Engineering and Technology Journal 40 (09) (2022)1231-1259

Engineering and Technology Journal
Journal homepage: https://etj.uotechnology.edu.iq

Remove Liquid Radioactive Wastes Utilizing Nanofiltration, Ultrafiltration, and Microfiltration Membranes

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HIGHLIGHTS

- The current short review addresses developments in treating and removing radioactive effluents (LRWs).
- Pressure-driven membrane separation could be used to treat radioactive wastewater instead of current procedures.
- Nanoparticle-incorporated polymeric membranes are used to handle radioactive waste.

ARTICLE INFO

Handling editor: Mustafa H. Al-Furaiji

Keywords:
Liquid radioactive waste; Decontamination factor; Concentration factor Nanofiltration; Ultrafiltration; microfiltration; Nanoparticles; Seeded ultrafiltration; Cleanup process.

ABSTRACT

Radioactive waste is generated from fuel cycle processes in nuclear reactors and nuclear power plants (NPPs) in electrical power production, radioisotope manufacturing in nuclear research centers, and medical, industrial, and agricultural applications. Also, natural chain-linked radioisotopes (NORM) are generated from processing and burning fossil fuels and producing oil and natural gas. Therefore, a planned and integrated radioactive waste management strategy must be adopted to protect human health and the environment from the dangers of this waste through published research on a comprehensive radioactive waste management strategy and the testing and dissemination of several treatment options. The main objective is to draw the scientific community's attention to the possibility of using pressure-driven membrane separation in treating radioactive wastewater compared to conventional methods. This short review addresses developments in the treatment and removal of radioactive effluents (LRWs) by pressure-driven membrane methods and improvements in routine treatment of dissolved radioactive ions by chemical treatment of the feed solution followed by membrane separation. Also, recent advances in treating radioactive waste use nanoparticles (NPs) incorporated in polymeric membranes.

1. Introduction

The growing population and the desire for contemporary technology in all aspects of life have resulted in a growth in electricity consumption, necessitating the creation of extremely huge amounts of this energy all over the globe to make up for the deficit. This energy can be generated from a variety of sources, including nuclear power plants (NPPs), which generate large amounts of electrical energy while also contributing to the reduction of carbon emissions generated by the combustion of fuels (e.g., natural gas and crude oil) in other power plants, which emit massive amounts of carbon dioxide into the environment, causing contamination [1]. Furthermore, nuclear energy may be utilized for various purposes, including: (agriculture, pharmaceuticals, industry, and nuclear research centers). However, the use of nuclear energy technology in many industries must adhere to safe environmental conditions and laws because of the radioactive waste it produces, damaging individuals and the environment in the long run. As a result, many nations that use this technology try to minimize the spread of radioactive waste in the environment by categorizing, processing, and storing it in a safe and regulated way [2]. Figure

http://doi.org/10.30684/etj.2022.134025.1218
Received 30 May 2022; Accepted 18 July 2022; Available online 07 September 2022
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1depicts the numerous sources that create these radioactive wastes and their classification based on their physical, chemical, biological, and radiological states since the risk of this radioactive waste rests in its states and the degree of radiation released from it. Furthermore, understanding the classification of the status and level of radioactive waste, such as exempt waste (EW), low-level wastes (LLW), low and intermediate-level waste (LILW), and high-level waste (HLW), facilitates the selection of safe treatment methods to eliminate the risks arising from the levels of radioactive contamination from these wastes, as well as to provide secure storage costs to limit its spread. On the other hand, the treatment of liquid radioactive wastes (LRWs) is more complex due to their huge volume. To that end, it is vital to concentrate on treatment approaches that minimize these amounts without creating secondary liquid waste that is often more harmful radiologically and chemically than the initial liquid waste [3].

