013). Meanwhile, vacuum membrane distillation has been shown to achieve over 99.76% removal of Cs^+, while maintaining 6.14 L/(m^2·h) of membrane flux during continuous operation (Jia and Wang, 2017).

4 Radionuclides removal from Fukushima Daiichi waste effluents

To cool the meltdown core following the Fukushima Daiichi nuclear power disaster, a significant amount of seawater was continuously injected into the core. This water, along with rainwater and groundwater that comes into contact with or flows through the nuclear reactor, inevitably contains significant amounts of radioactive elements (e.g., ^{137}Cs). Therefore, a substantial volume of radioactive wastewater is continuously generated and stored on the premises in tanks, awaiting treatment.

Several treatment systems have been developed to manage the radioactive wastewater from the Fukushima Daiichi nuclear power plant, including the Kurion system, SARRY system, desalination system, and the Advanced Liquid Processing System (ALPS) (Lehto et al., 2019). Initially, the Kurion and SARRY systems were used, which employed zeolite and silicotitanate as adsorbents respectively, for the removal of radioactive isotopes from the wastewater. The treated wastewater was further processed through a desalination system to meet the requirement for reintroduction into the reactor as cooling water (Sylvester et al., 2013). Subsequently, the ALPS treatment was introduced to reduce the levels of radionuclides to meet the standards for discharging treated water into the ocean. This system consists of multiple sets of adsorption equipment that remove various radioactive isotopes from the wastewater in stages. Among them, Cs adsorption equipment is the most commonly used, utilizing various adsorbents such as zeolite, synthetic zeolite, silicotitanate, hexacyanoferrate, impregnated activated carbon, titanium oxide, and ion exchange resins (IAEA, 2020; 2023).

4.1 Kurion/SARRY system + desalination system

During the initial phases, the Kurion and SARRY systems were employed for the adsorptive elimination of Cs^+ and Sr^{2+} ions from the radioactive wastewater. Subsequently, a desalination system (Fig. 5) was utilized to decrease the concentration of other radioactive nuclides. The ultimate objective of this procedure was to generate cooling water that adhered to the necessary safety regulations and could be reintroduced into the reactor core without any concerns.

Between 2011 and 2017, two distinct systems, known as the Kurion and SARRY systems, were deployed to eliminate radiocesium. The Kurion system utilized zeolite adsorbent, whereas the SARRY system employed silicotitanate adsorbent. Initially, both systems were concurrently operated, but as time progressed, the SARRY system took precedence as the main method for cesium removal, with the Kurion system serving as a backup. Throughout this duration, a total volume of 1.9 million cubic meters of radioactive wastewater underwent treatment, successfully eliminating the primary

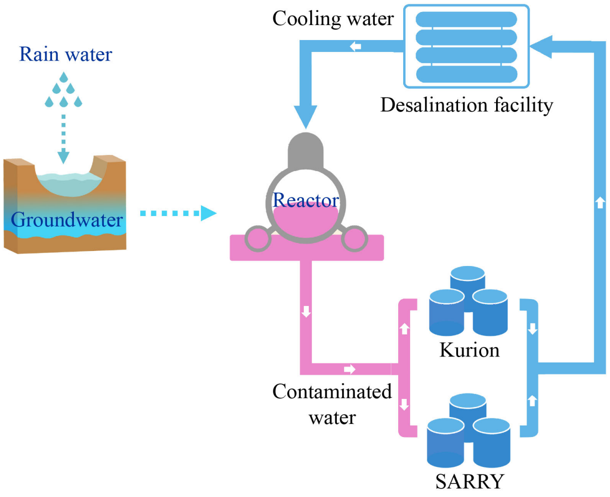

Fig. 5 Schematic diagram of Kurion/SARRY system + desalination system for the treatment of Fukushima Daiichi waste effluents.

radioactive constituent, ^{137}Cs (Lehto et al., 2019).

A desalination device was utilized to remove salt from the processed water, enabling its reintroduction into the core as cooling water. This device was located in a temporary plant that included reverse osmosis and evaporation concentration equipment. As a result of the treatment process, several million tons of reject from the reverse osmosis treatment have been generated and temporarily stored on-site, awaiting final disposal (Sylvester et al., 2013).

4.2 Advanced liquid processing system

After being processed through Kurion/SARRY system and desalination system, the contaminated water still retains various other radioactive nuclides. This issue has arisen due to the extensive storage within the Fukushima plant site, leading to radiation levels at the site boundary that significantly exceed the limits set by the Japanese government. In 2013, the annual dose at the site boundary reached an alarming 9.76 mSv/a, far surpassing the target of below 1 mSv/a. The water storage tanks alone accounted for over 90% of this dose.

