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Advances in Membrane Adsorption Techniques for the Removal of Radioactive Materials

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Abstract: *Radioactive wastewater treatment is a critical environmental and operational challenge for nuclear power facilities, medical isotope production, and fuel-cycle activities. This review provides a comprehensive assessment of membrane processes, adsorption technologies, and hybrid membrane-adsorption systems for the efficient removal of radionuclides from complex wastewater matrices. The fundamental mechanisms governing radionuclide retention-including size exclusion, electrostatic interactions, surface complexation, and ion exchange-are examined to elucidate the performance of microfiltration, ultrafiltration, nanofiltration, reverse osmosis, and advanced engineered membranes. Special emphasis is placed on hybrid treatment configurations that integrate adsorption, electrochemical processes, or multi-stage membrane operations, which collectively enhance removal efficiency, reduce fouling, and minimize secondary waste generation. Real-world applications from large-scale facilities, such as Fukushima Daiichi, the Bruce Nuclear Power Plant, and the Savannah River Site, are reviewed to demonstrate practical feasibility and operational constraints. Key performance indicators—removal efficiency, adsorption capacity, ion selectivity, and regenerability-are assessed to identify strategies for effective radionuclide retention, highlighting the promise of advanced membranes, high-affinity adsorbents, and hybrid systems for scalable, sustainable radioactive wastewater treatment.*

Keywords: *Continuous radionuclide removal, functionalized membranes, graphene oxide, hybrid systems, membrane adsorption, radioactive wastewater, titanate materials*

1. Introduction

Radioactive materials are substances that contain unstable nuclei capable of undergoing spontaneous decay and emitting ionizing radiation in the form of alpha, beta or gamma emissions. When these materials accumulate in environmental media-particularly water systems-they form radioactive waste, which represents one of the most persistent and hazardous classes of pollutants. Radioactive waste is generally categorized according to origin and activity level, including very low level waste (VLLW), comprising waste that can be safely disposed of alongside municipal, commercial or industrial waste, or can be disposed of to specified landfill sites, low-level waste (LLW) from scrap metal items, paper, plastics, and medical diagnostics, intermediate-level waste (ILW) from industrial and research facilities, and high-level waste (HLW) generated during nuclear fuel reprocessing and power production [1,2].

Soils and rocks naturally contain low levels of uranium, thorium, and their decay products (thorium and uranium concentrations in rocks are typically below 0.1%) [3,4]. A third important natural radioelement is the potassium isotope ^{40}K. Although ^{40}K accounts for only 0.0118% of total natural potassium, it contributes significantly to environmental radioactivity due to the relatively high abundance of potassium in rocks (approximately 2.5% of the Earth's crust) [5].

Radium-226(^{226}Ra), a decay product of uranium (^{238}U) and thorium-230 (^{232}Th), is a radionuclide of particular concern in the environment. Groundwaters generally contain much higher concentrations of ^{226}Ra than surface waters [6]. In contrast, oceans exhibit comparatively low ^{226}Ra levels, with activity concentrations ranging between 4 and 19 Bq/m^3 [7]. Several studies indicate that, depending on geological settings, average radium concentrations in groundwater can range from 50 to 900 Bq/m^3 [6]. ^{226}Ra decays to radon-222 (^{222}Rn), which can remain dissolved in water.

Groundwater often contains a wide range of dissolved ^{222}Rn, from 1 to 1000 kBq/m^3. ^{226}Ra is also used as a target material to produce actinium-225 (^{225}Ac) via nuclear reactions.

Globally, many drinking water sources contain measurable levels of radium, radon, and uranium isotopes. In the European Union, the key legislative framework for controlling radionuclides in drinking water is Council Directive 2013/51/Euratom, which requires Member States to take corrective actions when prescribed parametric values are exceeded [8]. Distinct regulatory approaches are implemented in regions outside Europe [9]. Table 1 summarizes the drinking water quality guidelines applied in the United States and Europe for controlling ionizing radiation.

Table 1: Water quality guidelines regarding radionuclides in the US, and Europe [8,9]

United States	**Radionuclides**	**Maximum Contaminant Level (Bq/m^3)**
	Combined Radium-226 and Radium-228	185
	Gross Alpha (excluding 222Rn and uranium)	555
	Uranium	0.372
Europe	**Nuclide**	**Derived Concentration (Bq/m^3)**
	Radium-226	500
	Radium-228	200
	Lead-210	200
	Uranium-234	2800
	Uranium-238	3000
	Polonium-210	100

The primary anthropogenic sources of radionuclide contamination include nuclear power generation, mining and milling of uranium, radiopharmaceutical production, nuclear medicine applications (e.g., iodine (^{131}I), technetium (^{99}Tc)), laboratory-scale research activities, and accidental releases associated with reactor failures or improper waste management practices [10].

Radionuclides can persist for long periods due to their physicochemical stability and, in some cases, high solubility, thereby increasing the potential exposure risks to living organisms and the environment [11-14]. Chronic exposure, even at low concentrations, can induce genotoxicity, bioaccumulation in plants [15] and aquatic organisms, disruption of biogeochemical cycles, and long-term soil and groundwater degradation. In humans, internalized radionuclides may deposit preferentially in specific organs (e.g., Sr in bone tissue, Cs in soft tissues), where they cause cellular damage, increase cancer risk, and potentially lead to systemic toxicity [16-18]. The mobility of radioactive isotopes necessitates efficient, selective, and scalable remediation, as conventional methods—precipitation, ion exchange, evaporation, and solidification—suffer from low selectivity, high energy use, sludge issues, and poor performance in dilute or complex systems (Fig. 1) [14].

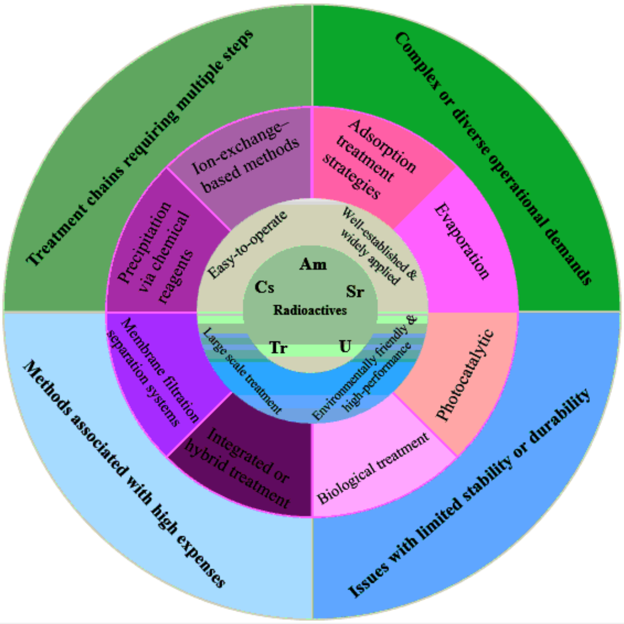

Fig. 1. Schematic illustration of the main challenges and treatment strategies associated with radionuclides

These constraints have accelerated research interest in membrane-based technologies, particularly membrane adsorption systems that integrate the hydraulic advantages of pressure-driven membranes with the physicochemical selectivity of functionalized surfaces [19-21].

In this context, membrane adsorption has emerged as a promising hybrid platform for radionuclide removal from contaminated water. By combining size-based separation with surface-mediated interactions such as adsorption, ion exchange, and tailored surface functionalization, these systems offer enhanced selectivity, reduced secondary waste, and compatibility with continuous-flow operation. As membrane architectures-from RO, NF, and UF to ceramic, polymeric, and hybrid nanostructured designs-continue to evolve, membrane adsorption is emerging as a strong option for sustainable nuclear wastewater management.