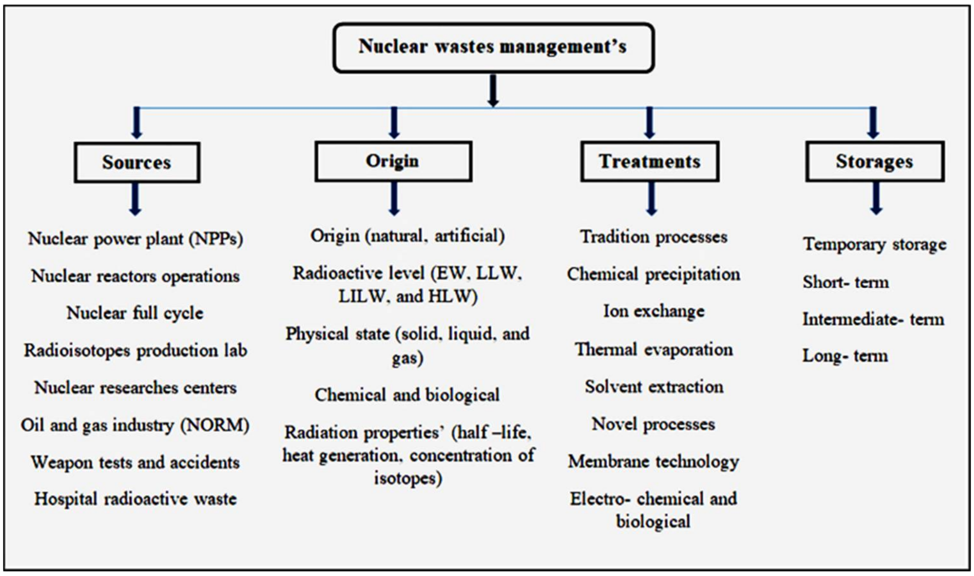

Figure 1: Nuclear wastes managements

Traditional procedures such as precipitation with sedimentation, ion exchange, and evaporation use energy or add an additional phase, resulting in secondary waste creation (sludge from sedimentation tanks, spent sorbent from ion-exchange columns, or effluents from resin regeneration). This radioactive waste will need further treatment and purification. All of these disadvantages can be avoided by using membrane methods, which have advantages such as (requiring relatively low energy amounts compared to other separation processes such as distillation, easy scaling-up by adding modules, operating at relatively low temperatures, and being capable of forming a hybrid system combined with other separation processes) [4,5,6]. Table 1 outlines some of these methods, stressing their potential benefits and downsides. Membrane-based approaches have tremendous promise in the treatment of radioactive waste.

Table 1 shows the importance of using membrane separation methods, particularly pressure-driven methods classified according to the size of the pores, such as reverse osmosis (RO), nanofiltration (NF), ultrafiltration (UF), and microfiltration (MF), as these membranes were used to eliminate various radioactive materials, such as cesium (137) and coblte (^{60}Co). Kumar et al. [8] evaluated whether commercially available RO membranes such as cellulose acetate, polyamide, or thin-film composites (polysulfone-polyamide) can successfully treat wastewater contaminated with heavy metals and radioisotopes. Salt rejection (SR) efficiency for cesium and molybdenum surrogates was shown utilizing commercially available membranes. Furthermore, it was discovered that polysulfone-polyamide composites outperform other membranes in terms of SR efficiency under varying operating parameters such as salt concentration, pressure, and temperature. Pressure-driven processes may be employed to remove suspended particles, boric acid, and colloids from liquid radioactive waste [7]. Despite the benefits of membrane separation methods, there are obstacles to using them in the treatment of liquid radioactive waste, particularly waste containing dissolved radioactive isotopes such as cesium (^{137}Cs), iodine (^{133}I), strontium (^{90}Sr), and cobalt (^{60}Co), because these isotopes have small ionic radii, allowing them to pass through the membrane pores (e.g., NF, UF, and MF). Another barrier is the material used to make the membrane since large quantities of radioactive liquids affect the characteristics and morphologies of membranes in particular (polymeric membranes). To circumvent this hurdle, the material produced by the membrane must be carefully selected when employed in high-level wastes [9]. Furthermore, ceramic or metallic membranes may be utilized to provide various benefits, including excellent radiation resistance, corrosion resistance, and high-temperature resistance. Despite this, there are significant concerns when employed in radioactive water treatment, such as its fragile construction, difficulty in manufacturing, and costly, laborious cleaning procedures for contaminated ceramic or inorganic membranes. As a consequence, these membranes have been replaced with polymeric membranes incorporating inorganic nonmaterial, which have features that combine organic and inorganic membranes. It also uses ceramic, mineral, or inorganic membranes restricted to contaminated water with high radioactivity. Still, modified polymeric membranes embedded with nonmaterial have obtained the same capability at a far lower cost than ceramic or inorganic membranes in recent decades.

Table 1: Summary of various radioactive waste processes [4][7][9]

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 Large choice of products ensuring high selectivity	Affected by high salt content Blockage problems Regeneration and recycling often difficult
Evaporation	DF>10^4 to 10^6 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 recycle 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 10^2–10^3, economical Established for large scale Operations	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