To tackle this problem and achieve the prescribed dose levels for the plant site perimeter, as well as further purify the contaminated water to meet the Japanese government's environmental discharge standards, the Fukushima Daiichi Nuclear Power Plant has implemented three sets of ALPS since 2013. These facilities have been specifically designed to treat the contaminated water that has already undergone cesium purification and desalination, with the aim of eliminating 62 additional radioactive nuclides apart from tritium (IAEA, 2022).

Table 4 lists the key parameters of ALPS. Out of the three sets of ALPS implemented, only two of the initially installed systems are currently operational. The first ALPS system, launched in March 2013, consists of three stages: pretreatment, adsorption, and end-polishing. Equipped with three parallel units, this system has a processing capacity of up to 750 m^3/d. The second ALPS system, installed in September 2014, closely resembles the first one, with the exception of the removal of ferric hydroxide co-precipitation and the addition of two additional sorbent beds. Unfortunately, the third ALPS system, constructed in October 2014, was only operational for a period of 1.5 years. Based on reports and operational data evaluating the performance of the ALPS device, it is evident that the currently deployed ALPS equipment exhibits notable efficacy in removing typical nuclides such as Cs-137, Sr-90, Co-60, and I-129 in the majority of situations.

Figure 6 presents the flow chart outlining the operation of the ALPS system. This system comprises a pre-treatment unit and a multi-nuclide removal unit (IAEA, 2020). The pre-treatment unit incorporates iron and carbonate precipitation devices, effectively eliminating alpha nuclides, ^{60}Co, and competing cations such as Mg^{2+} and Ca^{2+} (Lehto et al., 2019). The remaining radioactive nuclides are subsequently extracted through a series of interconnected adsorption towers that contain inorganic ion exchangers, activated carbons, chelating organic resins, and hybrid organic/inorganic sorbents. According to previous reports, seven different types of sorbents were used in this phase. The cesium-selective adsorbent (hexacyanoferrate) and strontium-selective adsorbent (titanate) are considered critical components of the ALPS system. Once the contaminated water undergoes purification via the ALPS system, the majority of radioactive nuclides, excluding tritium, are successfully removed. The purified water is then stored in tanks within the ALPS facility. Additionally, the sludge and used adsorbents generated throughout the purification process are carefully contained within high-integrity containers (HIC). Japanese authorities have proposed releasing the ALPS-treated water into the ocean after further dilution. Between August 24th and September 11th, 2023, Japan discharged approximately 7800 t of treated wastewater using the ALPS, followed by dilution, into the ocean. The second discharge operation commenced on October 5th, 2023, with an anticipated volume of around 7800 tons, matching the quantity released during the initial phase. As a considerable amount of data regarding Fukushima Daiichi waste effluents remains undisclosed, there is an ongoing and substantial public concern.

5 Concluding remarks and prospects

Removing radioactive cesium ions is a vital but challenging task. On the one hand, radiocesium is a crucial fission product of uranium and widely present in radioactive wastewater, and its presence can cause harmful radiation. On the other hand, the small hydrated radius and highly reactive chemical behavior of cesium ions, along with their similarity to Na^+ and K^+, present significant difficulties for their selective removal from

Table 4 Key parameters comparison of three sets of Advanced Liquid Processing System (ALPS)

Systems	Operation time	Parallel units	Processing capacity (m^3/d/unit)	Pretreatment method	Number of adsorption towers	Removal capability
First ALPS	Mar 2013–	3	250	Coagulation and Precipitation	14 + 2 stages	62 nuclides, below detection limit
Second ALPS	Sept 2014–	3	250	Coagulation and Precipitation	18 stages	
Third ALPS	Oct 2014–Mar 2016	1	500	Filtration	20 stages	