2. Fundamentals of Membrane-Based Processes in Radionuclide Treatment

Membrane-based processes have become essential technologies for the treatment of radionuclide-contaminated waters due to their operational flexibility, scalability, and ability to efficiently handle both dilute and complex aqueous matrices. Unlike conventional separation methods, membrane processes rely primarily on pressure-driven transport and physicochemical selectivity determined by pore structure, charge, and surface properties of the membrane material. Although size exclusion is the dominant mechanism in many membrane operations, the separation of radionuclides is also strongly affected by electrostatic interactions, hydration effects, and, in some cases, specific affinity of functional groups incorporated into advanced membrane materials. The overall performance of these processes depends on membrane type (RO, NF, UF, MF), membrane material (polymeric, ceramic, hybrid), and key operational conditions such as pH, ionic strength, transmembrane pressure, and the presence of competing ions. Nanofiltration (NF) membranes are sensitive to ionic charge and electrostatics, reverse osmosis (RO) membranes separate mainly by hydrated ion size, and ultrafiltration (UF) and microfiltration (MF) effectively remove colloidal or particulate.

2.1. Membrane Materials Used in Radionuclide Treatment

Membrane materials play a decisive role in determining radionuclide retention efficiency, chemical stability, fouling resistance, and long-term operational reliability in radioactive wastewater treatment systems. Accordingly, ceramic, polymeric, and hybrid/composite membrane platforms offer distinct structural and chemical characteristics, which directly influence their suitability for functional modification and selective radionuclide removal. These material-dependent properties ultimately dictate how effectively membranes can be engineered or enhanced to target specific radionuclide species under complex water chemistries. In radioactive environments, particularly those involving high-energy alpha, beta, or gamma emissions, polymeric membranes may undergo radiolytic degradation, which can compromise their mechanical integrity, chemical stability, and long-term operational performance. In recent years, nanocomposite membranes-formed by incorporating nanoparticles, metal oxides, carbon-based nanomaterials, or functional nanofillers into polymeric or ceramic matrices-have gained considerable interest due to their enhanced selectivity, radiation resistance, and surface reactivity toward radionuclide species.

Binkle et al. developed a ceramic-membrane-based purification method for radionuclide-contaminated waters. This work discloses a novel method for purifying water containing radioactive nuclides-particularly radium-bearing groundwater-through a two-stage system comprising chemical pre-treatment and filtration via at least one ceramic membrane. In the pre-treatment phase, finely precipitated manganese dioxide (MnO_2) is either introduced or generated in situ to adsorb and immobilize radionuclides and associated heavy metal ions. Thereafter, the chemically conditioned feed is passed through a ceramic filter membrane, wherein the particulate MnO_2-bound radionuclides and colloids are separated. The document further describes a preferred pore size range of the ceramic separation layer (80-800 nm, particularly 100-300 nm), and a modular membrane unit designed for robust long-term operation under harsh conditions including high acidity/basicity and radioactivity. Unique advantages of this approach include reduced secondary waste volume (compared to sand filters), lower long-term fouling, and the capability to either bypass or follow up with pressure-driven membrane separation (e.g., RO) as necessary. The method therefore introduces ceramic filter membranes as a viable solution for radionuclide removal in high-load or extreme environments within the nuclear water-treatment domain [22].

2.1.1. Organic (Polymeric) Membranes

Polymeric membranes, including polyamide (PA), polysulfone (PSf), polyethersulfone (PES), polyvinylidene fluoride (PVDF), and polyacrylonitrile (PAN)-based structures, remain the most widely used materials due to their cost-effectiveness, ease of fabrication, and compatibility with pressure-driven processes such as RO, NF, UF, and MF. Although polymers are generally less stable under high-radiation conditions compared to ceramics, advances in cross-linking, surface modification, and antioxidant incorporation have improved their resistance to radiation-induced degradation. Polymeric membranes offer considerable flexibility for surface functionalization, allowing immobilization of ion-exchange groups, chelating ligands, or nanoparticle adsorbents to improve selectivity toward radionuclides such as Cs^+, Sr^{2+}, UO_2^{2+}, and Co^{2+}.

Bolisetty et al. reported a novel hybrid membrane composed of amyloid fibrils derived from inexpensive proteins and activated carbon, designed to treat radioactive wastewater from clinical and nuclear sources. In their work, the authors demonstrated the remarkable removal of radionuclides including Tc-99m, I-123 and Ga-68 with efficiencies above 99.8% in a single-step filtration. The removal mechanism was attributed to a combination of steric trapping within the amyloid network and strong adsorptive interactions offered by activated carbon. They further applied the membrane to a real hospital wastewater sample containing I-131 and Lu-177, using SPECT/PET imaging to confirm the accumulation of the radionuclides within the membrane structure. Their findings highlight the potential of converting large volumes of liquid radioactive waste into much smaller volumes of solidified material and suggest a promising, scalable strategy for nuclear or hospital wastewater management [23].

Kim et al. evaluated the applicability of commercial polymeric desalination membranes for the removal of critical radionuclides-Cs^+, Sr^{2+}, and Co^{2+} from saline aqueous systems. In their study, thin-film composite (TFC) polyamide nanofiltration (NF) and reverse osmosis (RO) membranes were systematically characterized using FT-IR/ATR spectroscopy, thermogravimetric analysis, and SEM imaging to elucidate structural and chemical features influencing ion rejection. Performance tests conducted with single-solute and mixed-solute feed solutions demonstrated that RO membranes achieved consistently high nuclide rejection (>93% in single-ion feeds and >98% in mixed feeds), whereas NF membranes provided substantially higher water flux (exceeding 230 $L \cdot m^{-2} \cdot h^{-1}$) but comparatively lower radionuclide retention. The observed differences in performance were linked to the stronger steric and hydration-radius–dependent exclusion in RO membranes, whereas NF membranes exhibited charge-regulated transport behavior. These findings indicate that commercially available polyamide desalination membranes are strong candidates for nuclear wastewater treatment, especially when high-efficiency removal of multivalent radionuclides is needed [24].

Sancho et al. tested the feasibility of a two-stage membrane process-ultrafiltration (UF) followed by reverse osmosis (RO) - to remove both organic matter and residual ^{125}I from real hospital RIA waste streams. Their results demonstrated that the integrated UF-RO configuration is capable of producing an effluent that meets operational and radiological discharge criteria, while also reducing waste volume for final disposal. The UF step is particularly advantageous for RIA wastes, which typically contain proteins, antibodies, and organic macromolecules that would otherwise accelerate RO fouling. Overall, the authors conclude that UF-RO membrane treatment represents a technically feasible and operationally suitable solution for handling low-activity ^{125}I-containing hospital wastes, addressing both regulatory requirements and the specific chemical–biological complexity of RIA effluents [25]. Consequently, these studies demonstrate that polymeric membranes-when appropriately modified or configured-can achieve high radionuclide removal efficiencies across diverse nuclear and clinical wastewater matrices.