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

Fouling is another disadvantage of using pressure-driven membranes, as seen in Table 1. Membrane fouling is problematic for various reasons, including operating factors, feed characteristics, and membrane properties. Starting flux, feed water concentration, cross-flow velocity, temperature, charge, solution pH, and ionic composition are all important aspects in forming membrane fouling. Other factors contributing to fouling include membrane material, structure, and surface characteristics (hydrophilicity/hydrophobicity) [10]. Many efforts have been made by researchers to reduce this complex problem by improving the fabrication of membranes used in radioactive and non-radioactive wastewater treatment through numerous strategies that enhance and develop membranes, such as surface improvement by grafting and coating, radical polymerization, and blending with nanoparticles (NPs) as additives to the polymeric membranes. Also, these various strategies effectively reduce membrane fouling and remove dissolved radioactive ions from contaminated water. Furthermore, several membrane cleaning techniques were applied in accordance with established protocols to minimize fouling, which will be discussed in Section VI of this review. Orabi et al. [11] created a novel composite membrane composed of polysulfone (PS) and cellulose acetate (CA) grafted with chitosan (Ch-g-PDA). The innovative composite membranes were put through uranium adsorption tests. The results demonstrated the membrane's reusability in six adsorption-desorption cycles, with adsorption efficiency of 85.8 and 83.8 percent of the initial values for synthesized PSu/C-g-PDA and CA/C-g-PDA, respectively, indicating the stability and efficiency of the composite membranes in U removal from waste solution. Cross-linking was employed in another technique to remove cesium. Ding et al. [12] employed a simple and successful crosslinking technique to attach copper ferrocyanide/silica/polyvinylidene fluoride (CuFC/SiO2/PVDF) to the hollow-fiber composite membrane. The PVDF hollow-fiber membrane with embedded SiO2 was employed to stabilize the dispersion of CuFC nanoparticles for cesium (Cs) removal. The composite membrane had a high Cs rejection rate and membrane flux at the three layers of CuFC, and 0.5 percent SiO2, and its Cs rejection rate was unaffected by pH fluctuation (pH = 4-10). The CuFC/SiO2/PVDF hollow-fiber composite membrane demonstrated improved removal performance (more than 90%) in real surface water and simulated water with a low Cs content. Furthermore, nanoparticles incorporated in polymeric membranes may be employed. This method can achieve great efficiency in eliminating radioactive ions. Abbas et al. [13] created a unique NaY zeolite nanoparticle-modified Polyethersulfone (PES) membrane to remove ^{137}Cs from actual nuclear liquid waste. On seedless static aging, the zeolite was produced using the hydrothermal technique. The PES membrane matrix was then impregnated with various zeolite contents to adjust the membrane's features and ion exchange capabilities. The membrane produced with 0.15 percent NaY had the best removal rate (90.%), with a decontamination factor (DF) of 10.2 at pH 7.5. As a result, a second agent, copper ferrocyanides (CuFC), was added to the feed solution to improve 137Cs removal efficiency and decontamination factor. Consequently, Cesium retention was 99.2 %, and decontamination factors were 121.2. Table 2 describes different ways to modify membranes to extract radioactive ions from liquid waste.

Table 2: The strategies of modification of membranes separation

Membranes	Strategies	Radioactive ions	Removal efficiency	Ref.
Polysulfone (PS) and cellulose acetate (CA).	Incorporated with pre-synthesized chitosan grafted p-phenylenediamine (Ch-g-PDA).	Uranium (U)	85.8,and 83.8 %	[11]
Polyethersulfone (PES)	Blinding with Na-Y zeolite nanoparticles, and CuFC	Cesium (^{137}Cs)	99.2%	[13]
Poly-vinyl alcohol	Embedding potassium copper hexacyanoferrate (KCuHCF)	Cesium (^{137}Cs)	99%	[14]
Polyvinylidene fluoride (PVDF)	Blinding with SiO_2 and CuFC nanoparticles	Cesium (^{137}Cs)	99.4%	[15]
Polyvinylidene fluoride (PVDF)	Blinding with Prussian blue(PB)-SiO_2 nanoparticles	Cesium (^{137}Cs)	97.5%	[16]

So far, many radioactive waste removal technologies, such as membrane separation, have been devised and used. The number of radioactive ion removal papers from the Web of Science database during 21 years. From 2000 to 2012, around (51) publications and major papers were created each year; however, after 2012, research articles focusing on cesium removal considerably increased to approximately (194) due to the Fukushima Daiichi nuclear power plant disaster in 2011. In comparison to other pollutants, the number of publications on radioactive ions has increased due to the development of nanotechnology use with pressure-driven membranes (RO, NF, UF, and MF), as well as the increased interest in radio-ions among nuclear researchers due to the increased use of nuclear power plants around the world. The pressure-driven membrane separation approach for removing liquid radioactive waste will be discussed in this study (LRWs).

This research looked at pressure-driven membrane treatment techniques for liquid radioactive waste, with an emphasis on (NF, UF, and MF) due to their selective ability to remove radioisotopes. Furthermore, the operating conditions of these membrane types are better suited for radioactive wastewater treatment than RO membranes, which need more energy. Also, in terms of the use of pressure-driven polymeric membranes (NF, UF, and MF), they can be used as primary units before electrical membranes, the feasibility of which is to reduce unwanted parts and the effect of radiation. In contrast, electrical membranes are used in the processes of treating contaminated water in specialist applications, and besides, their cost is very high, and their applications are limited. It is also influenced by things like as (conductive and saline water). Consequently, utilize pressure-driven membranes as primary units to cleanse polluted water before it reaches the electrical membrane separation units and to maintain the main units operational for an extended period. In contrast, liquid membranes have limited uses, notably in the cleanup of radioactively polluted water. The first section of this review will focus on removing dissolved radioactive ions from contaminated water in the input solution, followed by membrane separation procedures (for example, NF, UF, and MF). The second section covers the use of current methods to embed nanoparticles in membranes, focusing on specific interactions between nanoparticles and polymeric membranes. Finally, the effects of membrane fouling on pressure-driven separation processes and the clean-up techniques employed to decrease fouling are discussed.

