



Decontamination of Aqueous Nuclear Waste via Pressure-driven Membrane Application – A Short Review

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HIGHLIGHTS

- Review of advances in liquid radioactive waste processing using membrane technology
- Membrane processes show enormous potential in radioactive science and technology
- Focus on nanoparticle materials for removing hazardous isotopes from liquid wastes

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ABSTRACT

Radioactive waste poses significant risks to human health and the environment due to the emission of gamma rays, beta particles, and alpha particles from natural or artificial radioactive elements. This paper reviews the essential sources of radioactive waste, their potential risks to humans and the environment, their classification depending on their chemical, physical, and radiological features, and the treatment techniques utilized to prevent them from leaking into the environment. In addition, the current study focuses on using membranes exclusively and membrane technologies in the remediation of radioactive liquid wastes. The use of pressure-driven membrane technologies, such as reverse osmosis, nanofiltration, ultrafiltration, and microfiltration, is particularly emphasized. Additionally, the use of nanomaterials like titanium dioxide, zinc oxide, silica, alumina, silver oxide, zeolite, copper ferrocyanides, and carbon nanotubes embedded with membranes to improve their effectiveness and enhance their applicability decreases the risks associated with radioactive contamination via modifying membrane properties including flow rate, the result of rejecting radioactive particles dissolved or suspended in contaminated water, hydrophobic and hydrophilic membrane's ability and among other properties. Moreover, incorporating nanoparticles into reinforced membranes enhances the mechanics of nanocomposite membrane surfaces via processes like adsorption and ion exchange with various radioactive ions. Besides increasing nanocomposite membranes' performance in reducing the hazardous radionuclides effects, it modifies membrane properties, such as enhancing antibacterial capabilities, antifouling, mechanical stability, and thermal stability. The article review also discusses hybrid pressure-driven membrane processes in disposing of radionuclides generated in nuclear power plants, hospitals, research facilities, and decontamination projects to protect human health and the environment.

1. Introduction

Nuclear technologies are widely used in diverse industrial and scientific research, as well as and in agricultural, medical, and environmental applications. Radioactive waste will surely be formed as a result of due to the production and use of radioactive materials; these wastes must be handled with considerable attention due to the inherent chemical, radiological, physical, and biological hazards. As a result, radioactive wastes could not be directly dumped into the environment since they endangered human health and the ecosystem. The primary goal of radioactive waste management is to reduce the volume of radioactive

materials to the smallest possible volume with the least amount of radioactivity to store or dispose of, particularly for long-term wastes of radioactive activity [1]. Radioactive waste is generated at all stages of nuclear research reactor operations, as well as operations involving the manufacture and use of radioisotopes. Furthermore, many sources of radioactive waste are created by nuclear facilities, such as the mining and milling of thorium and uranium nuclear fuel cycle process, nuclear power plants, dismantling and decontaminating nuclear research reactors, and power plants. Also, oil and gas applications during the drilling and exploration process for phosphate ore include radioactive waste production, to name a few. These activities are associated with the associated water exiting from phosphate reservoirs and oil or gas wells in a sludge phase (liquid or solid) known as naturally occurring radioactive (NORM) [1,2].

Natural occurring radioactive materials (NORM) are one of the sources of radioactive materials in various mediums, with chains of uranium and thorium found in soil, radium-226 and radium-228 present in associated water in oil reservoirs, and radon-222 present in primary storage tanks after oil or natural gas extraction operations [3]. As a result of the existence of various sources that generate this waste, it was necessary to consider radioactive waste classification. According to the International Atomic Energy Agency classification, radioactive waste can be classified according to sources (natural, artificial), state (solid, liquid, gas), physical properties (radiation dose, radiation level, half-life), chemical properties (toxicity, ability to corrode, solubility, radii ions, heat generation), and other properties. The classification of radioactive waste aids in safely managing it at nuclear plants and research centers regarding storage and treatment techniques, understanding the limits of waste that meet the release limit, and handling it per the safe management of it at nuclear plants and research centers in terms of storage and treatment techniques, understanding the limits of waste that meet the release limit, and handling it in accordance with the As Low As Reasonably Achievable (ALARA) principle. Additionally, alternative classifications that can aid in the management and treatment of liquid radioactive wastes (LRWs) include low-intermediate liquid waste (L-ILW) and high-liquid waste (HLW) [3-5]. The most difficulty in managing and treating radioactive wastes is the production of liquid radioactive wastes (LRW) at nuclear research centers, hospitals, universities, and research institutes' laboratories. The reason for this is because of the vast volumes produced, the complicated chemical compositions, and the changing levels of radioactivity. According to regulatory bodies' criteria, there are several procedures for treating and conditioning effluents for radioactivity before ultimate disposal and waste discharge. [6]. Liquid radioactive waste treatment focuses on minimizing the volume of liquid waste by eliminating dissolved radioactive isotopes in the liquid phase and concentrating it as solid waste. The solid waste is then solidified using cement, bitumen, and asphalt before being carefully kept to avoid its spread into the environment [7]. The processed liquid radioactive waste is subject to the release requirements for waste that fulfills the International Atomic Energy Agency (IAEA) and national criteria, as well as the safety release criteria of environmental protection organizations [8]. There are many treatment methods for these LRWs, including traditional methods and new methods, depending on the classification of liquid radioactive waste and its characteristics, as well as determining the treatment procedures used through operating costs, the amount of waste and its characteristics, and the purpose of the treatment procedures, including the size of the decontamination factors (DF), and the reduction of volume coefficients. Chemical precipitation, evaporation, distillation, solvent extraction, and ion exchange are prevalent ways for treating effluents, other than electrochemistry, biological approaches, and membrane separation are emerging technologies at nuclear power plants (NPP) and nuclear research institutions [9,10].

Although traditional methods have a relatively high decontamination factor (DF), they have some drawbacks in LRW treatment. For example, the process of ion exchange and sedimentation associated with sedimentation and evaporation requires a large amount of energy, in addition to the formation of high radioactivity secondary waste from the treated primary waste [11]. Modern technology has been used to treat LRW in recent decades. Membrane technologies are the most commonly used in effluent treatment because they have many advantages over traditional separation technologies, such as low energy requirements compared to other separation processes, ease of expansion by adding additional units, and the ability to work at relatively low temperatures and pressure conditions, as well as the ability to create an integrated hybrid system with other membrane separation processes and conventional separation processes [12,13]. On the other hand, there are drawbacks to this technology, including the need to select a membrane that will be in close contact with the radioactive solution for an extended specific time which may alter their separation and permeation characteristics. Therefore, several types of research on the impacts of irradiation on membrane stability have been undertaken to determine the membrane operation threshold in a particular application. They demonstrated that the threshold magnitudes of radiation doses that produce a structural difference in the membranes are great significant enough for most polymers. Thus it is preferable to use polymeric membranes utilized in low and medium-level radioactivity [14-17]. In another case, ceramic membranes are preferable since their resistance to chemicals and the impact of radiation. Zakrzewska-Trznadel and Harasmowicz investigated the permeate of the membrane with complexation for higher radioactive activity processing. MEMBRALOX and CÉRAME INSIDE ceramic membranes have been utilized [18]. Due to the importance of membrane technology in the treatment of LRW at many nuclear plants, the membranes utilized had to be developed, despite the low selectivity of the available membranes. As a result, numerous investigators sought to improve membrane performance (permeability, retention, separation factors, and permeate flux), membrane properties (membrane thickness, pore size, pore size distribution, charge density, hydrophobicity, and hydrophilicity, chemical and physical specifications) by combining different materials and adding additives or functional groups into the membrane matrix, and modifying membrane surface through the surface coating, blending, and grafting or plasmas treatment technique [19-21]. Membrane surfaces contain functional groups like carboxylic acids and primary amines, which can be added through chemical grafting. Surface grafting methods offer flexibility in customizing surface characteristics and precision in grafting at specific spots. This simple, controlled approach increases membrane chemical stability and performance [22]. Plasma modification is a flexible surface treatment that is commonly used to introduce chemically reactive functional groups to polymer surfaces to increase hydrophilicity and produce low-fouling membrane surfaces. Zhao et al.[23] showed that low-temperature plasma-

induced grafting increased the presence and antifouling properties of PVDF and PES/PSF membranes. Blending modification is a feasible method for large-scale production of polymer membranes, influencing morphological features like casting solution, manufacturing parameters, and additives. Unlike conventional grafting procedures, this approach creates hydrophilic surfaces and hollow fiber membranes without pre- or post-treatment. This approach creates hydrophilic surfaces and hollow fiber membranes without pre- or post-treatment, unlike conventional grafting procedures [24].

The present review study focuses solely on pressure-driven processes, with the ultrafiltration process being singled out due to its uniqueness in LRW treatment as documented in research and effective procedures for treating industrial radioactive waste (IAEA, TRS No. 370) [5] and practical applications. Many nuclear facilities and worldwide research institutes use the ultrafiltration process to remove radioactive isotopes, colloidal particulates, and suspended particles, and recover boric acid. In addition to advances in ultrafiltration membranes for eliminating dissolved monovalent radioisotopes (cesium-137, potassium-40) and divalent radioisotopes (strontium-90 and cobalt-60), which were previously restricted to reverse osmosis and nanofiltration techniques [25,26]. One of the most important advantages of the ultrafiltration process is the removal and processing of LRW, for example, ease of operating conditions is one of the excellent advantages of such a process. Operating pressure is relatively low when compared to reverse osmosis. A significant decontamination agent (DA) can be obtained in the removal of actinide in the colloidal or semi-colloidal form and some fission products. Ultrafiltration can eliminate alpha activity below the detection limit from a waste stream containing about 2.0 Bq/mL. Ultrafiltration can also retain dissolved radioactive ions by improving selectivity using the "seed ultrafiltration" technology (SUF). The process can be used to remove water and/or wash sludge from the sedimentation process and can gain a solid content of 30-40% by weight, greater than sedimentation or centrifugation. Ceramic ultrafiltration membranes can be widely used in the nuclear field to clean the core coolant of pressurized water reactors (PWRs) directly, removing the activity of the colloidal form with pH, temperature, and radiation resistance [26,27].