2.1.2. Hybrid and Composite Membranes

Hybrid or composite membranes integrate the structural advantages of polymers with the functional properties of inorganic materials. These membranes may include mixed-matrix membranes (MMMs) containing adsorbents (e.g., titanates, zeolites, Prussian blue, graphene oxide), thin-film nanocomposites (TFNs) embedding nanoparticles within polyamide layers, or mixed matrix membranes (inorganic–organic composites) prepared via sol-gel processes. Hybrid membranes provide enhanced mechanical strength, higher adsorption capacity, and tailored selectivity for various radionuclides. Their multifunctional nature enables simultaneous size-based separation and affinity-driven adsorption, making them highly attractive for nuclear wastewater remediation. Taken together, these attributes-combined with promising laboratory-scale performance-suggest that hybrid/composite systems hold strong potential for future implementation, despite the fact that many are still at the experimental stage and have not yet been fully validated using real radioactive waste streams. Yeszhanov et al. developed hybrid membranes by depositing polyvinyl chloride (PVC) nanofibers**,** produced via electrospinning, onto polyethylene terephthalate (PET) track-etched membranes (TeMs). The resulting hybrid membrane demonstrated excellent performance in direct-contact membrane distillation (DCMD) for the treatment of low-level liquid radioactive wastes and saline solutions, maintaining high vapor fluxes and stable radionuclide rejection due to its reinforced structure. Additionally, the enhanced surface roughness and hydrophobicity supported efficient water–oil separation, illustrating the versatility of hybrid membrane architectures for complex wastewater streams [26]. Zhang et al. developed a composite molybdenum disulfide (MoS_2)/polyethylenimine (PEI) nanofiltration membrane specifically engineered for the efficient removal of radionuclides from contaminated water. The membrane was fabricated by integrating two-dimensional MoS_2 nanosheets into a PEI matrix on a porous support, creating a hybrid selective layer with enhanced charge density and abundant functional groups. The study demonstrated that MoS_2 nanosheets significantly improved membrane hydrophilicity, surface charge, and adsorption affinity toward metal ions, while PEI contributed amine-rich coordination sites capable of binding positively charged radionuclides. As a result, the MoS_2/PEI NF membrane exhibited high rejection of key radionuclides, including Cs^+, and Sr^{2+}, driven by a synergistic combination of adsorptive interactions (coordination, electrostatic attraction) and size-based nanofiltration. Performance testing under a range of pH, ionic strength, and competing-ion conditions showed that the membrane maintained strong stability and selectivity, indicating its suitability for real wastewater environments. Furthermore, the membrane demonstrated good antifouling behavior and reusability over multiple filtration cycles. Overall, these findings highlight MoS_2/PEI composite nanofiltration membranes as promising next-generation candidates for radionuclide sequestration in aquatic systems [27].

In practical applications, further investigation is required regarding the ability of membrane materials to withstand harsh environments. Under relatively high levels of radioactivity, the surface structure of the membrane will inevitably deteriorate, leading to performance degradation and reduced service life. Therefore, to overcome these issues and further advance the treatment of radioactive wastewater, it is necessary, on one hand, to optimize process parameters, improve process flow, and reduce contact time; on the other hand, research and development of antifouling membranes, ceramic membranes, and similar advanced materials should also be considered. Azman et al. developed a positively charged polyamide reverse osmosis (RO) membrane using a polyethyleneimine (PEI) spray-coating technique to enhance the rejection of radionuclides in water treatment. The study demonstrated that the PEI-modified RO membrane exhibited improved surface charge density, higher hydrophilicity, and enhanced structural stability under operational conditions. As a result, the modified membrane achieved significantly greater removal efficiencies for various radionuclide ions compared to the unmodified polyamide membrane. Their findings indicate that surface-engineered RO membranes can provide more effective radionuclide separation, offering a promising pathway for improving membrane performance in radioactive wastewater treatment [28].

2.2. Mechanisms of Radionuclide Treatment

The removal of radionuclides by membrane processes is governed by a combination of physical, chemical, and electrostatic interactions occurring at the membrane-solution-ion interface. In pressure-driven membrane systems (MF, UF, NF, and RO), separation efficiency is determined not only by physical parameters such as pore size and hydraulic pressure, but also by ion valence, hydration radius, membrane surface charge, and the interactions between radionuclide species and membrane functional groups. The mechanisms responsible for radionuclide removal can generally be classified into four major categories:

- Size Exclusion: Dominant in MF and UF, this mechanism relies on the inability of suspended solids, colloidal particles, and radionuclide-colloid complexes to pass through membrane pores. It is particularly effective for particle-associated radionuclides such as ^{137}Cs, ^{60}Co, and ^{90}Sr.
- Charge-Based Exclusion (Donnan Interactions): Electrostatic exclusion, which plays a key role in NF and some RO membranes, depends on repulsive or attractive interactions between membrane surface charge and ionic species. In NF membranes, the rejection of divalent and multivalent ions is strongly influenced by solution pH and ionic strength, whereas in RO membranes the hydration radius becomes a key controlling factor for ion transport. Positively charged membrane surfaces can enhance the retention of cationic radionuclides (e.g., Am^{3+}, UO_2^{2+}, Sr^{2+}). This mechanism is especially significant in polyamide-based NF/RO membranes.
- Adsorption and Surface Complexation: Radionuclide ions may adsorb onto membrane surfaces or form complexes with functional groups such as –COOH, –OH, $–NH_2$. This mechanism contributes to the removal of radionuclides even at low concentrations. It is particularly relevant in metal oxide-coated inorganic membranes (ZrO_2, TiO_2, Al_2O_3), where strong surface complexation and inner-sphere complex formation occur between radionuclides and metal-oxide functional groups.
- Ion Exchange Interactions: Certain advanced or hybrid membranes exhibit ion exchange functionality, allowing radionuclide ions to replace exchangeable ions within the membrane matrix. This mechanism is particularly important for high selectiv removal of species that require ity, such as U(VI), Th(IV), and Ra^{2+}, and is commonly incorporated via grafted chelating ligands, sulfonic groups, or amine-rich polymers.

The relative contribution of each mechanism varies depending on membrane type, radionuclide characteristics, solution chemistry (e.g., pH, ionic strength), and interfacial reactions occurring at the membrane surface. Therefore, achieving optimal radionuclide removal requires careful integration of operational parameters (pressure, pH, pretreatment steps) with membrane material design (hydrophilicity, surface charge, functionalization). Additionally, in radioactive environments, radiolytic degradation of polymeric membranes can significantly alter surface functional groups and pore morphology, ultimately influencing these mechanisms and reducing long-term performance. Recognizing and mitigating this degradation pathway is essential for reliable membrane operation in nuclear wastewater treatment.

2.3. Types of Pressure-Driven Membrane Processes Used in Radionuclide Treatment

The membrane process can be selected according to the composition of the radioactive liquid waste, the speciation of radionuclides in solution, and the type of separation membrane employed. Based on differences in pore size, membranes can be classified into several categories, including reverse osmosis, ultrafiltration, nanofiltration, and microfiltration.

2.3.1. Microfiltration (MF)

Microfiltration (MF) membranes retain larger particles or macromolecules in the size range of approximately 0.1-1 µm [29]. In nuclear wastewater treatment, MF is typically applied as a pretreatment or clarification step to remove coarse particulate matter formed after chemical precipitation. Under pressure gradient, particles larger than the pore size of the membrane are retained, enabling efficient solid-liquid separation. Due to their relatively large pore size, MF membranes primarily remove suspended solids and colloidal aggregates rather than dissolved radionuclide ions. Consequently, MF alone is generally insufficient for direct radionuclide separation and must be integrated with additional treatment processes [30]. In nuclear power plants, MF is commonly used for pretreatment or for concentrating particulate matter generated after precipitation. For high-level radioactive wastes, ceramic MF membranes are preferred because their robust structure allows removal of radioactive particles with high decontamination and concentration factors. The performance of MF membranes has been demonstrated in multiple nuclear facilities, confirming their reliability in removing suspended particulates prior to downstream polishing processes. Zhao et al. treated low-level plutonium-containing wastewater using a combined flocculation-sedimentation-microfiltration process. By optimizing ferrous sulfate dosage and adjusting solution pH, a plutonium removal efficiency exceeding 99.9% was achieved.

Furthermore, for mixed wastewaters containing uranium, americium, and plutonium, the coupled flocculation-MF system achieved a single-stage total α-emitter removal efficiency of 99.87% [31].

2.3.2. Nanofiltration (NF)

NF membranes typically contain nanopores on the order of 1-2 nm and exhibit moderately charged surfaces functionalities [29]. Unlike other pressure-driven membrane processes, NF includes through both convective and diffusive transport while also incorporating an electrostatic separation mechanism-primarily Donnan exclusion- arising from the dissociation of active charged groups on the membrane surface upon contact with aqueous environments. As a result, NF membranes retain multivalent ions far more effectively than monovalent ions, making it particularly suitable for radionuclide-containing waste streams where many species exist in multi-charged ionic or complexed forms. NF is widely used in nuclear power plants for the recovery of boric acid and the removal of organic contaminants from contaminated cooling circuits. In addition to its recyclability benefits, boric acid facilitates the separation of divalent radionuclides present in reactor wastewater. NF membranes have therefore been employed to selectively remove divalent fission products-such as ^{60}Co and ^{90}Sr from nuclear effluents. The separation mechanism relies on controlling the surface charge of the NF membrane, which is governed by the solution pH, thereby enhancing electrostatic exclusion of the targeted radionuclide ions.