2. Treatment of Lrws Utilizing Pressure-Driven Membranes

In the past few decades, numerous membrane separation techniques have been established and used in potable water filtration and, more recently, in treating other processes and waste liquors. Certain membrane techniques may remove both dissolved and particulate contaminants. Pressure gradients are the most well-known and widely used techniques in industrial wastewater treatment. These methods include RO, nanofiltration, UF, and MF. It has been established that pressure-driven membrane separation techniques may be used effectively to remove radioactive compounds, with certain particular benefits, including more traditional procedures. After discovering appropriate membrane materials and their long-term verification in traditional water filtration areas, the nuclear industry has accepted these membrane techniques as a viable option for treating LRWs. In most situations, membrane procedures are employed as one or more treatment phases in complicated waste treatment systems that combine conventional and membrane treatment methods. These combination technologies have provided greater treatment capabilities, especially in circumstances when traditional procedures alone could not execute the work as efficiently or effectively. Pressure-driven membrane separation technologies may replace current radioactive waste processing procedures or supplement current radioactive waste treatment systems. In fact, most current radioactive waste processing systems use a hybrid of membrane separation and traditional processes. In certain systems, the membrane separation technique is the primary treatment step, but in others, it supplements traditional methods, boosting their efficacy.

Such combination systems can produce high-quality treated effluents with an acceptable quantity of residual radioactivity for release. Furthermore, the amounts of secondary radioactive waste residues are reduced and may be adequately conditioned to fulfill waste form standards for disposal [4,7]. This review highlights the use of pressure-driven membrane technologies to treat aqueous low and intermediate-level radioactive wastes. NF, UF, and MF are the membrane processes explored in depth.

2.1 Nanofiltration

NF is used in recovering boric acid and organic materials in nuclear power plants from contaminated cooled cycles in the nuclear reactor. In addition to its ability to recover, boric acid is also used to separate divalent radioactive ions dissolved in dirty water. For example, NF membranes were used to separate bivalent radioisotopes such as (cobalt-60 and strontium-90), a nuclear fission product in nuclear plants. The separation mechanism of these isotopes relied on controlling the surface charge of the nanofiltration membranes by changing the pH.

Gherasim et al. [17] studied the effect of pH change on the surface charge of a commercial NF membrane (AFC 40) in removing a cobalt isotope from contaminated water. The pore size characteristics of the NF membrane were determined by liquid-liquid displacement porosimetry and uncharged solute rejection. The results found a range of 0.54–0.56 nm. These values are significantly higher than the Stokes radius of the Co (II) ion (0.335 nm). Also, it found the pH dependence of the zeta potential (ζ) of the AFC 40 membrane measured in cobalt nitrate solutions of various concentrations in the range of 15–1000 mg Co/L. It can be observed that the AFC 40 membrane exhibits an amphoteric behavior in cobalt nitrate solutions with concentrations lower than 500 mg/L, with an isoelectric point (IEP) at pH = 4.1 for solutions containing 15–250 mg Co/L; for 500 mg Co/L solution the IEP is shifted to a higher pH value (about pH 4.6) and no IEP is observed when the solution contains 1000 mg Co/L. The amphoteric behavior is determined by the presence of both amine ($–NH_2$) and carboxyl (–COOH) groups in the polyamide top layer. The feed pH influences both the permeate flux and the ions retention by the AFC 40 membrane. The Data shows from this study that the permeate flux slightly increases, and the cobalt nitrate rejection decreases when increasing the feed pH in the range under investigation. These variations may be caused by several mechanisms, such as modification in the membrane's pore size, electro-viscous effect, and osmotic pressure gradient. Thus, when the membrane is charged, the pore size (pore volume) is reduced because of the repulsion between the charged groups of the membrane; the polymer adopts an extended configuration, which will produce an increase in rejection and a decrease in permeate flux. Additionally, the rejection of cobalt nitrate was very high (about 96% and 92%) for feed solution at pH 3–4 and significantly decreased to about 78% and 66% when increasing the feed pH to 5-6, respectively. Another study used it to separate radioactive uranium dioxide by commercial NF membranes. These oxides are generated by converting uranium hexafluoride gas into solid uranium dioxide dissolved in contaminated gaseous water. Oliveira et al.[18] tested two commercial NF membranes: NF and SW Dow/Filmtec. The transfer properties of the membranes were tested by (hydraulic permeability, flow permeability, and salt rejection) before and after being immersed in waste for 24 hours. Flow and permeability hydraulic system was used for forwarding permeation, cell displacement, and magnetic stir, feeding tank 800ml, with the maximum operating pressure of 6.2 bar. The system was compressed by dry air to carry the tank solution feeding the cell, and the solution permeated through the NF membrane. The effective membrane area is 40 cm^2. The data found in this test, between the permeable flow versus the pressure of the NF and SW membranes, shows that an increase in pressure leads to a linear increase in permeate flux. The SW membrane had the highest value, 7.8 L/m^2.h.bar, indicating the property of high water content, that is, the material's high affinity for water, considering that both membranes have pore sizes in the range from 1 to 10 nm. The rejection of NF and SW membranes was 64% and 55%, respectively. The application of NF membranes can be viable for treating and recovering uranium from waste "carbonated water", observed a decrease of the initial flow of 58% with the NF membrane and 31% with SW. This decrease can be due to concentration polarization, a phenomenon inherent in all PSM. When a solution permeates through a selective membrane about a solute, there is an increase in solute concentration in the interface membrane/solution and, therefore, an increase in osmotic pressure of the solution near the membrane, decreasing the driving force for the separation and therefore the permeate flux. NF membrane technology has been applied in several nuclear power plants. This type of filtration is used to remove boric acid and radioactive ions from water from the cooling cycles of nuclear reactors and remove uranium from drinking water as another application of this technology. In Bugey Nuclear Power Plant in France, NF was applied to separate ionized silica and boric acid. The presence of silica accumulated during plant operation resulted in the formation of silicates, causing fuel cladding corrosion [19]. NF allowed separation with 92 % recovery of silica and 16.5% of boron. The membranes retained the corrosion-activated cations (Sn^{2+}, Ag^+, and Co^{2+}). Australian Nuclear Sciences and Technology Organisation (ANSTO) tested different commercial NF membranes in cross-flow membrane cells to treat uranium mill effluents [20]. The rejection of uranium was greater than 75%. Some tested membranes showed potential for separating radium, sulfate, and manganese. Different NF membranes from Osmonic Desal (Desal 5 DK, Desal 5DL, and Desal %1 HL) and Dow (NF90 and NF45) were examined for uranium removal from drinking water [21]. The uranium complexes, which mainly occur in natural water, e.g., divalent anion complex $UO_2(CO3)_2^{2-}$ and four-valent anion complex $UO_2(CO3)_3^{4-}$ were rejected between 95% and 98% by four membranes and between 90% and 93% by one membrane, namely NF90.