2. Radioactive Wastes

Many actions involving the usage of radionuclides and creating nuclear energy, including all stages of the nuclear fuel cycle, resulting in the generation of radioactive waste. Other activities that generate radioactive waste include the medical or industrial use of radioisotopes and sealed radiation sources, defense and weapons programs, and the (mostly large-scale) processing of mineral ores or other materials containing naturally occurring radionuclides (NORM), which must be managed as radioactive waste in some cases, phosphate ore processing and oil or gas prospecting are two examples. Also, other resources such as nuclear accidents or to remediate regions impacted by previous practices generate radioactive waste [5]. The radioactive waste produced is as diverse in terms of shape, activity concentration, and contamination as it is in terms of producing activity. It can be either solid, liquid, or gaseous. Concentrations of activity range from extremely high levels connected with spent fuel and remainders from fuel reprocessing to extremely low levels related to radioisotope use in research laboratories, hospitals, etc. The spectrum of half-lives of radionuclides included in radioactive waste is also vast [28]. Nuclear power generation creates a variety of radioactive waste, including spent fuel (if declared waste), other high-level waste (HLW) generated primarily from chemical reprocessing of spent fuel, and very low-level waste (VLLW), low-level waste (LLW), and intermediate level waste (ILW) generated as a result of reactor operations, reprocessing, decontamination, decommissioning, and other activities in the nuclear fuel cycle [29]. It is vital to define the features and type of radioactive waste and classify it according to its level of radioactivity. The rationale is that understanding its features saves the effort required to treat, store, transport, and manage it properly. Table 1 summarizes the physical, chemical, radiological, biological, and other features that influence identifying suitable waste treatment procedures and restricting its spread due to threats to human health and the environment [9,30].

Based on the information presented in the classification of radioactive waste through the radioactive level and the characteristics summarized in Table 1, it is possible to employ traditional and modern methods or to integrate them in the treatment of radioactive waste in a way that ensures non-proliferation limit its impact and deals with it in the long term without harming the environment.

3. Membrane Filtration Process

Membrane filtration is a physical separation mechanism that separates molecules into permeate and concentrates components, allowing certain fluid substances to flow while inhibiting others. This process is used in various applications. Membrane separation processes are influenced by membrane design, trans-membrane transport mechanisms, driving forces, and other features. Some processes have been tested in commercial applications, while others are empirical or under development. Existing separation procedures are used in various industries, including water remediation. [32], pharmaceutical [33], food processing [34], petroleum [35,36], saltwater desalination [37], and liquid wastes of radioactive activity [38]. Membranes are classified by their fabrication material, shape, and average pore size. According to the substance can be classified into two polymeric (organic) and inorganic membranes. The most common polymers used in the production of polymeric membranes are polyether sulfone (PES), polysulfone (PSf), polyamide-imide (PAI), polyvinylidene fluoride (PVDF), and polyacrylonitrile (PAN). This might be ascribed to their multiple benefits, which include high flexibility, strong film-forming capabilities, high perm selectivity, chemical stability, good mechanical qualities, low cost, and widespread commercial availability. Most polymeric membranes are stable up to 105 Gy radiation exposures [39,40]. On the other hand, metals, ceramics, and zeolite membranes are the most frequent inorganic membranes. They are resistant to harsh chemical characteristics, can endure thermal properties, and are significantly more resistant to radiation impact. Nevertheless, inorganic membranes' major drawback is their high production costs [38,41]. Membranes can be divided into two types based on their cross-section morphology: asymmetric

and symmetric [39]. Membranes are categorized into four main categories used in pressure-driven processes based on pore size, as shown in Figure 1, including Microfiltration (MF), Ultrafiltration (UF), Nanofiltration (NF), and Reverse Osmosis (RO). Porous membranes are used in microfiltration (MF) and ultrafiltration (UF). Compared to UF and MF, nanofiltration (NF) has a dense membrane structure with a very small pore size. RO is a high-pressure-driven filtering method that employs a denser membrane than NF [40].

Table 1: Important characteristics of radioactive waste that may be used as parameters for classification [31]

Properties	Influencing factors
Radiological	Half-lives of radionuclides Heat generation The intensity of penetrating radiation Activity concentration of radionuclides Surface contamination Dose factors of relevant radionuclides Decay products
Physical	Physical state (solid, liquid, or gaseous) Size and weight Compatibility Dispersibility Volatility Miscibility Free liquid content
Chemical	Solubility and chelating agents Potential chemical hazard Corrosion resistance/corrosiveness Organic content Combustibility and flammability Chemical reactivity and swelling potential Gas generation Sorption of radionuclides
Biological	Potential biological hazards Bio-accumulation
Other	Volume Amount arising per unit of time Physical distribution

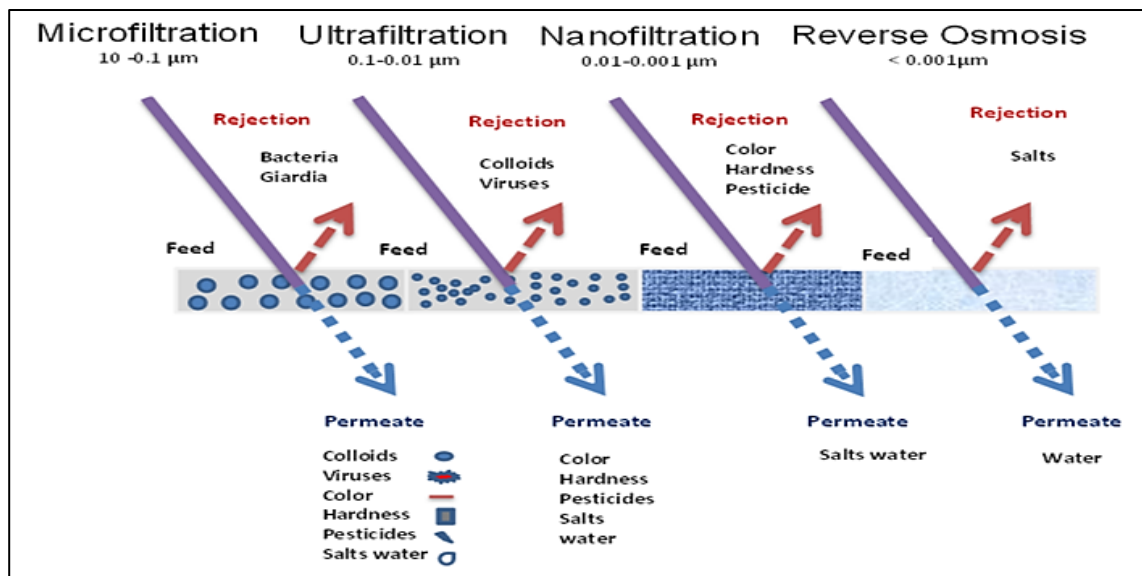


Figure 1: Classification of pressures-driven membranes and schematic illustrations of their operation according to the pore size

In nuclear power plants, membrane technologies have been used to treat radioactive washing waste, combined laboratory waste (organic liquid wastes), recycle boric acid solutions and reprocess liquid waste. Low and medium levels of radioactive waste are handled via membrane-based systems. Figure 2 depicts using two RO units and one UF unit to treat low-level effluents from radioactive liquids, including isotopes like cesium-137, strontium-90, and cobalt-60 [42].

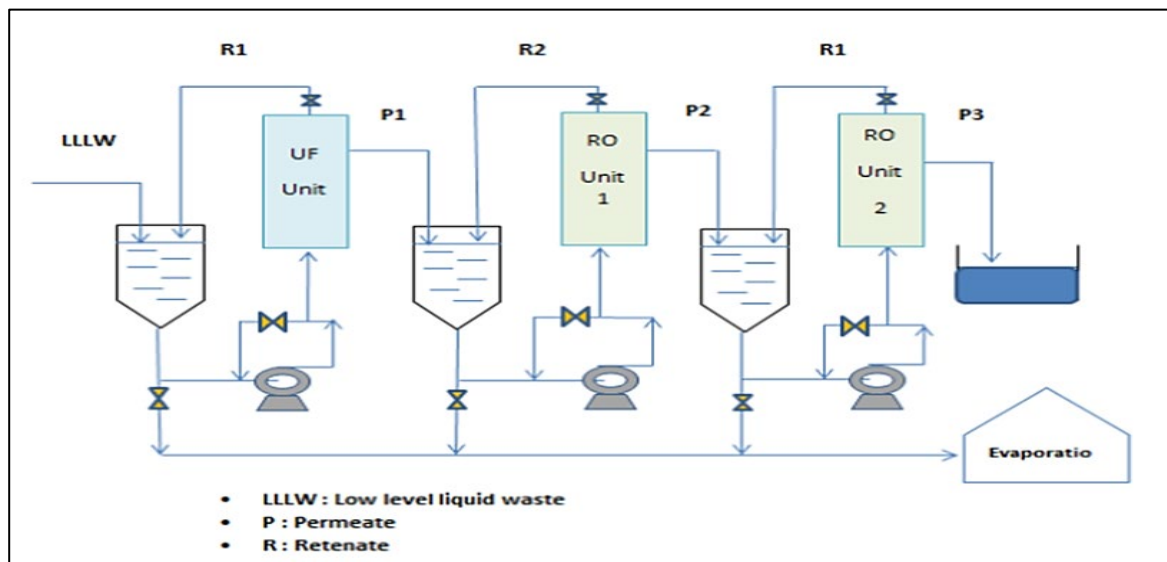


Figure 2: Schematic diagram of Ultrafiltration (UF) and two units of (RO) Plants for low-level liquid remediation

Because of the benefits and advantages of membrane technology, considerable attempts have been made by nuclear power plant researchers and employees to develop pressure-driven membrane units for the treatment of liquid radioactive wastes (LRW). The nuclear power facilities that have used membrane processes are listed in Table 2.