Kaptakov et al. investigated NF efficiency for extracting ^{238}U and ^{232}Th from aqueous solutions under controlled physicochemical conditions. The study evaluated the influence of pH, ionic strength, and operating pressure on radionuclide rejection. Their results showed that NF membranes exhibit significantly higher retention of multivalent actinide ions compared to monovalent salts due to combined effects of steric hindrance and Donnan exclusion. Uranium and thorium were removed with rejection efficiencies typically exceeding 95% at mildly acidic to neutral pH, where both species exist predominantly as multivalent hydroxo complexes. Although increasing ionic strength slightly decreased rejection due to charge screening separation remained effective. The study concluded that NF is a viable pretreatment or polishing step for liquid radioactive waste streams containing ^{238}U and ^{232}Th, offering high selectivity, relatively low energy demand, and operational simplicity [32].

Kaptakov and Milyutin evaluated polymeric NF membranes for treating real radioactive waste streams, including evaporator bottom residues from the Kola Nuclear Power Plant (Kola NPP) and Beloyarsk Nuclear Power Plant (Beloyarsk NPP), as well as low-level wastewater from the Frumkin Institute of Physical Chemistry & Electrochemistry of the Russian Academy of Sciences. A polymer NF membrane (NanoNF-1812) operated at 6.5-7.0 atm in circulation mode was applied. For the Kola NPP bottoms, single-stage NF achieved a purification coefficient of ~2.8 for ^{60}Co while ^{137}Cs was essentially unretained. In contrast, a five-stage NF scheme at Beloyarsk NPP achieved a purification coefficient of ~388 for ^{60}Co. In testing the low-level wastewaters, the NF process achieved removal rates of ~75% for ^{137}Cs, ~91% for ^{90}Sr, ~99.5% for ^{241}Am, ~91% for ^{152}Eu and ~99.5% for ^{239}Pu. The authors concluded that NF is highly effective for radionuclides present as colloidal species or multivalent ions, but less effective for monovalent ions such as Cs^+. Moreover, combining NF with selective adsorption (e.g., using a ferrocyanide sorbent Temoksid-35 for Cs) can generate a final salt residue that no longer qualifies as radioactive waste under regulatory standards [33].

2.3.3. Ultrafiltration (UF)

Ultrafiltration (UF) membranes typically possess pore sizes ranging from 0.001 to 0.1 μm, allowing dissolved solutes to permeate while retaining colloidal and suspended particles [29]. UF is widely used as a pretreatment step prior to reverse osmosis and can be effectively integrated with adsorption, precipitation, or complexation processes to enhance separation efficiency. UF membranes are extensively used in the treatment of contaminated wastewater generated from nuclear research laboratories, medical facilities, and the nuclear industry operations. Radioactive liquid wastes arising from various processes in nuclear power plants generally display a complex matrix containing organic and inorganic substances, colloids, suspended particles, and dissolved radionuclide ions. This complexity necessitates membrane-based separation mechanisms capable of selectively retaining heterogeneous particulate and macromolecular fractions.

Zhang et al. investigated the effect of low concentrations of cationic surfactants on metal-ion removal during UF. Their results demonstrated that when the concentration of CTAB was maintained below the critical micelle concentration, the removal efficiency of Cs^+ increased from 24-33% to approximately 50%, whereas the rejection of Sr^{2+}, Co^{2+}, and Ag^+ exceeded 90% [34].

Yu et al. evaluated the feasibility of using ultrafiltration for the removal of silver colloids from radioactive waste streams through a series of laboratory-scale filtration experiments. The authors characterized the colloidal fraction present in both model and/or real radioactive effluents and subsequently examined the retention performance and hydraulic behavior of an UF stage under the tested operational conditions. The study reports that UF achieved >98% removal of silver colloids, demonstrating the strong capability of UF membranes to retain particle-bound radionuclide species. Notably, a substantial increase in permeate flux (≈146.9%) was observed during the optimized runs, indicating favorable hydraulic performance under the selected operating regime. These findings support the application of UF as an efficient physical barrier for colloid-associated radionuclides and highlight the need for further pilot-scale investigations to evaluate long-term stability, fouling behaviour and integration options with downstream polishing units [35].

Similarly, numerous studies on micellar-enhanced ultrafiltration (MEUF) for heavy-metal removal-including applications to Pb(II), Cu(II), Cd(II), and Zn(II)-demonstrate that surfactant–metal complexation combined with UF provides high selectivity and enhanced rejection, offering a strong scientific basis for adapting these mechanisms to radionuclide separation processes [36,37].

2.3.4. Reverse Osmosis (RO)

Reverse osmosis (RO) is a pressure-driven separation process in which a transmembrane pressure gradient forces the solvent to permeate through the membrane while rejecting dissolved solutes. RO membranes exhibit strong retention performance toward a broad spectrum of inorganic ions and are widely applied for the concentration and purification of contaminated aqueous streams [29]. As an alternative to ion exchange and evaporation, RO has been used for the treatment of radioactive wastewater since the 1980s; however, many of these applications remain classified or otherwise inaccessible. This constraint is also emphasized in the International Atomic Energy Agency (IAEA) report addressing membrane-based treatment of liquid radioactive waste [38]. Radionuclides such as ^{90}Sr and ^{137}Cs have been successfully removed using RO membranes, achieving ion rejection efficiencies exceeding 99.7%. Ahmad et al. developed a thin-film composite RO (TFC-RO) membrane modified with spray-coated titania nanosheets (TNS) to improve long-term stability and antifouling performance during radionuclide-containing wastewater treatment. In their approach, a bilayer of oppositely charged titania nanosheets was uniformly deposited onto the polyamide selective layer, enhancing both physicochemical resilience and separation efficiency. Using cesium ions (Cs^+) as a model radionuclide, the modified membrane exhibited substantially increased water permeability-up to 1.53 $L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$, corresponding to a 45.8% improvement over pristine TFC-and near-complete Cs^+ rejection, outperforming the unmodified membrane's 99.32% rejection. Long-term filtration tests further demonstrated that the TNS-coated membrane maintained its water flux and ion-rejection characteristics under prolonged operation, indicating enhanced durability and fouling resistance. These improvements were attributed to the stabilizing effect of the TNS bilayer, which mitigates surface fouling, while preserving ion selectivity, positioning this tailored TFC-RO membrane as a promising candidate for practical radionuclide wastewater treatment [39]. Shaltout et al. designed a prototype spiral-wound thin-film composite RO system for the removal of ^{137}Cs from liquid radioactive waste streams. To enhance the retention of small cesium ions, the system incorporated two key modifications: (1) addition of mono- and dibasic acids (stearic acid, tartaric acid, citric acid, and EDTA) to the feed solution, thereby increasing the effective ionic radius of Cs^+ trough complex formation; and (2) chemical modification of the RO membrane structure, as confirmed by SEM, FTIR and EDX analyses. Under optimized conditions-specifically, the use of 1.5 $g \cdot L^{-1}$ EDTA in the feed-the retention coefficient for cesium (R_{Cs}) approached ~98%. Elution experiments with 0.25 M HCl achieved effective desorption of Cs^+ from the loaded membranes. Langmuir isotherm modeling indicated that chemical sorption on the membrane surface constitutes the dominant mechanism of Cs retention. The authors conclude that the modified RO-TLC system provides a promising pretreatment approach for ^{137}Cs removal [40].

Table 2 summarizes the main membrane types used for radionuclide separation, outlining their pore sizes, operating mechanisms, and primary advantages and limitations.