2.2 Ultrafiltration

UF and MF membranes are used in nuclear power plants to separate colloids, macromolecules, and boric acid from nuclear reactor cooling cycles. Also, UF membranes are used to treat contaminated wastewater in nuclear research centers, hospitals, and the nuclear industry. The majority of these radioactive liquid waste generated from different activities in nuclear power plants are of complex composition, as they include many materials of different sizes and diameters, such as (organic and inorganic materials, colloids, large particles, and dissolved radioactive ions), which depend on the particle separation mechanism in membrane technology. UF or MF relies on the size of the membrane pores in relation to the size of the particles to be separated in these membranes. The ability of these membranes was limited to separating particles and materials of large diameters. At the same time, the dissolved radioactive ions can pass through the pores of the UF and MF membranes. For this reason, many nuclear power plants have used combined system methods of UF or MF membrane units followed by separations of dissolved radioactive ions by RO or NF membrane units. Membrane techniques are applicable in treating radioactive liquid wastes produced in sanitary centers, specifically those from in vitro tests containing radionuclides such as (I-125). Among the main membrane processes, the RO technique is the most suitable one to carry out that treatment, as it can retain almost all the I-125. To optimize the process, pre-treatment is necessary before the RO stage to remove the macromolecules and the largest compounds to avoid fouling and to increase the life of the RO membranes. Arnal et al.[22] employed UF membrane type (HG 05) as a pretreatment unit. The UF membrane unit was working at pressure (2-6) bar, while the RO membrane type (KNOWS-SS10) unit was at (20-40) bar. The best results are found when combining UF and RO techniques: UF is used to retain the macromolecules and the largest compounds, and RO is used in the second stage to retain the ion elements (I-125). The results obtained with RO membranes are shown over 97% in all cases so that RO membranes can retain nearly all the (I-125) present in the solution. Liquids from radioimmunoassay (RIA) are low-medium activity wastes and potentially infectious since they may contain pathogens from patients' blood. These wastes cannot be discharged due to chemical and biological, and radiation content. Sancho et al.[23] proposed the treatment of these wastes by membrane technology (UF and RO). The treatment's purposes are removing radioactivity and pathogens to make decontaminated liquid suitable for discharge; and volume reduction of final waste. Experiments with real RIA wastes of different activity levels were performed. Overall results obtained in the full treatment by UF and successive passes of RO produce final values of permeate activity suitable for discharge. Also, the overall DF values and average activity removal percentages have been much higher in all cases than those found in the literature for other treatments for (I-125) removal as shown in Table 3 .Furthermore, these values are similar, or even higher in some cases, to the ones found in other treatments of radioactive wastes by UF and RO.

Table 3: Overall results were achieved by UF and RO membranes

Characterizations	1UF + 2 RO	1UF + 2 RO	1UF + 3 RO
Initial concentration waste (kBq/L)	2.80	240	2.80
Treatment duration (h)	5	5	6
Volume initial waste (L)	63	60	60
Volume final permeate (L)	48	45	40
Final permeate concentration (kBq/L)	0.060	0.699	0.0652
Decontamination factor (DF)	46.7	343.35	3681
Volume Reduction Factor ($\boldsymbol{VRF}$)*	21	20	15
PH	6.6	6.7	7
Total storage time	1.5 years and 2 months	1 year and 1 month	~150 days

$$*VRF = \frac{Feed\ \ solution\ volume\ (Vf)}{Concentrate\ volume\ after\ treatment\ (Vc)}$$