Table 2: Remediation of liquid wastes of radioactive activities via membrane process (LRW)

Membrane process	Nuclear centers	Modules/ type membrane	Solution treated	Ref.
(RO)	Chalk River Laboratories in Canada (AECL)	Tubular and spiral wound /cellulose acetate	Reactor coolant clean-up with the boric acid recovery	[44]
(NF)	Australian Nuclear Sciences and Technology Organization (ANSTO)	Commercial nanofiltration membranes	Remediation of uranium mill effluents	[45]
(UF)	Mound Lab in (UAS)	Tubular UF membrane	Process alpha emitters (^{241}Am , ^{238}Pu , ^{237}Np and ^{233}U) polluted waste from fuel reprocessing	[46]
(MF)	Rocky Flats in (USA)	Tubular MF membrane (0.1 μm)	Groundwater remediation and wastewater polluted with isotopes of heavy metals, uranium, and toxic organic particles	[47]

The nuclear industry prefers RO, NF, UF, and MF as pressures-driven separation processes over alternative membrane technologies for the following reasons [48]:

- They have a long track record of success in industrial water and wastewater remediation applications.
- They have established technologies with over 30 years of design and operation.
- There have already been many successful applications in the nuclear sector.
- The process design is well known, and computer simulation programs are accessible.
- Process configuration flexibility could improve performance.
- They could be integrated into a system with traditional remediation processes.
- For pressures-driven processes, a wide range of membrane materials and kinds are available, and these membranes may be tailored to the properties of the pollutants to be dumped.

They are appropriate for meeting broad process targets and requirements. Pressures-driven membrane devices eliminate anything from big particles to ionic substances.

Pressure-driven membrane separation techniques might be used as the primary technology for radioactive liquid waste cleanup or as an adjunct to the process. Most modern radioactive waste remediation strategies include membrane separation and conventional techniques. Membrane separation may be regarded as the principal remediation unit in some systems. However, in other circumstances, it might be used in conjunction with recognized techniques to improve performance. Furthermore, certain integrated systems may provide an economic cleanup strategy with acceptable doses of radioactive for eventual disposal. Table 3 outlines some of these methods, stressing their potential benefits and downsides. Membrane-based approaches have tremendous promise in the treatment of radioactive waste.

Table 3: Summary of various radioactive waste processes [26]

Processes	Advantages	Disadvantages
Precipitations	Suitable for large volumes and high salt content waste. Easy and non-expensive.	Low decontamination factor (DF)* Efficiency depends on solid–liquid separation step.
Ion exchange	Good chemical, thermal and radiation stability Wide choice of products ensuring high selectivity	Affected by high salt content Blockage problems Regeneration and recycling often difficult
Evaporation	DF>104 to 106 Well established technology High volume reduction, suitable for a variety of radionuclides	Process limitations (scaling, foaming, corrosion, volatility of certain radionuclides) High operation and capital costs
Solvent extraction	Selectivity enables removal, recovery or recycling of radionuclides/actinides	Generates aqueous and organic secondary waste
Liquid membrane	Simultaneous extraction and stripping, selectivity due to target-selective carriers Low operating cost	Membrane life, effect of radiation on membrane in long run
Reverse Osmosis (RO)	Removes dissolved salts DF 102–103, economical Established for large scale Operations	The high-pressure system, limited by osmotic pressure Non-back washable, subject to fouling
Nanofiltration (NF)	Negatively charged surface Separate single-charged ions from multi-charged ions retained Economical, provide high fluxes at low pressures Established for large scale Operations	Organic membranes subject to radiation damage Fouling
Ultrafiltration (UF)	Separation of dissolved salts from particulate and colloidal materials Good chemical and radiation stability for inorganic membranes	Organic membranes subject to radiation damage Fouling
Microfiltration (MF)	High recovery (99%) Low fouling when air backwash	Sensitive to impurities in waste stream

$$*DF = \frac{\text{Concentration of radioactivity waste before treatment}}{\text{Concentration of radioactivity after treatment}}$$

3.1 Reverse Osmosis

RO is a pressure-driven process that employs a semipermeable membrane. The membrane holds dissolved components in the feeding water, allowing clean water to flow [49]. In the reverse osmosis process, the pressure exerted to push the water flow into the less concentrated solution must be greater than the osmotic pressure of the feed solution. By applying a pressure larger than the osmotic pressure, clear water flows from the high solute quantity section over a membrane to the low solute amount part, resulting in water separation from the solution. The reversal of the regular osmosis process is referred to as "RO" [49]. In reverse osmosis, a pressure larger than the osmotic pressure (2-10 MPa) is given to a concentrated solution, causing the solvent to flow from the concentrated side of a semipermeable membrane to the diluted side. RO membranes are classified as asymmetric films and composite thin films. The asymmetric RO membrane comprises a single polymer with a thin selective skin-supported layer supported by a porous sub-layer. The dense skin layer has a thickness ranging from (0.1 to 1m). This layer influences flow and selectivity. However, the porous layer has little impact on the membrane's separation properties. Asymmetric membranes are frequently created using a phase inversion technique based on the Loeb-Sourirajan method. Since the invention of asymmetrical cellulose membranes in 1962, significant progress has been made in cellulose membranes (CA, cellulose triacetate, etc.) and liner aromatic polyamide to be used widely in specimen asymmetric membranes over the last twenty years. The polymer surface can be defined as a non-porous layer in reverse osmosis membranes. The reverse osmosis membrane comprises a "dynamic" polymer network that allows pure water molecules to flow through. Solution and diffusion govern the transport process, which holds nearly all ionic species in place [50-52]. RO is gaining international acceptance in desalination and water treatment applications. The use of reverse osmosis in the cleanup of radioactive waste liquids is considered a developed technique. This method has been used to remove radionuclides from low-level activities, such as waste streams in nuclear power plants. Garrett uses RO to remove all contaminants from solutions except dissolved gases and tritium [53]. RO testing findings for decontaminating wastewater and underground water were discussed. Data revealed that RO might retain strontium-90,

uranium-238, technetium-99, and cesium-137. Except for tritium-3, these compounds may be decreased to amounts lower than required. Reverse osmosis has been utilized as a single unit to extract boric acid from power station cooling systems. In addition, RO membranes based on ion exchange technology are employed as part of an integrated purification system for treating radioactive waste liquids [54]. In many nuclear plants, the RO system may be used with other membrane methods such as UF, MF, and NF to treat radioactive waste liquids. Table 4 illustrates some of these cases.

Table 4: Examples of utilizing reverse osmosis membranes in nuclear facilities to treat liquid wastes of radioactive activities (LRW)

Membrane process	Facility	Wastes processed	Ref.
Reverse osmosis with conventional pretreatment	Nine Mile Point nuclear power Plants (NMP) (USA)	Boiling water reactor (BWR) floor draining with different other wastes	[55,56]
	Pilgrim Nuclear power Plants (USA)	BWR floor draining and various other wastes	[57]
Reverse osmosis with Ultrafiltration pretreatment	Wolf Creek nuclear power Plants (USA)	Pressurized water reactor (PWR) floor draining, reactor outage waste, spent resin sluice water, etc.	[58,59]
	Comanche Peak nuclear power Plants (USA)	Floor draining, resin sluice water, boron recycle water	[60,61]
	Dresden nuclear power Plants (USA)	Inventory of TRU (trans-uranium) polluted batch of liquid waste	[61]
	Bruce nuclear power Plants (Canada)	Aqueous wastes from steam generator chemical cleaning	[62]
Reverse osmosis with Microfiltration pretreatment	AECL (Atomic Energy of Canada Ltd) Chalk River Lab (Canada)	Nuclear research wastes	[62-64]
	Savannah River site (SRS) (USA)	Reprocessing/defense wastes	[65,66]
Reverse Osmosis (two steps) with Ultrafiltration pretreatment	Australian Nuclear Science and Technology Organization (ANSTO)	Upgrading remediation Plants	[67,68]
Reverse Osmosis (three-step RO Plants)	Institute of Nuclear Chemistry and Technology (INCT), Poland	Remediation of liquid low-and medium-levels wastes of radioactive activity (LLLRWs and LMLRWs)	[69]

3.2 Nanofiltration

The nanofiltration membrane (NF) is a dense membrane structure that operates on a pressure-driven mechanism. NF membranes were first discovered in the 1980s. Its characteristics were found by comparing reverse osmosis (RO) to ultrafiltration (UF) [70]. An NF membrane's pore size is typically (1 nm). The composite membrane approach is the most efficient method for fabricating NF membranes. An ultra-thin dense layer is formed on a porous substrate to produce a composite membrane. However, alternate methods for providing NF composite membranes might be used [71,72]. The relatively small charge on NF membranes in an aqueous solution is due to solute adsorption charge or surface functional groups [73]. In the NF polymeric membrane manufacturing approach, ionized groups (including sulfonic acid and carboxylic groups) are usually used to form a charged surface in a feed solution. These groups can be essential, acidic, or combined depending on the specific chemical used during the synthesis process [74]. For removing small organic molecules and inorganic salts, NF membranes are comparably effective as reverse osmosis membranes. Furthermore, NF has a high retention of divalent ions and a low retention of monovalent ions. The NF membrane separation process is essentially dependent on particle size differences and charge influences (for ionic components) via steric and electrostatic interactions based on Donnan exclusion [75-77]. According to Shon et al. [78], the charge impact and solution diffusion processes separate components with low molecular weight or ionic species. Components having high molecular weight, on the other hand, are separated via a sieve method. Due to its low operating pressure and high permeate flow, NF might be used instead of reverse osmosis (RO) in various applications. Nanofiltration (NF) membranes are becoming more popular for water treatment because of their superior retention versus UF and lower energy usage than RO. These characteristics enable the NF membrane to be used in various applications. Water purification, medicines, biotechnology, wastewater cleanup, and saline water desalination utilize nanofiltration. In industry, the NF method is used to separate colors in the textile industry [79] and to remove heavy metals from wastewater [80]. NF has also been used for coke wastewater remediation [81], paper manufacture [82], oil/water system remediation from petroleum businesses, and eliminating acid sulphate from mine water [83]. In nuclear facilities, such as reactors, medical isotope production laboratories, and other nuclear technology industries, generate radioactive waste. Due to the severe concerns, the radioactive wastes could not be dumped into the environment immediately. Instead, the material should be pre-stored and subsequently treated to reduce waste to the least possible volume and activity levels. One option that might be used to achieve this purpose is the NF membrane. The NF membrane offers attractive qualities that make it preferable to alternative separation strategies in radioactive activity waste removal wastes, such as great selectivity, simplicity of module assembly, and cheap cost. Special additives might be used to differentiate a monovalent ion from a multivalent species to improve the selectivity of NF membranes. In the nuclear industry, boric acid is recovered from dirty cooling using an NF membrane in the nuclear reactor. Table 5 shows some instances of NF applications and studies carried out with the objective of implementation in nuclear power plants and research facilities.