Table 2: Comparison of Pressure-Driven Membrane Processes Used for Radionuclide Separation

Membrane Type	Pore Size	Separation Mechanism	Advantages	Disadvantages
Microfiltration (MF)	0.1÷1 µm	Sieving (size exclusion)	• Highly effective for removing suspended solids and colloids • Low operating pressure and energy demand • Protects downstream membranes as a pretreatment step	• Cannot remove dissolved radionuclide ions • Limited selectivity • Often requires coagulation/flocculation
Ultrafiltration (UF)	0.001÷ 0.1 µm	Size exclusion + retention of colloids/complexed species	• Efficient removal of macromolecules, colloids, and metal-surfactant complexes • Suitable as pretreatment or polishing • Stable flux with moderate energy requirements	• Free radionuclide ions can not be removed unless complexed • Membrane fouling may occur • Often requires integration with adsorption/complexation
Nanofiltration (NF)	~1 nm	Steric hindrance + Donnan electrostatic exclusion	• High rejection of divalent /multivalent radionuclides (e.g., Sr^{2+}, Co^{2+}, UO_2^{2+}) • Partial desalination at lower energy than RO • Suitable for coolant purification and boric acid recovery	• Limited retention of monovalent ions (e.g., Cs^+) • Sensitive to feed pH and ionic strength • Susceptible to fouling and scaling
Reverse Osmosis (RO)	< 0.5 nm (dense polymer active layer)	Solution-diffusion mechanism	• Highest radinuclide removal efficiency (> 95–99.9%) • Removes ions, organics, and particulates • Well established for radioactive wastewater treatment	• High energy consumption • Requires high-pressure operation • Significant fouling potential; pretreatment needed • Generates radioactive brine requiring management

Overall, the comparison shows that each membrane type fulfills a distinct functional niche in radioactive wastewater treatment. MF and UF are most effective as pretreatment or polishing steps for removing colloids and particulate-bound radionuclides, whereas NF provides selective rejection of multivalent species through combined steric hindrance and Donnan exclusion. RO remains the most robust and comprehensive option for radionuclide removal due to its dense active layer and solution-diffusion mechanism, although these advantages come at the expense of higher operating pressures, energy consumption, and concentrate management requirements.

3. Fundamentals of Adsorption Processes in Radionuclide Treatment

Adsorption is widely recognized as one of the most effective technologies for the remediation of radionuclide-contaminated wastewater, owing to its high operational stability, low cost, simplicity of implementation, minimal secondary waste generation, and relatively low energy requirements. These attributes make adsorption particularly suitable for the treatment of low-concentration radioactive effluents, where other separation processes may become inefficient or economically impractical [41-43]. At its core, adsorption involves physicochemical interactions between radionuclide species and active sites on a solid sorbent surface. These interactions are governed by several fundamental mechanisms, including electrostatic attraction, ion exchange, surface complexation, and intra-particle (pore) diffusion. A clear understanding of these principles is essential for evaluating the performance of different classes of sorbents-ranging from natural materials to advanced engineered platforms such as MOFs, COFs, POPs, graphene-based materials, and titanate-derived sorbents-used in radionuclide removal applications.

3.1. Adsorbent Materials Used in Radionuclide Treatment

The adsorbent represents a critical component of adsorption-based treatment technologies, as its physicochemical properties directly determine the efficiency, selectivity, and overall feasibility of radionuclide removal processes. Various categories of adsorbents-including natural, composite, and engineered materials-along with their corresponding adsorption capacities, are summarized in Table 3. Adsorbents exhibiting high sorption capacities and strong selectivity toward specific radionuclide ions are particularly desirable, as they enable efficient decontamination even at low contaminant concentrations and enhance the overall performance of adsorption systems.

3.1.1. Natural adsorbents

Traditional adsorption processes often rely on activated carbon, which, although effective, is relatively expensive. Therefore, renewable natural materials such as zeolites and clays have been widely employed in radioactive wastewater treatment due to their abundance, renewability, and low cost. In recent years, bio-based adsorbents derived from natural alginate, chitosan, and other biopolymers have gained increasing attention, offering additional advantages such as environmental compatibility, tunable surface functionality, and improved adsorption performance. Clay minerals are also regarded as ideal adsorbents due to their non-toxic nature and large specific surface areas. Zeolite- an aluminosilicate with a porous framework- is extensively used for the adsorption of potentially toxic elements (PTEs) and radionuclides, exhibiting strong ion-exchange capacity and high surface area [44]. Muslim et al. examined the efficacy of bentonite clay as natural adsorbent for the removal of ^{137}Cs from real radioactive wastewater via batch adsorption experiments. The Iraqi bentonite was characterized by XRD, SEM, EDX, BET, and FT-IR before and after adsorption, and operational parameters-such as pH (6-8), solid-to-liquid ratio (1 $g \cdot L^{-1}$), contact time (1.5 h), and temperature (298 K) were optimized. Under these conditions, a removal efficiency of ~ 95.66% for ^{137}Cs was achieved. Equilibrium data were best described by the Freundlich isotherm model ($R^2 \approx 0.998$), while the kinetics conformed well to a pseudo-second-order model ($R^2 \approx 0.983$). Thermodynamic analysis indicated that the adsorption is an endothermic physisorption process ($\Delta H° = +15.01$ kJ/mol), spontaneous ($\Delta G°$ negative), and accompanied by a small change in entropy ($\Delta S° \approx +0.076$ kJ/mol·K). These findings highlight bentonite's potential as a cost-effective, eco-friendly sorbent for ^{137}Cs decontamination [45]. Abbas et al. developed a magnetic Fe_3O_4@zeolite NaA nanocomposite via hydrothermal synthesis, exploiting its high surface area and cation-exchange capacity to treat actual radioactive wastewater. Systematic adsorption experiments were performed to optimize key parameters-such as adsorbent dose (10 g $\cdot L^{-1}$), pH (7), and contact time (60 min). A very high adsorption capacity of 2744.44 $mg \cdot g^{-1}$ for ^{137}Cs was achieved under real-wastewater conditions. The adsorption mechanism was attributed to chemisorption, driven primarily by ion exchange between Cs^+ and Na^+, as well as physical entrapment within the zeolite pores. In addition, the nanocomposite's magnetic properties allowed rapid separation from solution, making the material a promising, cost-effective, and scalable adsorbent for managing liquid radioactive waste while minimizing secondary environmental risk [46]. Zhuang et al. synthesized a novel fibrous chitosan biosorbent via wet spinning using a LiOH/KOH/urea/water solvent system to remove Co^{2+} and Sr^{2+} from aqueous solutions. The fibrous morphology produced a high specific surface area (~ 16.9 m^2g^{-1}), uniform fiber fineness (~ 24.1 µm) and improved mechanical strength, all of which contributed to a significantly faster adsorption rate compared to spherical chitosan. The maximum adsorption capacities were found to be 31.3 mg/g for Co^{2+} and 20.0 mg/g for Sr^{2+} - substantially higher than those observed for conventional spherical chitosan (22.5 $mg \cdot g^{-1}$ and 8.9 $mg \cdot g^{-1}$, respectively) [47].

3.1.2. Composite adsorbents

Due to common drawbacks of natural adsorbent materials-including low adsorption capacity, insufficient selectivity, and limited regeneration potential- research efforts have increasingly focused on composite attention. These materials, produced through chemical or physical methods, are engineered to display tailored adsorption properties. By integrating specific pore architectures, surface functional groups, and controlled chemical compositions, composite adsorbents can be optimized to efficiently capture targeted contaminants in gaseous, liquid, or solid phases.