According to the results in Table 3, the most suitable management for RIA wastes would consist of short storage of wastes after generation (around one month) and later UF treatment followed by 3 consecutive passes of RO. This management would mean a significantly shorter storage time than the other tested alternatives to preserve hospital storage capacity. After treatment, a permeate suitable for discharge (from the point of view of radioactivity) would be produced after 120 days of later storage. Before discharge, the lack of pathogens should be checked. Furthermore, managing the waste volume can be reduced by around 30%. The UF and RO membranes experimental tests were conducted with real RIA wastes. Sancho et al. [24] utilized the UF membrane in pre-treatment RIA radioactive liquid wastes, the 4040-TFV-P100 of Hydranautics. It is a spiral wound polysulfone membrane, the most resistant polymeric material to radiation, with a molecular cut-off of 100000 Da and an effective membrane area of 6.5 m2. The permeate obtained in the UF stage was later treated with the RO membrane CPA2–2540 of Hydranautics, a composite polyamide spiral wound membrane, with an effective area of approximately 2.6 m^2. The characteristic parameters of the real wastes used were included in this experiment: conductivity (11–15.5) mS/cm, pH (6.5–7), and concentration activity (101000) KBq/L. The experiments carried out with real wastes proved that UF has an important role in the partial reduction of feed radioactivity. It can be seen that radioisotope rejection by UF membrane was higher than 50% most of the time, even reaching a maximum value of 80%. This proves that part of the (I-125) remains labeled to the proteins and other organic compounds, most of which are removed in the UF stage. Perm-selective performance of RO membrane has been excellent from radioisotope removal, with values higher than 90% in the first RO pass and higher than 95% in the following passes. Experimental results prove that UF and RO combination is suitable for treating radioactive liquid wastes from RIA. Many nuclear power plants employ UF membranes as an important part of radioactive water separation processes. These applications have been collected and published in the International Atomic Energy Agency publications. These publications are based on the UF membrane separation capabilities and the purpose of establishing these units to reduce the risks of these wastes. The Salem nuclear power plant used UF to treat low-level radioactive wastes originating from floor drains from pressurized water reactors (PWR), laboratories, sampling points, and auxiliary equipment drains. The purification system consisted of tubular UF modules and a demineralization unit. UF membranes protected the ion exchange beds removing particles smaller than 0.05 µm, oil, grease, colloids, and metal complexes. The number of such radioisotopes, like $^{58/60}$Co, ^{54}Mg, and ^{100}Ag, was also reduced by UF modules [25]. The UF was tested at Seabrook nuclear power plant with the aim of colloidal ^{58}Co removal to discharge limits and reduce total suspended solids. The processed waste contained floor drains from PW and spent resins tank drain-down. The UF plant took the sidestream from the wastewater feed line and operated in parallel with other processing systems. More than 90% of TSS was removed by a UF system[26]. Waste originating from floor drains, equipment drain tanks, and reactor coolant systems was processed with UF in Callaway nuclear power plant. Four UF modules were used to treat reactor coolant water. 70% of radioactivity and suspended solids were removed with UF, and permeate was additionally polished with ion exchange to the minimum detectable level. Only one UF module was used for the treatment of floor drain water; after removing 89% of radioactivity and suspended matter, the permeate was discharged to the floor drain system for recycling. The full-scale plant configured in 2001 consisted of UF unit and ion exchange system processing the waste for direct disposal. The UF protected the ion exchange beds against the foulants that could shorten the life of the resigns[27]. In Diablo Canyon nuclear power plant, two UF systems for processing spent media transfer liquid – sluice water containing a high concentration of radioactive submicron particles – were tested. This kind of waste was considered the most difficult waste for the previous system to process. The new treatment unit was a mobile tubular UF plant. The product from this plant was finally cleaned in ion exchange columns, and the reject needed additional processing. The second UF skid-mounted plant used a solid collection system, resulting in no additional rejection processing. Both systems produce liquid free from particulate activity that can be introduced into the ion exchange columns without the danger of serious fouling and deterioration of their performance [28]. At Mound Laboratory (USA), UF was tested to treat wastes from a fuel reprocessing plant. The waste feed contained 5–100 mg/L suspended solids and less than 16,000 Bq/L specific radioactivity; the main radionuclide was ^{238}Pu. The installation comprised 32.3 m long tubular UF elements, 6.5 m^2 of total surface area, and 4.5 m^3/h of productivity. 80–99% of α-emitters were removed in the test operation and high retention of transuranic elements in full-scale installation was observed (^{241}Am = 98.9%, ^{238}Pu =98.6%, ^{237}Np = 69.1%, and ^{233}U= 93.7%) [29].