Table 5: Examples of applications and studies employing NF membrane in the remediation of (LRW)

Membrane process	Facility/ country	Wastes processed	Ref.
Nanofiltration	Bugey Nuclear Powers Plants in (France)	Ionized silica and boric acid separation	[71]
Different (NF membranes) types; Osmotic Desal (Desal5 DK, Desal5DL and Desal percent1 HL) and Dow (NF90and NF45)	Institute for Water Research and Water Technology, D-65201 Wiesbaden, (Germany)	Selective strontium removal from a sodium nitrate aqueous.	[72]
Nanofiltration with complexation (two steps)	(Korea)	The radioactive strontium retention from liquid wastes of radioactive activities.	[73, 74]
Nanofiltration (with EDTA and DTPA)	(France)	Separation of gadolinium (III) and lanthanum (III) in aqueous.	[75,76]
NF/complexation (with tetrahydroxylated bis-crown-6calixarene)	(France)	Separation of traces of cesium /sodium from a greatly salted aqueous medium	[77]

3.3 Ultrafiltration

3.3.1 Ultrafiltration membranes

The ultrafiltration (UF) membrane technique concentrates and filters medium to high molecular weight components at working pressures ranging from 1 to 10 bar and pore sizes ranging from 0.01 to 0.1 μm . UF membranes function at lower pressures and with higher permeate fluxes than reverse osmosis [78]. The selectivity of Ultrafiltration membranes is determined by the difference in the pore size, molecular structure, and surface charge of the components to be separated. The phase-inversion approach is routinely used to construct most UF membranes with an asymmetric porous structure. Polysulfide (PS), polyether sulfone (PES), polyacrylonitrile (PAN), aromatic polyamides, and polyvinylidene fluoride (PVDF) are materials used in the manufacture of ultrafiltration membranes due to their low cost, wide pH range, and temperature resilience. Many ultrafiltration membrane applications have made use of these polymers [79]. One of the most fascinating applications discussed in this paper is using UF membranes to remove LRW. Section 3.3.2 discusses some of the research on this topic. Table 6 also includes several implemented situations. Polymeric membranes may be created in four shapes: flat sheets, tubes, hollow fibers, and spiral wounds. However, fouling has been a concern for UF polymeric membranes in the industry, limiting their usefulness. Fouling diminishes membrane efficacy and raises operational expenses due to maintenance and decreased membrane lifetime [80]. Therefore, the fouling reduction has been the focus of its practical implementation in recent years by altering UF polymeric membranes. To reduce fouling in UF membranes, several techniques have been used to improve UF polymeric membrane performance, including (1) surface modification via surface coating and grafting; (2) bulk improvement via the membrane blending technique with hydrophilic additives; and (3) radical polymerization. (3) the employment of inorganic nanomaterials as nano-fillers in polymer matrices to achieve the necessary membrane performance [81-84]. Additives are essential in ultrafiltration membrane fabrication, such as polyethylene glycol (PEG), polyvinyl pyrrolidone (PVP), polyethylene oxide (PEO), ZnCl_2 , and LiCl as inorganic salts, and water is the most often used additions. Organic substances include alcohol and glycerol. In the rule of additives, the additive might be used as a single component or as part of a combination [85]. Because of its hydrophilic and solubility, polyvinyl pyrrolidone (PVP) is an excellent pore-forming agent that modifies the microstructure of the membrane. Furthermore, PVP is recognized as an anti-biofouling agent, reducing biofouling of membrane surfaces by increasing the hydrophilicity of membrane surfaces [81,86,87]. Also, combining PVP-grafted copolymers or directly grafting on the surface of a polymeric membrane might alleviate the problem of membrane fouling [88]. Another significant pore-forming substance is polyethylene glycol (PEG), which has a hydrophilic characteristic. PEG has various molecular weights, including PEG200, PEG400, PEG6000, and PEG20,000 [89]. Another method of UF membrane modification is incorporating a cross-linking agent into the casting polymer solution to chemically bond the polymer and produce insolubility and toughness [90].

The operational flow mode in ultrafiltration equipment reduces membrane fouling too. UF membranes might function with either cross-flow or dead-end flow patterns. In dead-end filtration, the feed solution flows perpendicular to the membrane surface. In a cross-flow pattern, the flow is parallel to the surface of the membranes [91]. Because solids create at the surface of membranes, requiring regular back flushes and cleaning to maintain high flux, dead-end flow is advantageous in batch processes with minimal suspended particles. Cross-flow patterns are preferred in continuous operations because particles are constantly discharged from the surface of membranes, resulting in a thinner cake layer and lower resistance to penetration. [92]. Improved UF membranes have evolved in recent decades for various industrial processes used in wastewater remediation, water purification, medical devices, and food and chemical industries [93-96].

3.3.2 Ultrafiltration application in LRW

The primary function of ultrafiltration membranes is to remove colloids and other particles (suspensions) from feed liquid waste streams that are then processed by reverse osmosis systems. Actinide wastes are usually colloidal or suspended in radioactive activity wastes and can be easily removed using an ultrafiltration membrane [97]. Because the pore size of ultrafiltration membranes is too large to separate radioactive soluble ions from contaminated water, radioactive ions are size boosted by complexation, sorption, or precipitation methods before being an ultra-filtered process. The expression "seeded ultrafiltration" refers to using these three methods, as seen in Figure 3 [98]. Ultrafiltration is frequently used in conjunction with

complexation or precipitation. Precipitation generates less soluble particles (carbonates, phosphates, oxalates, ferrocyanides, or hydroxides), which are subsequently filtered via a UF membrane [99].

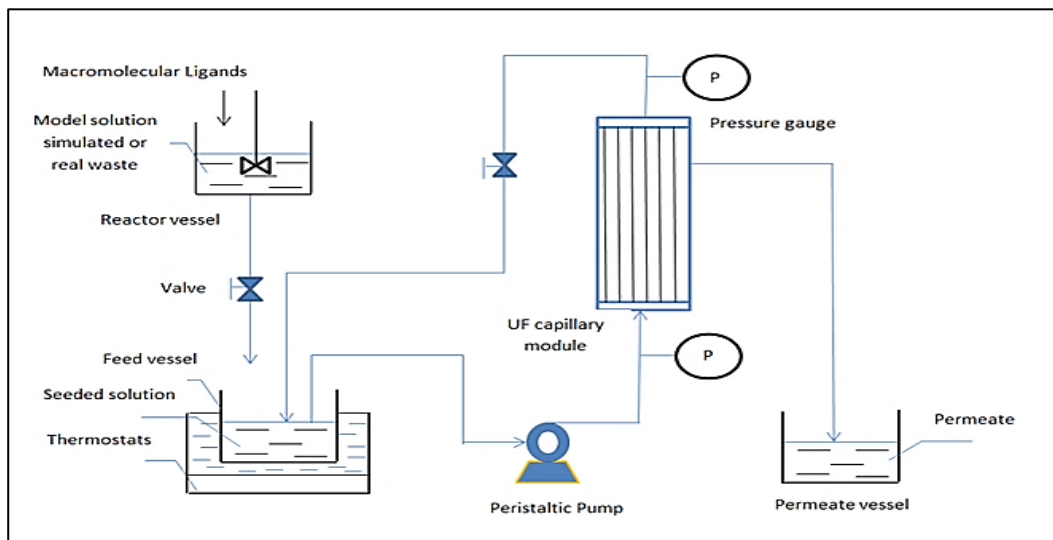


Figure 3: Schematic diagram of lab unit utilized for seeded Ultrafiltration test

Complexation is a technique used to improve the selectiveness of the UF process in eliminating dissolved very small metal ions from dilute aqueous solutions by adding high molecular weight chelating compounds to the entering waste solution to produce macromolecular complexes. The solution is subsequently filtered using an ultrafiltration membrane system, which rejects macromolecular complexes while allowing un-complexed ions to flow through. Most LRW activities generated by various nuclear plants, including reactor cleanup and recycling boric acid solution, are treated using seeded ultrafiltration. It is also commonly utilized in waste remediation stations to remove contaminated aqueous wastes [100]. Using complexing agents in combination with UF might give significant decontamination factors (DFs) for certain ions. As a result, it is vital to review prior efforts in the field of uses and applications of UF as a pre-remediation unit for RO. It additionally becomes critical to evaluate seeded ultrafiltration applications in eliminating soluble radioactive ions, which are used in many research institutes and laboratories. There is a lot of research, applications, and cases on this topic. Some of them, as well as their varied applications, will be addressed.

Ultrafiltration (UF) membrane utilized in combination with water-soluble polymers or surfactants containing meta-selective chelating agents (carbonate (CO_3^{2-}), dihydroxybenzenedisulfonic acid (DBSA), methylsulfonatonitrioltriacetic acid (SNTA), decyl nitrilotriacetic acid (DNNTA), undecyl-8-hydroxyquinoline (C_{11}HQ), and decylthiourea) has been studied by Scamehorn et al. [13]. Each of these ligands has been used to choose a certain ion. To remove and recover radionuclides and related harmful non-radio contaminants from groundwater or aqueous waste solutions, colloid-enhanced ultrafiltration (CEUF) methods are used. This project's target metal ions include uranium, thorium, lead, cadmium, mercury, and chromium. In lab scale testing, 99%-99.9% of each metal tested was removed in a single separation step.

Rao et al. [101] conducted a comprehensive study on cesium and strontium removal, respectively. Precipitation-UF membrane has been performed utilizing copper Ferrocyanides and calcium phosphate precipitation techniques. Supernatants have been exposed to Ultrafiltration (UF) utilizing a membrane with a pore size of $0.2\ \mu\text{m}$ on intermediate-level and low-level wastes produced in different aspects of the nuclear fuel cycle and had different chemical structures. The factors of decontamination (DF) have been calculated at 2 and 24-hour intervals with and without UF. The DF gained utilizing low-level wastes solution that has chemical properties comparable to cesium (^{137}Cs) has been in the (200-300) range for cesium (^{137}Cs) and strontium of (200) with LLW solution that has chemical properties like groundwater. Two hours of settling are sufficient for strontium (^{90}Sr) before UF. In the case of cesium, the DF magnitudes are not significantly affected by UF. Nevertheless, because the copper flocks Ferrocyanides precipitate are feathery, the UF has aided in separating solid-liquid. The influence of ionic strength and the existence of TBP on cesium and strontium removal efficacy has also been investigated.