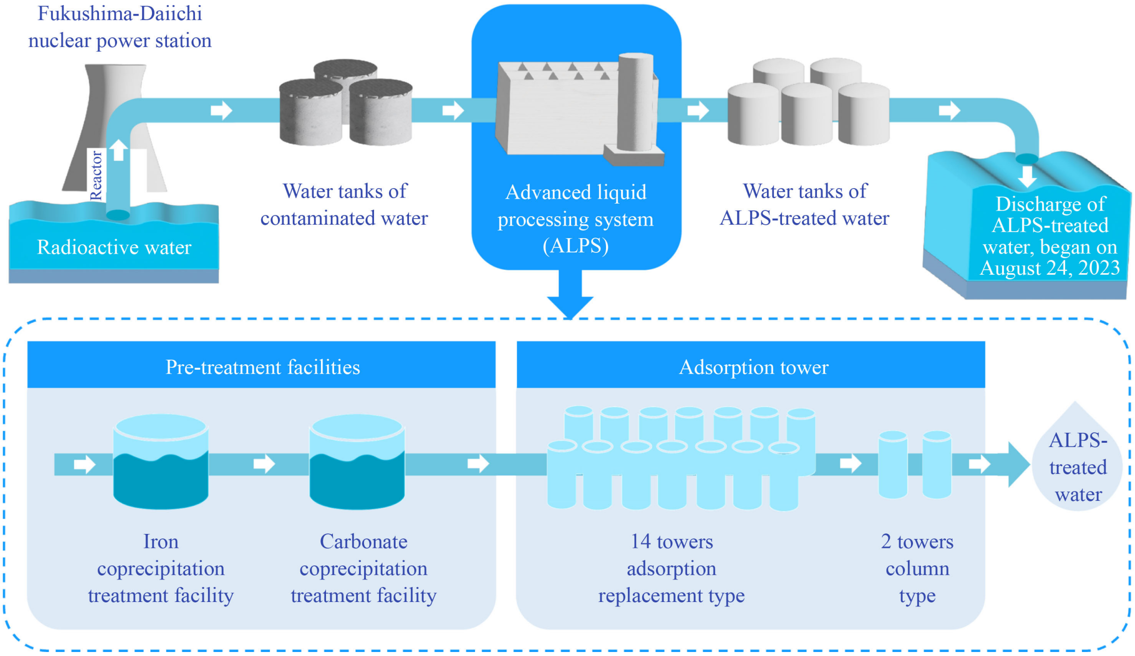

Fig. 6 Flow chart of the ALPS used in Fukushima Daiichi nuclear power plant.

aqueous solutions. Therefore, innovative and efficient techniques for Cs^+ removal from radioactive wastewater are essential.

Adsorption stands out as a remarkably favorable technique for treating radioactive wastewater that contains low concentrations of radionuclides but spans a significant volume. Presently, the primary emphasis lies in the advancement of adsorbents that are both highly efficient and cost-effective for the removal of Cs^+. Numerous inorganic (e.g., hexacyanoferrates, carbon-based materials, clay minerals, geopolymers, MOFs, and others), organic (e.g., resins, Cs^+-imprinted polymers, macrocyclic ligands based adsorbents, and others), and biological materials (microbes, industrial and agricultural wastes, and biopolymers) have been attempted for Cs^+ removal, with promising results. Among them, hexacyanoferrate is widely recognized for its high adsorption capacity and selectivity for Cs^+. To enhance the adsorption selectivity of cesium ions, two approaches can be pursued: Firstly, by leveraging the coordination complex adsorption mechanism, innovative adsorbents with highly specific ligands can be developed. Secondly, by harnessing emerging porous materials such as MOFs and COFs, adsorbent materials with suitable pore sizes and environments can be synthesized, enabling the exchange of cesium ions with other ions within the pores. Moving forward, there remains a need to advance the development of adsorbent materials that possess exceptional adsorption capacity, selectivity, and resilience to radiation. Additionally, it is imperative to delve into the intricate physicochemical reactions and interfacial separation dynamics of Cs^+ within adsorbents. This entails comprehensive investigations into its behavior within complex systems and a thorough exploration of its adsorption mechanisms. This exploration will provide a scientific basis and technical support for developing new technologies to treat wastewater containing Cs^+.

Membrane separation is also an effective method for separating cesium ions, with reverse osmosis (RO) being one of the most commonly used techniques. RO membranes with smaller pore sizes can be directly used for the efficient retention of cesium ions. RO has several advantages, such as high commercialization level, good efficiency for cesium ion retention, and high water fluxes, making it the mainstream membrane technology used in the Fukushima Daiichi nuclear disaster. Other membrane separation techniques have also been explored for cesium ion separation, such as membrane distillation and forward osmosis. These two technologies have been shown to be capable of selectively separating cesium ions at the laboratory level, but further exploration and practical applications are still needed. Despite of these advantages, the membrane method still generates concentrated retention liquid that needs to be treated, with higher concentrations of radioactive nuclides. In addition, prolonged operation poses certain challenges to the radiation stability of the membrane materials.

The treatment of nuclear waste effluents at the Fukushima Daiichi plant has been ongoing since 2011 and is expected to continue for many years. The removal of the major radioactivity component, ^{137}Cs, has been a critical and challenging focus of the treatment efforts. To address this issue, several treatment systems have been developed, including the Kurion system, SARRY system, desalination system, and the ALPS. Currently, ALPS plays a significant role in the treatment process, using a range of adsorption materials to remove radionuclides, excluding tritium, to meet the regulatory standards for discharge. Hexacyanoferrate is the primary adsorption material used for the removal of cesium ions. Starting from August 23, 2023, the discharge of Fukushima nuclear contaminated water into the ocean will continue for approximately 30 years. Up to this point, many details regarding the treatment of Japan's Fukushima nuclear wastewater remain undisclosed, which has also raised concerns in the international community about the stability and effectiveness of this system.

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