2.3 Microfiltration

In nuclear power plants, MF is used for pre-treatment purposes or the concentration of coarse particles after precipitation. MF employed with high-level radioactive waste is ceramic membranes; these membranes are used to rid radio particles with high decontamination and concentration factors. The MF membrane's ability in nuclear power plants to treat liquid radioactive waste was tested in many nuclear facilities. At AECL Chalk River Laboratory (Canada), the possibility of application of MF for groundwater and soil decontamination [30][31] was demonstrated. The hollow fiber MF system consisted of 40 cross-flow filtration modules, ~6 cm in diameter, 50 cm long, and 1 m^2 surface area. The membranes were manufactured from polypropylene, with a nominal pore size of 0.2 μm. Feed was introduced tangentially, and cross-flow velocity was kept on the level avoiding deposition and fouling. Membranes were periodically back-washed by pressurized gas from the filtrate side. The air pulse 90s in duration removed all accumulated solids from the membrane surface. Additionally, periodic chemical cleaning was applied with an alkaline detergent. The system demonstrated the usability of MF for the treatment of soil leachate and the removal of radionuclides from groundwater. 120 m^3 of groundwater contaminated with 90Sr were cleaned during pilot plant tests. The radioactivity of ^{90}Sr was reduced from 1700–3900 Bq/dm^3 to 2 Bq/dm^3. From that time, about 20,000 m^3 of groundwater was treated. MF was applied to clean the groundwater from uranium isotopes, toxic organic compounds, and heavy metals at Rocky Flats (United States). The installation was equipped with tubular MF modules with a pore size of 0.1 μm. The system's removal efficiency of uranium isotopes was 99.9%[7]. The Idaho Nuclear Technology and Engineering Center (INTEC) belong to the Idaho National Engineering and Environmental Laboratory, and the cross-flow MF for the remediation of radioactive waste was developed [32]. Solids formed from precipitation and radioactive substances' absorption during reprocessing nuclear fuel required separation from the liquid before solvent extraction and ion exchange. The cross-flow filtration with Mott sintered Hastelloy filter for that purpose was tested. The filter was characterized by 0.480 in inner diameter, 6 in length, and 0.5 μm pore diameter. Its performance was tested for different solids loading from 0.19 to 7.94 wt.%. Filtrate flux rates for each solid loading displayed a high dependence on transmembrane pressure and negative dependency on axial velocity. Cross-flow filtration seemed to be a viable method for the removal of undissolved solids from INEEL radioactive slurries. Cross-flow MF for radioactive waste dewatering was evaluated in Berkeley Nuclear Laboratories[33]. Pilot tests of five diverse types of units were established as cross-flow MF — the most appropriate for the thickening of mineral waste slurries. The ceramic and stainless-steel membranes were used in tabular form: Pall PSS (2.5 μm limit of separation), Fairey Microfiltrex FM4 (1 μm), and APV Ceraver (1.4 μm). Cross-flow MF was effective in dewatering a range of simulated radioactive wastes. MF membranes were applied in centrifugal membrane filtration technology developed by SpinTek membrane Systems, Inc. The technology supported microporous membranes rotating at high speed under pressure to separate suspended and colloidal solids from liquid streams yielding solid-free permeate and concentrated retentate. Energy and Environmental Research Center tested different ceramic membranes in the SpinTek STC-X4 static cell. Based on those experiments, a 25-μm TiO_2/Al_2O_3 membrane was selected for evaluation of the performance of centrifugal membrane filtration using SpinTek ST IIL [34]. It was assessed that the technology can be applied to tank wastes, contaminated groundwater, landfill leachate, and secondary wastes from other remediation processes, including decontamination and decommissioning systems within the U.S. Department of Energy (DOE). The innovative membrane system coupled with centrifugal force gave this crosscutting technology the advantage over conventional UF or MF processes. By application of this system the streams containing relatively large particulate sizes, unlike in hollow-fiber filters, can be processed. It is also possible to maintain higher permeate fluxes and higher solid concentrations than in tubular or spiral-wound units. The combination of the ceramic membrane with the low-fouling characteristics for the centrifugal system resulted in SpinTek's ability to treat hazardous wastewater to a slurry-type level and reduce tank sludge volume. Spin ST-IIL was tested at Los Alamos Nuclear Laboratory (LANL) with the aim of a full-scale demonstration of the technology. The designed two-stage system operated on both surrogate and real radioactive wastes from LANL was expected to provide the results for the elaboration of the model for determining the applicability and economics of the SpinTek ST-II system to different DOE waste and process streams.

3. Feed Solution Treatment Followed By Membrane Separation

Processes of treating liquid radioactive waste in nuclear power plants and its applications are not limited to combining pressure-driven membrane units only. Researchers have sought alternative ways to remove dissolved radionuclides through the chemical treatment of the feed solution followed by membrane separation processes. One of the most important reasons for using these methods is to increase the selectivity of membranes, especially UF and MF membranes. In addition, chemical treatment methods followed by membrane separation processes saved energy consumption compared to the combined separation processes between UF membranes or MF with RO membranes. Thus the costs of setting up treatment plants are lower than the methods discussed in Section 2 of This review. Chemical treatment methods followed by the membranes also have limitations, including the quantities and types of chemicals used in the treatment of the contents of the feed solution, the proportions of addition to the feed solution must be taken into account so as not to cause an increase in secondary residues or damage to the membranes used after processing operations with additives, ease the recovery of these additives by filtration processes, and that they are cheap and environmentally friendly. Therefore, when using these materials, these limitations must be considered before starting to use them in radioactive water treatment. We address the materials used in treating the feed solution first, followed by a membrane separation (NF, UF, and MF). This method used several mechanisms, including precipitation, co-precipitation, complexation, and adsorption, to obtain selectivity for radioactive ions and increase the membranes' separation efficiency referred to in this section [35]. The reason for using this method is to get rid of dissolved radioactive ions that have small ionic radii compared to the big pore size for these types of membranes via increasing the ionic radii for these radioactive ions, which impede its penetration through the membrane pores. Additionally, these additives can convert dissolved radioactive ions into insoluble radioactive materials (solid-state) by materials that have macromolecular weights such as (polymers, oxalates, and phosphates) through precipitation, co-precipitation, and complexation mechanisms. As for the adsorption mechanism, materials additives that absorb radioactive ions are used, as it depends on the ion exchange mechanism between the ions of the additive material and the radioactive ions, for example, zeolites, active charcoal, and copper ferrocyanides.