Zakrzewska-Trznadel and Harasmowicz investigated membrane permeation with complexation for wastes of radioactive activity processing [18]. MEMBRALOX (tubes 250 mm long and 7/10 mm in diameter) and CÉRAME INSIDE (three-channel tubes) ceramic membranes have been utilized. To improve the UF separation impact, various soluble polymers, polyacrylic acid (PAA) with various molecular weight polyethyleneimine (PEI), and cobalt ferrocyanides (CoFC) have been employed. The complexing agents have been tested in the lab to identify the best process parameters. According to their research, decontamination factors increased substantially when macromolecular compounds were included. The efficacy of complexation by each ligand is greatly influenced by pH and alkali metal amount. The test demonstrates that cobalt hexacyanoferrate has been the most impactful complexing agent for binding cesium ions. When complexation by CoFC has been combined with UF (UF/Complexation), the DF of ^{137}Cs has been 109.8. Other metal ions had moderate DF. ^{154}Eu and ^{241}Am were also identified in permeate specimens derived from UF/CoFC testing. The DF for those radioisotopes has been 3.78 and 4.47, respectively.

Bisset et al. [102] created a novel polyethylene imine (PEI), a water-soluble polymer with N-methyl hydroxamic acid chelating groups. The study discusses the production of this chelating polymer, PEI-NMH, and its capacity to precisely separate chosen metal ions using UF, particularly in comparison to the previously described primary hydroxamic acid analog PEI-H. For

pH=1, PEI-NMH had a greater bound capacity to Fe³⁺ than thorium ion (Th⁴⁺), whereas PEI-H had the reverse impact. At pH=2, both demonstrated superior binding ability to other soluble chelated polymers. This indicates that slight changes in the chelating agent's structure substantially influence their binding ability.

The use of inorganic membranes for wastes of radioactive activity processing has been investigated by Zakrzewska-Trznadel [103]. The study examined using ceramic membranes (MEMBRALOX tubes with various pore sizes, 250 mm long and 7/10 mm in diameter, and CeRAM INSIDE three-channel tubes) in the Ultrafiltration range. Experiments have been conducted utilizing non-active and radioactive model solutions and genuine wastes of radioactive activity materials. To gain great decontamination factors, the technique has been designed as "seeded Ultrafiltration," the UF process improved by chemical complexation. Polyacrylic acid and polyacrylic acid salts of various crosslinking, polyethylene imine, and cyanoferrates of transitory metals have been investigated as complexing agents. A constant flow reduction induced by membrane fouling has been detected during the membrane-complexation process. The permeate flow reduction has been measured utilizing MEMBRALOX 5 kDa and a 0.5 g/dm³ Eu₂O₃ solution. In that event, membrane filtration was followed by complexation with a solution of NaPAA-8000 containing 10 g/dm³ (the amount corresponds to a ligand/ion ratio of 23:1). After 5 hours of operation, the flux fell by 16percent, and after 10 hours, the flow reduced by 78percent, indicating membrane blockage. Cleaning the membrane has been required to avoid complete blockage. It has been demonstrated that the hybrid technique had greater decontamination factors than UF membrane filtering alone. When (CoCF/NaPAA, 30,000 Da) has been added simultaneously, DFs have been obtained, as follows, ²⁴¹Am (101.4), ⁶⁰Co (55.38), ¹³⁷Cs (179.5), ¹⁵²Eu (96.39), and ¹⁵⁴Eu (26.95). Kedari et al. studied americium (III) recovery from nitric acid solutions utilizing anionic sodium dodecyl sulfate (SDS) and non-ionic polyethylene glycol ether (Tergitol 15-S-9) surfactant micelles [104]. In the batch, stirred cell Ultrafiltration unit, polyethersulfone membranes with nominal molecular weight cut-off (NMWCO); 3, 5, and 10 kDa have been employed to separate surfactant micelles. The impacts of NMWCO of the membrane, feed solution conditions, including the amount of surfactant, organic ligand 2-ethylhexylphosphonic acid mono-2-ethyl hexyl ester (H2A2), and metal ions on the efficacy of Am(III) removing and permeate flux have been studied. The permeate flux gained with Tergitol micellar solution is 74% more than that obtained with SDS micellar solution. The decrease in flux with increasing SDS amount is significant since the growing compactness of the SDS layer on the membrane induces membrane fouling. Still, it may also be since smaller micelles block membrane pores at a later step, as seen in Figure 4.

By utilizing SDS micellar solution up to pH= 3, the increased UF recovery of ²⁴¹Am almost completed the recovery of Am (III). Gross back extraction of Am (III) from the pseudo micellar phase is accomplished by raising the acidity of the metal-loaded rejection solution to greater than 0.5 M and adding the necessary quantity of nitric acid.

Singhal et al. [105] conducted lab simulation experiments to study the relationship ^{239,240}Pu with iron pseudo-colloids in groundwater specimens with dissolved organic carbon (DOC) at various levels from 10 to 60 mg L⁻¹. Ultrafiltration membranes have separated colloidal components into seven distinct size ranges ranging from 0.45 m to 1.1 nm. This research demonstrates that an aqueous environment rich in dissolved organic carbon (DOC) produced Fe pseudo-colloids via complexation between iron (Fe) and dissolved organic carbon. The DOC-coated Fe colloids preferentially absorb Pu. The surface shape and content of the Ultrafiltration membrane residue have been investigated. The findings support that the residue left on 10000 and 500 NMWL filter paper membranes with pore sizes of 1.6 and 1.1 nm, respectively, has greater amounts of carbon and Fe content than other fractions. Understanding the relationship between ^{239,240}Pu and Fe and DOC will aid in determining the long-term integrity of alpha-emitting wastes of radioactive activity storage facilities.

The membrane separation performance has been improved via additives that improved the rejection of the membranes by enlarging the size of target radionuclide species Svittsov et al. [106]. Polyacrylphosphonic acid (APA) has been demonstrated to be the best efficient cobalt binding agent, next to polyethyleneimine (PEI) and polyacrylic acid (PAA). Cobalt has been retentate at a rate of more than 98%. Cesium has been efficiently bound by nickel hexacyanoferrate that aided in its retention via a membrane. Cesium rejection might be boosted further in the presence of the surfactant dodecylsulfate. Surfactant created large micelles which adsorbed microparticles of nickel-cesium hexacyanoferrate. As an outcome, the cesium purification coefficient has risen about 200 percent compared to surfactant-free and 400 mg/L surfactants.

Zakrzewska-Trznadel and Khayet investigated the removal of ⁶⁰Co, including cobalt nitrate, by UF membranes utilizing water-soluble polyethyleneimine (PEI) [19]. The cobalt (Co²⁺) retention for surface-improved macromolecules (SMM) blended polyethersulfone (PES) membrane has been greater than 97%. SMM mixed PES, pristine PES, and commercial PES membranes have DF magnitudes of 223–163, 75, and 44 consecutive. Furthermore, SMM blended PES membranes had 4 to 5 times lower ⁶⁰Co adsorption than pristine PES membranes.

Reduced volume and the amount of long-lived radioactive materials are significant challenges in handling low-level liquid wastes of radioactive activity. Dang, Thi Thu Hien, et al. [107] conducted research and comparison of the performances of low-pressure reverse osmosis (LPRO) and polyelectrolyte-enhanced Ultrafiltration (PEUF) for the removal of target metals (Co and Sr) from simulated low-levels wastes of radioactive activity. This work employed chelating polymers, including polyacrylic acid (PAA) and polyethyleneimine (PEI). The rejection efficiencies of (Co and Sr) by the LPRO membrane have been almost constant regardless of PAA dose or pH levels. However, adding PAA resulted in much more membrane fouling. Figure 5 illustrates the removal efficacy of Co and Sr via a combination of a UF membrane and PEI. The result demonstrated elimination higher than 80% of Co ions at an NH: M (+2) molar ratio greater than 50; nevertheless, unlike the PEUF with PAA, PEI gained relatively low Sr removing less than (40%) since PEI has an excellent binding for Co but a low binding for Sr.

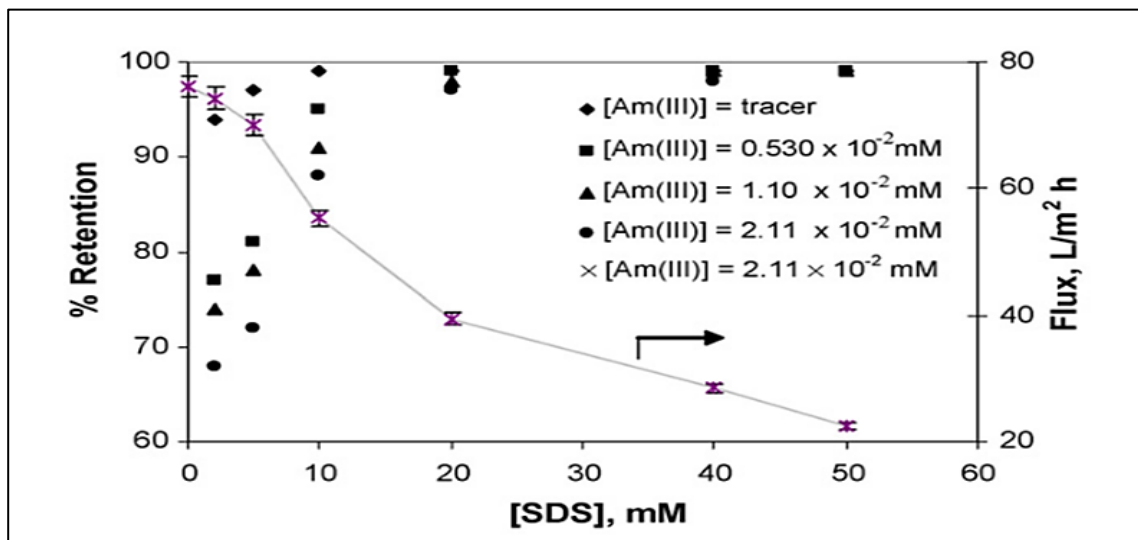


Figure 4: The influence of SDS amount on the retention of Am (III) and permeate flux. Feed solution SDS + Am (III) ($[Am(III)] = 241 Am(III) \text{ tracer} + Eu(III)$) at $pH 3.00 \pm 0.11$. Membrane—5000 Da NMWCO [104]. © 2009 Elsevier Ltd