Li et al. developed a novel, highly selective composite adsorbent by functionalizing silica (SiO_2) with a dithiol containing ligand, DPTESAM (di(p-thioethyltriethoxysilane) amine), yielding the DPTESAM@SiO_2 material. FT-IR, XPS, and SEM-EDX analyses confirmed the successful grafting of thiol-amine functional moieties onto the silica matrix, providing strong coordinative affinity toward Fe^{2+}/Fe^{3+} species. Batch adsorption studies demonstrated rapid and highly selective uptake of ^{55}Fe from complex nuclear waste solutions, characterized by high distribution coefficients and fast kinetics (with equilibrium achieved within minutes). The adsorption behavior followed a pseudo-second-order kinetic model, indicating a chemisorption-dominated mechanism, whereas pH-dependent performance revealed optimal removal under acidic conditions. Selectivity assessments further showed negligible interference from coexisting ions common to nuclear waste streams (e.g., Na^+, Cs^+, Sr^{2+}), highlighting the high specificity of DPTESAM@SiO_2 for ^{55}Fe. Furthermore, the adsorbent was also readily regenerable under mild conditions, maintaining a substantial portion of its capacity over repeated adsorption-desorption cycles. These results highlight DPTESAM@SiO_2 as a promising candidate for the efficient and selective removal of ^{55}Fe in nuclear waste management applications [48]. Prussian Blue Analogues (PBAs) have likewise emerged as highly effective composite materials for the selective capture of Cs^+ ions, often surpassing conventional Prussian Blue due to their tunable metal-cyanide frameworks. In systems such as Co-Fe and Fe-Fe PBAs, ion-exchange mechanisms dominate the cesium uptake process. Previous studies have also demonstrated that compositional modification, for example increasing Zn content in Zn-Fe-Co PBAs, can significantly enhance Cs adsorption capacity. Moreover, structural vacancies resulting from missing $[M(CN)_6]$ linkers generate additional active sites that facilitate efficient Cs^+ binding. Many PBAs also demonstrate notable stability during adsorption, exhibiting minimal Fe or Co leaching under aqueous conditions. Li et al. synthesized a Fe-Co Prussian Blue Analogue (PBAFe) through a simple precipitation method and reported excellent performance for Cs^+ removal. PBAFe showed rapid adsorption kinetics with short equilibrium times, and achieved maximum Cs^+ adsorption capacities reached ≈ 150-170 $mg \cdot g^{-1}$, depending on experimental conditions. It also maintained high selectivity in the presence of competing ions such as Na^+, K^+, and Ca^{2+}. Structural stability assesments confirmed minimal metal leaching across a broad pH range, reinforcing its suitability for radionuclide remediation [49].

3.1.3. Engineered adsorbents

Engineered adsorbents refer to materials whose adsorption functionality is deliberately tailored through structural design, surface modification, or nanoscale Engineering- where performance is governed primarily by human-driven manipulation rather than the intrinsic properties of the raw components. Metal-organic frameworks (MOFs), covalent organic frameworks (COFs), MXenes, and graphene-based materials have demonstrated significant potential for radioactive wastewater treatment. MOFs and COFs are widely investigated due to their exceptionally high specific surface areas and tunable porous architectures, enabling the rational design of adsorption pathways and binding environments. MXenes, characterized by excellent electrical conductivity and intrinsic hydrophilicity, can be engineered through surface termination control to enhance their affinity for heavy metal ions and various radionuclides. Similarly, graphene-based materials exhibit remarkable mechanical and chemical stability, and their surface chemistry can be precisely modified to introduce functional groups capable of selectively binding contaminant species within complex wastewater matrices. Post-synthetic modifications - including targeted functionalization and heteroatom incorporation-further expand the adsorption capacity and specificity of these platforms by creating well-defined interaction sites. Current research on engineered adsorbents focuses heavily on developing novel synthesis and modification strategies to enhance adsorption efficiency, selectivity, and long-term operational stability, particularly under harsh radiochemical environments. Hassan et al. synthesized an eco-friendly chitosan/graphene oxide (CS/GO) composite hydrogel for the simultaneous removal of ^{134}Cs and $^{152+154}Eu$ radionuclides from aqueous solutions. The hydrogel was prepared through a green cross-linking method and characterized using FT-IR, XRD, and SEM to confirm successful integration of chitosan and oxygen-containing groups of graphene oxide. Batch adsorption experiments were performed to optimize key operational parameters, including pH, adsorbent dose, contact time, and initial radionuclide concentration. The composite displayed high adsorption efficiencies for both radionuclides, attributable to the synergistic effect of chitosan's amino groups and the large specific surface area and functionalization of graphene oxide. Kinetic modeling indicated good agreement with the pseudo-second-order model, suggesting chemisorption-driven uptake, while isotherm analyses confirmed favorable sorbate-sorbent interactions. The hydrogel also exhibited regeneration capability over successive adsorption-desorption cycles, demonstrating its practical potential as a sustainable and practical engineered biosorbent for radioactive wastewater treatment [50].

Table 3: Adsorbent Materials Used in Radionuclide Removal

Category	Adsorbent Material	Adsorbed Radionuclide	Adsorption Capacity/ Efficiency	pH	Key Characteristics	Ref.
Natural	Zeolite	^{137}Cs	77 mg·g^{-1} /95.66%	6.0	High thermal/ chemical stability	[51]
	Zeolite	^{137}Cs	67.8%	7.23	Cost-effective/ selectivity	[52]
	Zeolite	^{90}Sr	93.5%	7.9	Cost-effective/ selectivity	[52]
	Bentonite	Eu	4.52 mg·g^{-1}	7.0	High Eu(III) uptake	[53]
	Bentonite	U	9.124 mg·g^{-1}	5.0÷7.0	Spontaneous, multi-surface complexation	[54]
	Bentonite	^{137}Cs	98%	6.0	Eco-friendly/ low-cost	[55]
	Kaolinite	^{137}Cs	75%	6.0	Eco-friendly/ low-cost	[55]
	Attapulgite	^{137}Cs	97%	6.0	Eco-friendly/ low-cost	[55]
	Wheat straw SMS-L	^{232}Th	87%	4.0	High selectivity	[56]
	Tree bark (Amla)	U(IV)	121.95 mg·g^{-1}, 84.3%	7.0	High affinity	[57]
	Lignin-phytic acid (PA)	Ce(III), La(III)	743.16/735.60 mg·g^{-1}	3.8÷4.3	Highly efficient treating	[58]
Composite	Titanium Phosphate (TiP)	Sr^{2+}	373.6 mg·g^{-1}	5.0	Porous, chemically stable	[59]
	Ag@ACF	I$^-$	372 mg·g^{-1}	2.0	Excellent stability	[60]
	alg@PB	Cs$^+$	136 mg·g^{-1}	2.0÷10.0	High capacity	[61]
	CAA@MgAlFe	U(IV)	533 mg·g^{-1}	12.0	Fast adsorption; stable	[62]
MOF-based	PA/PEI@Zr-MOF	I$_2$, DMNP	609 mg·g^{-1}	—	Dual-functional; rapid adsorption	[63]
MOF-based	UiO-66-Lys/PAN	^{60}Co	41.4 mg·g^{-1}	—	High stability	[64]
COF-based	COF-1	Eu(III)	—	—	High porosity	[65]
MXene-based	Ti$_3$C$_2$Tx MXene	U(VI)	High	—	Rich active sites	[66]
MXene-based	Dialdehyde cellulose-MXenes	U(VI)/Th(IV)	105.7 mg·g^{-1} 95.1 mg·g^{-1}	—	High stability/ selectivity	[67]
GO-based	ssFG	U (C2F, C6F, C8F)	3.45 eV, 1.20 eV, 1.12 eV *	—	Excellent performance	[68]
GO-based	PAO-N-rGO/CF	U(VI)	277.8 mg·g^{-1}/ 99.93 %	7.0	Thermal stability; multifunctional	[69]
GO-based	3D-Printed Ca-Alginate /Graphene Oxide Adsorbent	U(VI)	740.74 mg·g^{-1} / 97.7 %	5.0	High selectivity	[70]
CNT-based	CNTs	Co^{2+}	117.87 mg·g^{-1}	—	Reusable; high selectivity	[44]

Note: *values associated with adsorption energy in [eV].