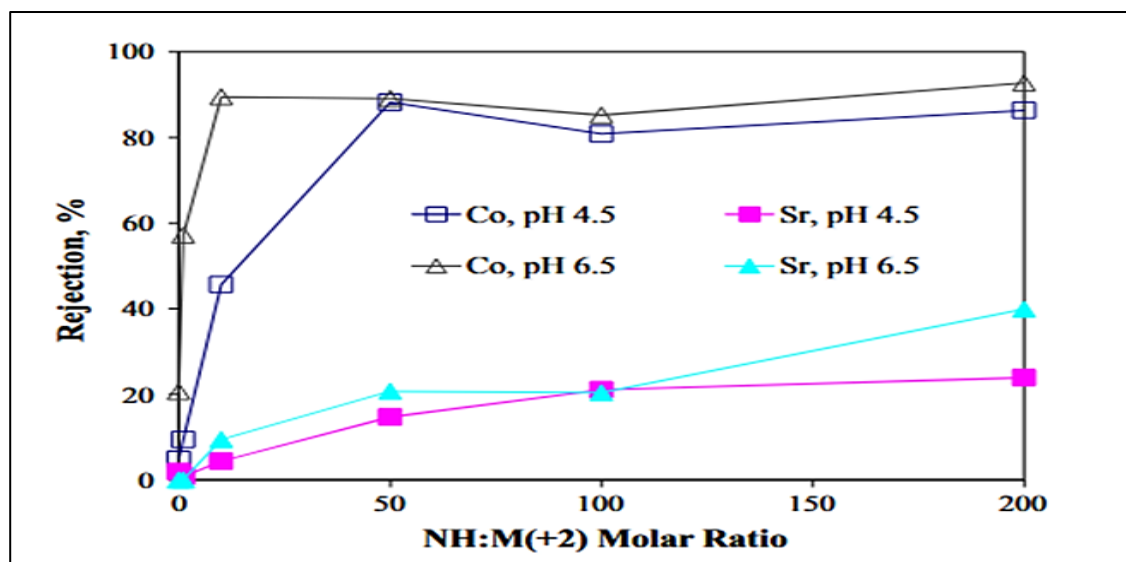


Figure 5: Removing efficiencies of Co and Sr ions at pH magnitudes of 4.5 and 6.5 utilizing PEUF systems with 750-kDa PEI at different molar ratios of amine groups to metal ions [107]. © 2016 Elsevier Ltd

Zhang et al. [108] examined the impact of the cationic surfactant hexadecyl trimethyl ammonium bromide (CTAB) on metal salt rejection during Ultrafiltration of low-level radioactive wastewater (LLRW). In the presence of CTAB at amounts lower than the critical micelle amount (CMC), nuclide rejection rose from 24–33% to 50% for Cs (I) and over 90% for Sr (II), Co (II), and Ag (I). It is demonstrated that the rejections enhanced since the CTAB fouling layer enhanced the surface of membranes to be more hydrophilic and positively charged on the surface of membranes. The nuclide content of the fouling layer increased with CTAB amount, and at 400 mg/L CTAB, 55–80 % of Co (II), Sr (II), and Ag (I) were adsorbed on the membrane. The production of Ag Br contributes to the great deposition of Ag (I). Depending on these findings, modest amounts of cationic surfactant could considerably improve cation UF rejections, although membrane fouling and nuclide deposition should not be overlooked. According to the study findings, at greater amounts of CTAB and nuclides, a lower CMC magnitude findings in the production of CTAB micelles, a thicker fouling layer as demonstrated in Figure 6, and reduced membrane porosity. As a result of the tight exclusion impacts, additional nuclide rejection and deposition processes occur.

Suping et al. [20] worked to modify the UF membranes (polyvinylidene fluoride) (PVDF) utilizing poly(vinylpyrrolidone) (PVP) thru a two-step surface grafting technique to improve the antifouling properties and membrane rejection of the simulated low-levels radioactive wastewater that included Cesium nitrate ($CsNO_3$), cobalt nitrate hexahydrate ($Co(NO_3)_2 \cdot 6H_2O$), strontium nitrate ($Sr(NO_3)_2$), and silver nitrate ($AgNO_3$). Small molecular PVP has been utilized to change the membrane pores, whereas cross-linked PVP has been utilized to modify the surface of membranes. The improved membrane decreased flux significantly more slowly than the pristine membrane. The flux recovery rates of the BSA-fouled membranes after water washing have been 98% for the improved membrane and 46% for the pristine membrane, respectively. Furthermore, the improved membrane rejected slightly the same nuclide and surfactant as the raw membrane, as demonstrated in Figure 7. Depending on these findings,

the two-step improved membrane is appropriate for use in LLRW remediation, with the benefits of less frequent membrane cleaning and a longer membrane lifetime.

Singha et al. [104] designed a hybridization process for the three-step system (a) complexations, (b) UF, and (c) Remediation of the resin bed. A five-liter lab-scale experimental capacity set-up has been utilized to demonstrate the nuclear Plants-produced waste system. Water-soluble polymers, including polyethyleneimine (0.1%), and complexing agents, including polyvinyl alcohol (0.1 %), have been utilized in the complexation step, about 0.12 percent total metal ions either individually or in combination, including Uranium 1000 ppm and other metals at pH (5.6). Then, they have been crossed thru the UF membrane system. Next, a (polyacrylamide hydroxamate) resin bed was utilized. According to the findings, initially complexing with PEI eliminates the metal during the major step. Adding PEI to the solution and subsequently passing it thru UF eliminated about 97% of the Fe (II), Cr (III), Al (III), and U (VI). Cu (II) has been eliminated at a rate of 40%. Sr (II) exhibited little retention. The hybrid operation has gained about 100% removal of the metals from the aqueous waste. Many nuclear Plants have utilized UF technologies, and numerous works have been performed to apply ultrafiltration for wastes of radioactive activity remediation. Some of these works are described in Table 6 below.

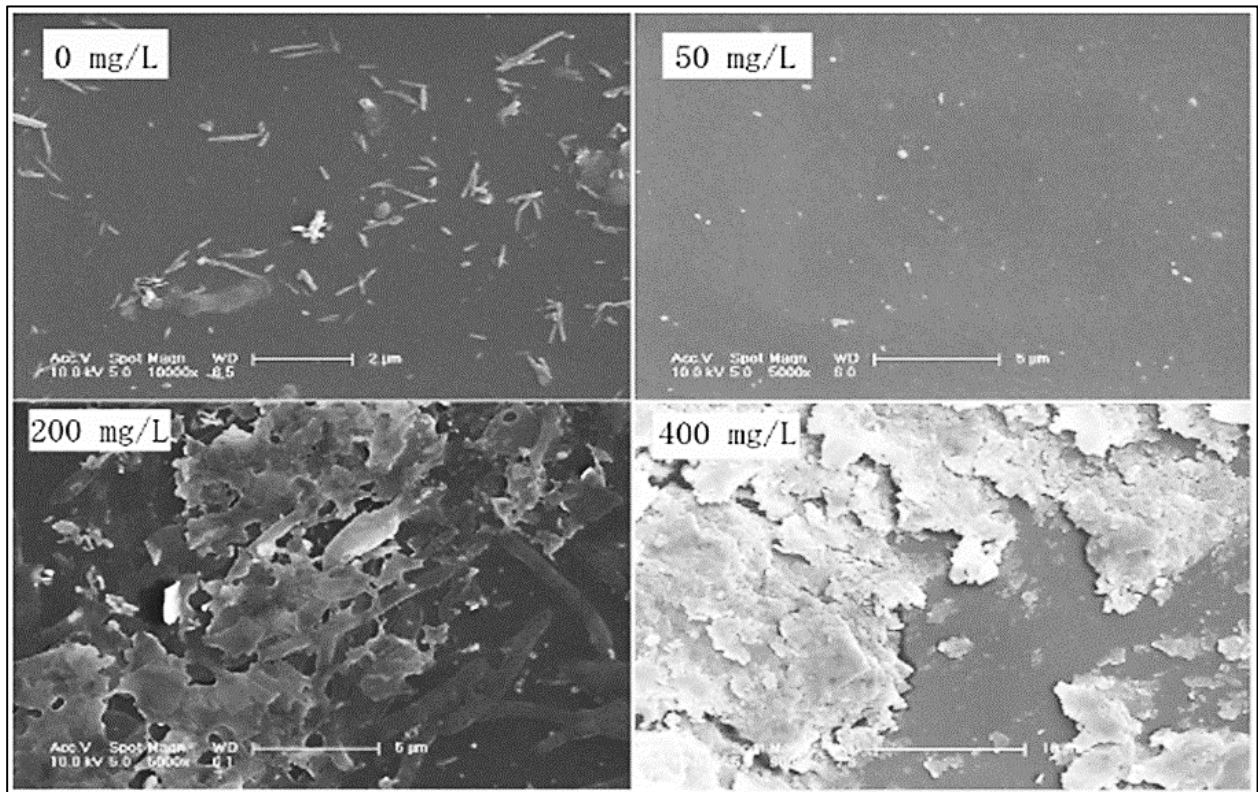


Figure 6: SEM images of membranes fouled by solutions of 0–400 mg/L CTAB (labeled on the images) and one mg/L nuclide [108]. © 2017 Elsevier Ltd

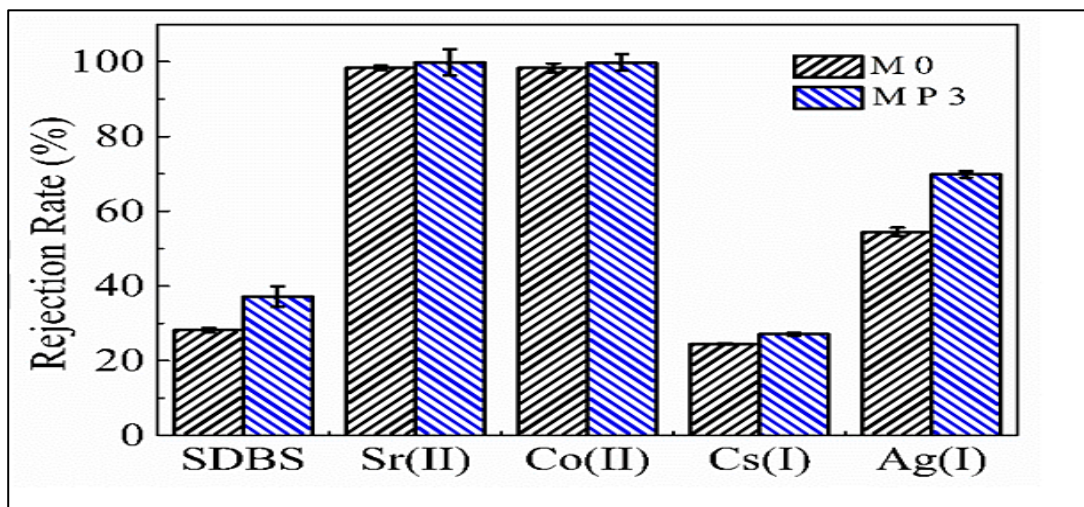


Figure 7: Nuclide ions and SDBS (200 mg·L⁻¹ SDBS, 0.1 mg·L⁻¹, pressure 0.1 MPa, 19.6 cm² PVDF, 25°C), rejection rates, M0 and MP3 represent the raw and improved membrane, respectively [20]. © 2018 Elsevier Ltd

Table 6: Examples of UF membrane processes applied for wastes of radioactive activities

UF type/process	Facility/country	Processed wastes	Ref.
Inorganic tubular Ultrafiltration / 2-steps UF.	Enhanced Actinide Removing Plants at Sellafield (UK)	The amount of solid material of a few percent has been accomplished. This is the first step, and then further sludge dewatering occurs at the later step.	[109]
Seeded Ultrafiltration	Harwell Lab, at UK,	Separate the radioactive floc and liquid.	[109]
Plate and frame UF modules	Paks nuclear powers Plants /Hungary	Cleaning and recycling polluted boric acid solutions.	[117]
Precipitation, co-precipitation with UF technique	Nukem, Hanau	Low-levels of waste arising from fuel fabrication Plants.	[110]
Ultrafiltration unit	River Bend nuclear powers Plants (USA)	Boiling Water Reactor (BWR) floor draining.	[38]
	Salem nuclear power Plants. (USA)	Pressurized Water Reactor (PWR) floor draining, laboratories, specimen points, and auxiliary equipment draining.	
	Seabrook nuclear powers Plants (USA)	PWR floor draining and spent resin tank draining down.	
Four Ultrafiltrations modules.	Callaway nuclear powers Plants (USA)	Floor draining, equipment draining, reactor coolant	
Two Ultrafiltrations' first step (mobile tubular UF plan), the second step (UF skid-mounted Plants)	Diablo Canyon nuclear powers Plants (USA)	Spent media transfer liquid.	
Seeded Ultrafiltration	Cadarache Nuclear Research Centre	Remediation of liquid laundry waste.	[111]
Ultrafiltration with complexation	Colorado, USA	Boric acid removing from water solutions.	[112]
Ultrafiltration with complexation	Los Alamos national laboratories	Decreasing the volume of radioactive sustains originating from decontamination and decommissioning nuclear facilities	[113,114]
Ultrafiltration with two complexation procedures	Bhabha atomic research center	1st, it has been utilized for removing radioactive. Next, it has been utilized to remove complex uranium and enhance this isotope from environmental waters.	[115,116]

3.4 Microfiltration (MF)

Microfiltration (MF) is the most common membrane method. It all started around the turn of the century with the creation of synthetic microporous membranes based on cellulose [132]. MF is based on the sieving concept, with pore diameters ranging from 0.1 to 10 μ m. Depending on the dimension, the separation process is based on sieving and particle separation; however, some charge or adsorptive separation may occur. Compared to other filtering procedures, the applied pressure in microfiltration is comparatively low (less than 2 bar). In MF membrane operation-dependent applications, polymeric and ceramic membranes are used in numerous industries [117]. Organic polymers, inorganic ceramics, metals, and glass make MF membranes. MF membranes are manufactured using a variety of procedures, such as phase inversion, track-etching, stretching, and sintering. The pore structure is frequently symmetrical, with porosities exceeding 80% [134]. Ceramic membranes, on the other hand, are resistant, with a long lifetime and minor fouling tendencies. Furthermore, its resistance to extreme circumstances enables various cleaning techniques to prevent fouling. Ceramic membranes are hence the optimum choice for industrial applications. However, the high initial cost is the most significant barrier restricting their utilization [118]. The characteristics of the MF membrane. Other sectors that have benefited from microfiltration membranes include water and wastewater remediation [137], heavy metal wastewater remediation [138], food and dairy industries [139], pharmaceuticals [140], oil removal from wastewater [141], and biotechnology [142]. Microfiltration is used in the treatment of radioactive wastewater. In the 1980s, the microfiltration technology for purifying radioactive wastewater was first used in industry. The microfiltration technology may effectively remove 241 Am from low-level radioactive effluent when combined with ferric chloride as a flocculent. Assume the initial wastewater radioactivity is 809.2 Bq/L, and the effluent radioactivity is less than one Bq/L. In such a scenario, the clearance rate may exceed 99.9%, and the decontamination factors (DF) have exceeded 1651.4, suggesting that the technology positively influences radioactive wastewater removal [119]. When the dose of Fe^{2+} is (35- 60 mg/L) in the process consisting of ferrous sulfate as a flocculent and coupled microfiltration, the removal rate of plutonium may approach 99.9% [120]. In treating radioactive wastewater, microfiltration is often used as a remediation procedure. It can be used in conjunction with adsorption and flocculation techniques. Using the combined technology of immersion microfiltration membrane reactors and adsorption complexation, the cesium quantity could be reduced from 106.87 g/L to 0.59 g/L, with a removal rate of 99.44%. The average number of DFs and quantity factors were 539 and 208, respectively [121]. The co-precipitation followed by MF (CPMF) approach combines the advantages of co-precipitation and MF. Precipitation alone was unable to remove particles from liquids, resulting in a poor factor of decontamination (DF). Nevertheless, a membrane's flux drops fast without a precipitation phase. The strontium content in the water specimen utilized in this investigation has been about 5 mg/L, comparable to low-level wastes. In

two intermittent runs, 2 doses of $\text{Na}_2\text{CO}_3 \cdot 10\text{H}_2\text{O}$ (sodium carbonate) (2000 and 1000 mg/L) have been utilized for ($\text{Na}_2\text{CO}_3 \cdot 10\text{H}_2\text{O}$) precipitation from the water specimen, that has been subsequently exposed to microfiltration, DFs of 237 and 158, as well as average amount variables (CFs) of 288 and 462, have been found, respectively [122]. Adsorption/MF hybrid system for selective removal of potentially harmful strontium and cobalt ions from simulated laundry wastewaters. A crystalline silicotitanate (CST) and four distinct iron oxides with varied crystal structures and adsorptive processes, such as surface coordination and ion exchange, have been created and used as adsorbents. Adding up to 30% adsorbents to the microfiltration system enhanced membrane permeability. For eliminating ^{60}Co and ^{90}Sr ions from wastewater, combining CST with an MF membrane appeared to be the most appealing option [123]. More information about these studies and the latest publication on MF applications is included. Table 7 summarizes some examples of hybrid processing to treat liquid radioactive wastes.

Table 7: The application of MF membrane in wastewater remediation [26]

Hybrid process	Target isotopes	Result	Ref.
Adsorption–microfiltration with polyvinylidene fluoride (PVDF)	^{137}Cs	DF = 208 Strong membrane fouling	[124]
Microfiltration with PVDF hollow fibers and $\text{Fe}(\text{OH})_3$ flocculation	^{241}Am , ^{90}Sr	99.9% ^{241}Am rejection, DF = 1650	[125]
Microfiltration with sodium carbonate, pore size 0.22 μm	Sr^{2+}	DF = 460	[126]