4. Hybrid Membrane Processes (HMPs)

In practical radioactive wastewater treatment, membrane technologies alone often face limitations, particularly when rapid and selective radionuclide removal is required. Hybrid membrane processes (HMPs)-formed by integrating membrane units with biological and/or physicochemical treatment methods-represent an advanced class of technologies that offer enhanced separation efficiency and improved operational robustness, and reduced treatment cost. In general, an HMP couples membrane filtration with complementary processes such as ion exchange, adsorption, catalytic ozonation, photocatalysis, coagulation and electrocoagulation, or humidification /dehumidification operations [71]. HMP configurations may also involve combinations of different membrane processes (e.g., MF/UF–NF, UF–RO) along with with conventional physical treatment techniques. In systems of the first type, a membrane step is integrated with a biological or chemical transformation process, while in the second category, membrane separation is combined with a physical operation. These hybrid configurations act as environmentally friendly “green” technologies that enhancing the capabilities of both conventional treatment methods and membrane-based molecular separation, thereby generating synergistic effects that minimize environmental and economic impacts. Pretreatment remains a critical element for ensuring membrane performance. Solid-liquid separation steps such as MF and UF are often combined with precipitation, adsorption or flocculation to reduce turbidity and particulate load. Although NF and RO can directly remove dissolved radionuclides, their applicability depends strongly on water quality parameters, making pretreatment evaluation essential. Man et al. proposed a hybrid process combining membrane electrolysis with nanofiltration to simultaneously recover tungsten and thorium originating from welding-rod (W-Th) industrial waste. Their system consisted of an electrolysis cell equipped with a sulfonated poly(ether ether ketone) (sPEEK) membrane and a nanofiltration unit employing a chitosan-polypropylene hollow-fiber membrane (C-PHF-M). Characterization of discarded W-Th electrodes indicated a composition of approximately 89.4% W, 7.1% Th, 2.5% O_2, and 1.1% Al. Optimal operating conditions for both membrane electrolysis and nanofiltration were achieved within a pH range of 6-9, under which soluble tungstate ions, solid aluminum hydroxide, and solid thorium dioxide were simultaneously generated. This hybrid configuration demonstrates the potential of combining electrochemical reactions with membrane-driven processes to efficiently treat mixed-metal radioactive industrial residues [72].

Large-scale treatment facilities have also been established at the Fukushima Daiichi Nuclear Power Plant to manage the continuous production of radioactive wastewater. Following the accident, seawater injection for emergency cooling, combined with infiltration of rainwater and groundwater, generated enormous volumes of radionuclide-contaminated water containing species such as ^{137}Cs. As these waters accumulated in storage tanks, the need for robust, high-throughput treatment systems intensified. Over time, several engineered systems-including the Kurion system, SARRY system, desalination units, and the Advanced Liquid Processing System (ALPS)-were developed and installed to remove radionuclides from the contaminated water. Early-stage systems such as Kurion and SARRY primarily relied on zeolite and silicotitanate adsorbents, respectively, to achieve efficient removal of radionuclide species [73].

4.1. Membrane-Adsorption Hybrid Systems

Membrane-adsorption hybrid systems integrate membrane filtration with adsorptive removal mechanisms (Figure 2), uniting the molecular-level size exclusion inherent to membrane processes with the high affinity and selectivity of engineered adsorbent materials.

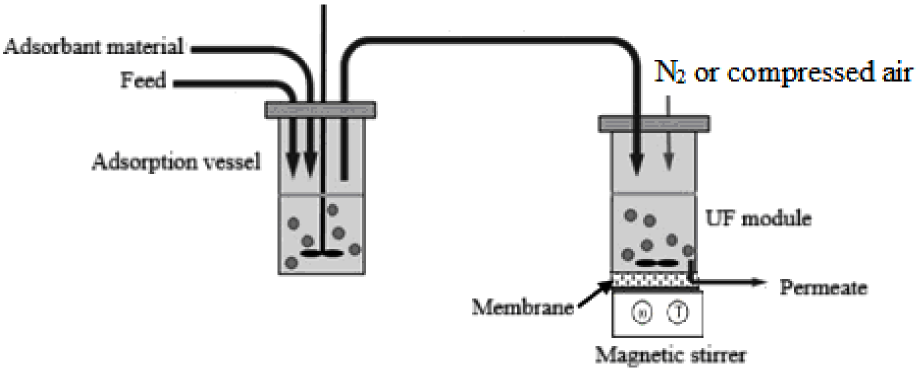

Fig. 2. Schematic representation of the hybrid adsorption-ultrafiltration process

Depending on the target radionuclides and wastewater characteristics, adsorption may be incorporated as a pretreatment, post-treatment, or simultaneous step within the membrane separation process. When applied as a pretreatment, adsorbents reduce membrane fouling, reduce the concentration of sparingly soluble radionuclides, and stabilize feedwater composition. In contrast, post-membrane adsorption is particularly advantageous for polishing permeate streams where residual radionuclides must be reduced to meet stringent regulatory discharge limits. A key advantage of membrane-adsorption hybrid configurations is their synergistic behavior. Adsorption decreases fouling potential and reduces ionic load, thereby improving membrane flux and extending operational lifespan. Conversely, membrane filtration removes suspended solids and competing ions, leading to enhanced selectivity and adsorption capacity. This mutual enhancement results in higher overall removal efficiencies, reduced chemical consumption, and minimized secondary waste generation.

A wide range of adsorbent materials -including activated carbon, zeolite, bentonite, ion-exchange resins, chitosan-based biosorbents, metal-organic frameworks (MOFs), and functionalized nanomaterials-has been successfully incorporated into membrane radionuclide treatment systems. The choice of adsorbent depends on factors such as the target ion’s charge density, hydration radius, coordination chemistry, and the competitive ionic environment. Recent studies have consistently demonstrated that coupling ultrafiltration or nanofiltration membranes with high-affinity adsorbents significantly enhances the removal of radionuclides such as cesium, strontium, europium, uranium, and other radionuclides from complex aqueous matrices. Overall, membrane-adsorption hybrid systems represent a versatile, efficient, and operationally stable approach for treating radioactive wastewater. Overall, membrane–adsorption hybrid systems provide a versatile, efficient, and approach. Their adaptability to variable feedwater chemistries and resilience under fluctuating loading conditions further underscore their suitability for large-scale applications.

Miśkiewicz and Kołtuniewicz investigated biosorbent-UF hybrid systems for the removal of radionuclides from low-level radioactive wastewater. Their study examined various natural biosorbents- including alginate-, chitosan-, and cellulose-based materials-as presorption media prior to UF, examining the effects of sorbent type, solution pH, and ionic composition on radionuclide uptake. The hybrid process significantly improved stand-alone UF, achieving >90-98% removal of key radionuclides such as Cs^+, Co^{2+}, Sr^{2+} and U(VI). Sorption capacities ranged from 20 to 80 mgg^{-1}, depending on biosorbent type and radionuclide, with optimal performance occurring at mildly acidic to neutral pH (pH 5-7), where functional groups (carboxyl, hydroxyl, and amino) exhibit maximal binding affinity. The authors demonstrated that coupling biosorption with UF reduces membrane fouling, enhances selectivity, and improves the separation of dilute radionuclide solutions, confirming the suitability of biosorbent-assisted UF hybrids for low-level radioactive wastewater treatment [74]. Fuks et al. investigated coal fly ash (CFA) as a low-cost adsorbent within an adsorption-membrane filtration (AMF) hybrid process using a PES ultrafiltration membrane (0.22 μm). Their results showed that CFA efficiently bind radionuclides in both batch and membrane-assisted modes and offering it as an economical alternative to traditional column-based adsorption for specific wastewater matrices [75]. In another study, Muslim et al. developed an adsorption-UF hybrid system for removing ^{137}Cs from real radioactive wastewater using natural clay as a low-cost sorbent. Integrating clay adsorption with UF significantly enhanced cesium removal efficiency relative to standalone processes. The clay exhibited strong affinity for Cs^+, and its integration into the hybrid system decreased membrane fouling and improved permeate quality. Under optimized conditions, the process achieved high removal efficiencies and produced effluent compliant with regulatory discharge standards. The authors concluded that the clay-assisted adsorption/UF configuration represents a robust and economically feasible option for treating cesium-containing radioactive wastewater [76].