3.5 Membrane Modification Utilizing Nanoparticle Additives

Nanoparticles (NPs) enhance membrane properties, improving antibacterial and anti-fouling, increasing rejection efficiency, and mechanical and thermal stability. These nanoparticles are dispersed in polymer casting solutions, providing innovative polymeric membranes with unique properties like electrical, magnetic, optical, thermal, and mechanical stability. Their wide surface area and strong behavior could enhance membrane separation activity in remediation procedures. Chen et al. [127] utilized a phase inversion approach to building hybrids PES membranes with halloysite nanotube and copper ions as nano-fillers, resulting in nano-hybrid membranes with adequate bio-fouling controls. Alsahly et al. [128] investigated the effect of embedding ZnO-NPs on the PVC performance of the membrane. An anti-biofouling polyvinyl chloride/zinc oxides (PVC/ZnO) membrane had been provided utilizing the phase precipitation technique for implementation in a University of Cape Town membranes bioreactors-submerged membranes bioreactors (UCT-MBR) for remediation of exact hospital wastewater, and the adding of ZnO nanoparticles, up With 0.1 g of ZnO, the membrane's purified water permeability (PWP) increased by 315%. With increasing ZnO nanoparticles up to 0.3 g, the cake layers build-up on the surface of membranes decreased from 52.8 to 10.42 m, but 0.4 g ZnO did not affect the cake film thickness. Compared to 29 days for a neat PVC membrane, the long-term performance of PVC-0.3 g nanoparticles has been enhanced to 70 days before membrane cleaning. The chemical oxygen demands (COD) removing efficacy of the UCT-MBR technique has been consistent across all membranes, averaging about 73.5%. Employing the induced phase inversion approach, Alsahly et al. [129] investigated the performance of the PPSU membrane of poly(phenyl sulfone) (PPSU) membrane contains utilizing varied amounts of ZnO nanoparticles as admixtures. According to this research, adding ZnO nanoparticles to the membrane increased its hydrophilicity. With 0.025 wt.% ZnO as additions, the flow of the PPSU membranes increased from 76 to 107 ($\text{L m}^{-2} \text{h}^{-1} \text{bar}^{-1}$) with no significant change in solvent rejection. The influence of inserting TiO_2 nanoparticles on PVC for long-term UF membrane performance for treating refinery wastewater has been explored by Al-Ani et al. [130]. These findings demonstrated the effect of TiO_2 nanoparticle amount on the long-term performance of PVC/ TiO_2 -NPs. They also verified that by modifying the characteristics of polymeric membranes with TiO_2 nanoparticles, it seems feasible to increase the lifespan of membranes throughout the long-term operation. Mechanical and thermal resilience, large surface area, hydrophilic nature, non-oriented porosity distributions, and regular spherical shape are all characteristics of zeolite nanoparticles. Because of these characteristics, it is commonly utilized in membrane separation processes such as gas separation, desalination, and Ultrafiltration. Rezakazemi et al. [131] investigated gas transport in polydimethylsiloxane (PDMS)/zeolite. Pure gas permeation tests were utilized to identify mixed matrix membranes (MMMs). Once compared to the clean PDMS membrane, the MMMs had greater selectivity of H_2/CH_4 and H_2 permeability, indicating that these membranes were particularly promising for gas separations, including H_2/CH_4 separation. Moreover, using a non-solvent induced phase inversion technique, Y.Yureki [132] investigated the adsorption and filtration techniques combined with NaX zeolite nanoparticle artificially inseminated polysulfone (PSf) membranes thru the produced nanocomposite membranes to eliminate the nickel and lead cations from wastewater. After 60 minutes of filtration, at 1 bar of trans-membrane pressure, the maximal adsorption rate of the hybrids membrane for lead and nickel ions was determined as 682 and 122 mg/g, respectively. The nanoparticles might also be utilized to extract colors from wastewater. Through the phase transition approach, Rana et al. [133] utilized graphene oxide nanoparticles (GO-NPs) to change the polyether sulfone (PES) membranes and construct mixed matrix membranes (MMMs). The addition of GO to the PES casting solution resulted in longer membrane lifetimes since improved fouling resistance and flux recovery efficacy (FRE) after backwash, as dye removal was greater than 99% for all membranes began studying and both dyes (AB-210 and RB), at dye amounts of 10, 50, 80, and 100 ppm and trying to operate pressures of 3 bar. Abdullah et al. [134] confirmed novel photocatalysts PES-WO_{2.89} nanoparticles membrane ultra-filtration for removing dyes, with the optimum 0.4 wt. percent loading of WO_{2.89} exhibiting smoother surface along with min average pore sizes and optimistic membrane manifesting the lowest contact angle and greater retention vs. Congo red dye. The Flux recovery ratio FRR of the nanocomposite membranes was further improved by subjecting them to UV irradiation, with the flux rising from 71.5% for the neat membrane to 83.3% at 0.4 wt.% content. In recent decades, carbon nanomaterials-depending polymeric membranes have been utilized in numerous

industrial applications since their unique structural and electronic properties, good mechanical properties, better antifouling performance, antibacterial agents, enhancement of the membrane characteristics, and great membrane rejection, including carbon nanotubes (multi-wall MWCNTs and single-wall CNTs), graphene oxide, and mesoporous carbon, etc [135-139]. Jalal et al. [140] investigated the effects of MWCNT-g-GO content on the performance and characteristics of PVC/MWCNT-g-GO membranes that were formed utilizing a traditional phase inversion technique to eliminate chemically oxygen demands (COD) from oily wastewater. The findings revealed significant impacts of MWCNT-g-GO on the membranes' structural morphology and fostered increased water permeation flux, which was 66% greater. Furthermore, natural materials combined with graphene might be utilized to enhance the characteristics of the polymer membranes. Ali et al. [141] looked into the effects of Gum Arabic-modify Graphite (GGA) that was manufactured utilizing a simple green technique and utilized for the first time as an addition to improving the characteristics of the PPSU Ultrafiltration membrane. Furthermore, compared to pristine PPSU membranes, all membranes manufactured with reduced GGA amount could impart greater hydrophilic nature and permeability properties. Silica nanoparticles have been utilized to change polymeric membranes in many studies. Al-Araji et al. [142] discussed the modification of PES membranes with inorganic nanoparticles and/or organic polymeric additives such as PEI and inorganic nanoparticles such as SiO₂, as well as how the amount and types of these admixtures affect the final nanocomposite's recoverability, selectivity, permeability, wettability, and stability over time, as well as the strengthening of incorporated inorganic nanoparticles into polymeric PES membranes.

On the other hand, nanoparticles could be utilized to eliminate radioactive isotopes from polluted radioactive wastewater by adding them directly to contaminated aquatic solution thru batch processes or to membranes to enhance their adsorption and ion exchange capacity in continuous processes. Ding et al. [21] utilized a copper ferrocyanides (CuFC) film and SiO₂ nanoparticles to study and manufacture new PVDF membranes. Their findings revealed that SiO₂ nanoparticles had been significantly deposited on the surface of the membrane, and the CuFC film had been securely connected to the SiO₂-NPs and the surface of the membrane. In filtering humic acidic media and pure surface water, Cs rejections might still gain 99.4% in 8 hours. The membrane has successfully eliminated Cs from water utilizing direct fast filtering, indicating that it may be a promising technique for radioactive wastewater remediation. Additionally, various materials have been utilized to eliminate radioactive ions to eliminate harmful radioactive ions in a contaminated liquid. Wang and Zhuang [143] highlighted the many materials utilized to eliminate radioactive ions via numerous organic and inorganic adsorbents as additives utilized with the different remediation techniques of radioactive isotopes, including metals-organic frameworks and ion exchanging resin, clay minerals, and metals hexacyanoferrates). Table 8 exhibits the most common materials employed to eliminate radioactive ions.

Table 8: Some of the materials utilized to eliminate radioactive isotopes from polluted liquids

Material / Additives	Radioactive isotopes	Treated wastes	Ref.
Copper ferrocyanides (CuFC)	Cesium ion (Cs)	Radioactive liquid waste	[144]
Prussian blue (PB)	Cesium ion (Cs)	Drinking water	[145]
Cobalt ferrocyanides (CoFC)	Cesium ion (Cs)	Wastewater	[146]
Natural zeolites (clinoptilolite)	¹³⁷ Cs, ⁶⁰ Co, ⁹⁰ Sr and ^{110m} Ag	liquid radioactive waste	[147]
NaY zeolite	Uranium (VI)	Aqueous solutions	[148]
TiO ₂ NP-chitosan and ZrO ₂ NP-chitosan	Gadolinium and samarium	Water	[149]
A and X zeolite	¹³⁷ Cs and ⁹⁰ Sr	Seawater	[150]
Graphene oxide (GO)	Cesium ion (Cs)	Radioactive liquid waste	[151]
chromic doped TiO ₂ nanotubes (Cr-TiO ₂)	¹³⁷ Cs and ⁹⁰ Sr	Groundwater and seawater	[152]
Multiwall carbon nanotubes (MWCNTs)	Cesium ion (Cs)	Aqueous solution	[153]
Ammonium molybdophosphate – polyacrylonitrile (AMP-PAN)	Cesium ion (Cs)	liquid radioactive waste	[154]
NaY zeolite –PES nanocomposite membranes	Cesium (¹³⁷ Cs)	liquid radioactive wastewater	[155]

4. Conclusion

Pressure-driven techniques, including MF, UF, NF, and RO, effectively treat radioactive effluents, removing insoluble compounds like colloids and heavy radioactive materials. These procedures are crucial for nuclear industry waste remediation, posing a significant threat to human and environmental health. LRW cannot be directly released into the environment unless treated by one of the remediation techniques. One of these techniques is membrane technologies and their various processes to eliminate radioactive materials or reduce their levels to release limits. The essential factor in choosing one of the pressure-driven processes utilized alone or combined with another auxiliary process depends on the feed solution's chemical, physical, and radiological characteristics. Reverse osmosis treats low- and medium-level radioactive materials and ions in liquids, followed by ultrafiltration or microfiltration. This process produces high decontamination and retention coefficients, making it widespread at nuclear sites. Additives improve the selectivity of NF membranes by distinguishing monovalent and multivalent ions. UF membranes remove colloids and particle foulants from waste flow, while ultrafiltration membranes remove dissolved radioactive ions through mechanisms like sorption, complexation, or precipitation. These processes are typically done before ultrafiltration. Complexation enhances UF process selectivity by adding chelating chemicals and creating macromolecular compounds, producing decontamination factors for specific ions. Membrane materials are chosen based on radioactive waste activity levels. Polymeric membranes are more widely used due to lower cost and higher packing densities. However, they have limitations like

contamination, limited lifespan, and low radiation resistance, making them unsuitable for industrial use. Ceramic membranes offer ionizing radiation resistance, pH range, and temperature resistance, making them the ideal choice. The microfiltration process involves sieving and particle separation, with low-pressure polymeric and ceramic membranes used for pre-remediation procedures. Preliminary procedures like precipitation, co-precipitation, and complexation can remove radioactive particles from contaminated liquids before membrane filtration. Fouling increases energy consumption, hydraulic resistance, and system components, affecting performance. Membrane technology offers superior performance but is heavily reliant on cost-effectiveness. The adsorption or ion exchange method of embedded nanoparticles with membranes is considered cost-effective due to its lack of huge requirements and secondary waste generation. However, advanced materials combined with membranes can offset this cost. Recyclable and environmentally friendly nanomaterials can solve the cost problem in nanocomposite membranes. Low-cost or renewable materials can also improve cost-effectiveness by refurbishing consumables, which typically have low adsorption capacities. Renovating these consumables can be cost-effective when included with membranes to remove radioactive isotopes from contaminated water.

Author contributions

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The data that support the findings of this study are available on request from the corresponding author.

Conflicts of interest

The authors declare that there is no conflict of interest.

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