Zhang et al. developed a MoS_2/PEI-based nanofiltration membrane via a layer-by-layer self-assembly technique, achieving exceptional removal performance for multiple target ions: 99.83% for Cs(I), 100% for Sr(II), 99.2% for Hg(II), 98.13% for Pb(II), and 98.5% for As(III). The membrane’s structure combines positively charged PEI, which provides abundant amine groups for electrostatic interaction and chelation, with MoS_2 nanosheets which provide low-resistance water channels. This structural design enhances both selectivity and flux. Long-term filtration tests and regeneration experiments confirmed excellent stability and reusability for trace level of radionuclide removal, demonstrating the membrane's strong potential for practical application in contaminated water treatment [77].

4.2. Membrane-Membrane Hybrid Systems

Several water treatment facilities in the United States and Canada have attempted to apply membrane-based processes for the treatment of real radioactive wastewater. Ultrafiltration and reverse osmosis systems were implemented to treat contaminated aqueous waste generated during the chemical cleaning of steam generators and heat exchangers at the Bruce Nuclear Power Plant in Canada [78]. The ultrafiltration unit consists of four parallel banks, each containing ten Zenon ZPF12 tubular membranes connected in series. The first-stage reverse osmosis system comprises two parallel banks arranged in series and operates at 6.5 MPa. The second-stage reverse osmosis system functions at 4.1 MPa and includes eight vessels configured as three parallel pairs in series, followed by two additional vessels in series. Each vessel is loaded with six Filmtec SW30HR spiral-wound elements.

At the Savannah River Site (SRS), a major U.S. Department of Energy complex responsible for processing nuclear materials from the national defense program, the Effluent Treatment Facility (ETF) incorporates three parallel treatment trains, each consisting of a three-stage system. Microfiltration is achieved using cross-flow ceramic membranes with a nominal pore size of 0.2 µm, while the reverse osmosis units employ high–salt-rejection spiral-wound elements. The first RO stage includes two pressure vessels containing six membrane elements each, whereas the second and third stages each utilize one vessel. Each train has a treatment capacity of 385 L/min, and the system design provides both equipment redundancy and flexibility in production flow. Since its commissioning in October 1988, the facility has consistently met discharge standards. The most significant operational challenge encountered has been bacterial biofouling of the microfiltration membranes [79]. Yang et al. investigated an integrated Reverse Osmosis-Membrane Distillation-Adsorption (RO-MD-Adsorption) process for the advanced treatment of high-salinity and low-level radioactive wastewater. Their study demonstrated that coupling these three processes significantly enhanced radionuclide removal while simultaneously reducing salinity to levels suitable for safe discharge or reuse. The results showed that the RO unit effectively removed the majority of dissolved radionuclides but was limited by osmotic pressure and membrane fouling under high salinity conditions. Integration with membrane distillation (MD) facilitated further concentration of the brine and improved water recovery, thereby overcoming the salinity limitations of RO. Finally, the addition of a polishing adsorption step ensured near-complete removal of residual radionuclides (e.g., Cs, Co, Sr), achieving overall removal efficiencies exceeding 99%. The authors concluded that the combined RO-MD-adsorption process provides a highly efficient, stable, and scalable treatment strategy for complex radioactive waste streams, providing improved operational reliability, reduced secondary waste generation, and enhanced suitability for nuclear facility wastewater management [80]. In addition, forward osmosis (FO) systems-such as those demonstrated by Şahin for treating complex wastewater streams-highlight the potential of low-energy osmotic-driven membrane processes to complement RO-and MD-based treatment trains, particularly in managing high-salinity or fouling-prone radioactive effluents [81, 82].

4.3. Performance Metrics and Efficiency in Radionuclide Treatment

The performance of membrane-, adsorption-, and hybrid-based radionuclide treatment systems is typically evaluated using key indicators such as removal efficiency, adsorption capacity, and ion selectivity. Removal efficiencies for common radionuclides (e.g., Co^{2+}, Sr^{2+}, Cs^{+}) often exceed 90% when optimized membrane materials or high-affinity adsorbents are employed, while adsorption capacities vary widely depending on surface area, functional groups, and overall solution chemistry. Selectivity coefficients are critical for distinguishing the affinity of membranes or adsorbents toward targeted radionuclides in the presence of competing ions such as Ca^{2+}, Mg^{2+}, Na^{+}, and K^{+}; these background ions can significantly influence removal performance by altering electrostatic interactions, complex formation, or adsorption site availability. Solution pH, ionic strength, and the presence of organic or colloidal matter further affect radionuclide retention by modifying membrane surface charge, hydration radius, and the degree of ion hydrolysis.

Guo et al. successfully produced a novel nitrogen-doped biochar (AL-N/BC-700) and investigated its adsorption mechanisms for U(VI). The material exhibited an exceptional U(VI) adsorption capacity of 25,000 mg/g - markedly higher than many conventional adsorbents-and achieved a desorption efficiency of 94.5% in the presence of Na_2CO_3. These results address critical challenges in radionuclide removal, including adsorption instability, high operational costs, low desorption efficiency, and interference from coexisting ions [83]. Sumalatha et al. synthesized potassium hydroxide–activated biochar from peanut shells (PSABC) via pyrolysis at 400 °C for 30 minutes. PSABC exhibited remarkable performance, achieving a maximum radionuclide removal rate of 99.02%. Even in the presence of high concentrations of competing Ca^{2+} ions, it maintained a removal efficiency of 53.55%, offering innovative pathways for radionuclide adsorption [84].
Repeated adsorbent regeneration and hybrid system use can damage structure, reduce capacity, and degrade functional groups, while membranes suffer fouling and radiolysis; evaluating materials under realistic conditions is crucial for their long-term radionuclide removal performance.

5. Challenges and Future Directions

Despite progress in membrane, adsorption, and hybrid technologies for radionuclide treatment, key scientific, technical, and economic barriers still hinder large-scale deployment [85]. Overcoming these limitations is essential for translating laboratory advances into reliable field systems.

- Scalability and economic feasibility: Advanced membranes, engineered adsorbents, and hybrid systems remain costly to produce and operate. Industrial-scale use requires more efficient membrane synthesis, lower-cost adsorbent fabrication, and modular, energy-efficient system designs.
- Long-term stability and fouling: Membrane fouling from organic matter, colloids, biofilms, and radioactive precipitates continues to reduce performance and raise operating costs. Chemical and radiolytic degradation further threaten long-term stability. Progress depends on antifouling surface strategies, radiation-resistant polymers, and regenerable or replaceable functional layers.
- Integration with other treatment methods: Membranes alone may not handle complex radionuclide mixtures. Coupling with ion exchange, oxidation, electrochemical processes, MD, or FO can provide synergistic removal. Membrane-adsorption hybrids are particularly promising for improving selectivity, extending membrane life, and lowering overall costs.
- Emerging materials and smart membranes: Advances in nanomaterials [86-88], MOFs, COFs, polymer–inorganic hybrids, and stimuli-responsive membranes offer new routes for selective radionuclide capture. Smart membranes with dynamic charge control, pH-responsive binding, or tunable pores may enhance removal of challenging species such as U(VI), Th(IV), Ra^{2+}, and Am^{3+}.

6. Conclusions

Membrane-based processes, adsorption technologies, and hybrid systems show strong potential for addressing radionuclide contamination in water. Pressure-driven membranes (MF, UF, NF, RO) enable effective physical and electrostatic separation, while adsorption offers high selectivity across diverse radionuclides. Hybrid membrane processes-especially membrane–adsorption and membrane–membrane configurations-provide synergistic gains in efficiency, stability, and operational flexibility. Overall, advances in membrane design, functionalization, and process optimization continue to enhance treatment performance, though challenges such as fouling, durability, cost, and scalability remain. Continued development of functional materials, radiation-resistant polymers, and smart membranes will be essential for next-generation remediation technologies.

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