3.1 Nanofiltration

Some nuclear effluents contain traces of radioactive elements in sodium salt media, from which radioactive cesium must be separated. Various processes (including liquid/liquid extraction and ion exchange) can perform such separation, but they produce additional waste. Therefore, NF has been selected as a new separation process that may allow a large volume reduction without generating additional waste. NF is a pressure-driven membrane process between UF and RO that can separate species at the ionic scale to separate dissolved radioactive ions from the highly salted aqueous medium. Some laboratories have been developing the NF-complexation technique since 1993 [35,36,37]. These laboratories have shown that using EDTA with a Nanomax 50 membrane allowed 100% retention (rejection) of Sr^{2+} and other polyvalent ions and only 10% retention of Na and Cs ions. Then, using a cesium-selective ligand such as resorcinarene, Cs/Na separation was also improved - but not satisfactorily. In fact, to remove radioactive traces of cesium in a highly salted aqueous medium, a minimum of 99% Cs retention along with a maximum of 10% Na retention would be required for industrial applications. Hwang et al. [38] used three types of NF (NTR7410, NTR7250, and NTR729HF) membranes with precipitation and complexation by polymer (poly acrylic acid(PAA)) to eliminate strontium (^{90}Sr) from liquid waste. PAA with a molecular weight of 5,000 was added to the feed solution as a complexing agent, and further strontium removal (>50% removal) was achieved at higher pH as the molar ratio of [COOH] to [Sr] increased from 0 to 27. However, NF fluxes were affected by the addition of PAA, particularly when a rapid change of feed pH from 9 to 5 was done. This could be explained by the reasoning that the sorption of associated PAA onto the membrane surface was more facilitated by forming larger amounts of less charged PAA with a sudden pH drop. In another study, Macnaughton et al. [20] separated milled uranium from radioactively contaminated water using various commercial NF membranes. In addition, they studied the membrane properties (pore size and surface charge) and the effect of changing the pH on the membrane surface charge in rejecting radioactive ions. Pointed out that most NF membranes have negatively charged, allowing positive radioactive ions to pass through its pores. Changing the pH of the solution before the separation process begins is crucial in getting rid of the positively charged radioactive ions. Also, this study clarified that pore size is a very influential factor in rejecting radioactive ions, so uranyl sulfate was used as an additive to convert dissolved uranium ions into large complexes that NF can separate. The results showed a rejection rate of 75% for milled uranium by complexation- NF. More than one type of pressure-driven membrane separation process can also be combined with complexity. In a study conducted by Zakrzewska [39] combined NF and UF with complexity to separate dissolved radioactive ions from polluted water collected from the Institute of Atomic Energy in Warsaw. NF and UF ceramic membranes were used with a group of poly (acrylic) acid complexes and poly(acrylic) acid salts of different crosslinking, polyethyleneimine, and cyanoferrates of transient metals. The experiments included changing the ratios of adding complexes to the feed solution and studying the pH and its effect on the decontamination factor. Results showed that the best conditions for UF–complexation resulted in high decontamination factors found in multistage arrangements, especially when radioactive wastes contain one dominant component, as was cesium (137 Cs) in experiments. DF was (179.5) with CoCF/NaPAA complex agent. By operating a pressurized water reactor (PWR) in Hungary, low-level liquid radioactive waste was generated with the isotope cobalt-60, so a research team [40] removed this isotope from the contaminated water by using three types of NF membranes (MWCO: 200, 400, 1000) with the addition of ethylene diamine tetraacetic acid (EDTA) Complexing material for Cobalt-60 isotope selectivity from contaminated water. Experiments with the rejection of the NF membrane MPF-34 (F 200 MWCO) showed a good separation ability up to (R>90%) at pH (9.5) and at a temperature of 25 ^{o}C. The filtration system was designed and implemented to separate the americium (^{241}Am) isotope from erbium (^{152}Eu) by using a NF membrane made from polyethylene glycol (PEG), The active membrane area is 0.25 m^2, and molecular weight cut-off (MWCO) is estimated at 2500 Da with a complex di ethylene tri aminepentaacetic acid (DTPA) added to the feed solution as shown in Figure 2. From Figure2the complexation- NF filtration system for the separation of radioactive isotopes; through this system the effect of pH and the amount of additive to the feed solution was studied, followed by separation by NF. The observed 241Am and 152Eu rejections increase with increasing pH during the filtration system. Furthermore, at low pH (<2.5) the rejection of (^{241}Am) is higher than the rejection of ^{152}Eu even with a 3700-fold excess of ^{152}Eu. At pH 1.7, the rejection of ^{241}Am and ^{152}Eu is 41% and 33%, respectively.

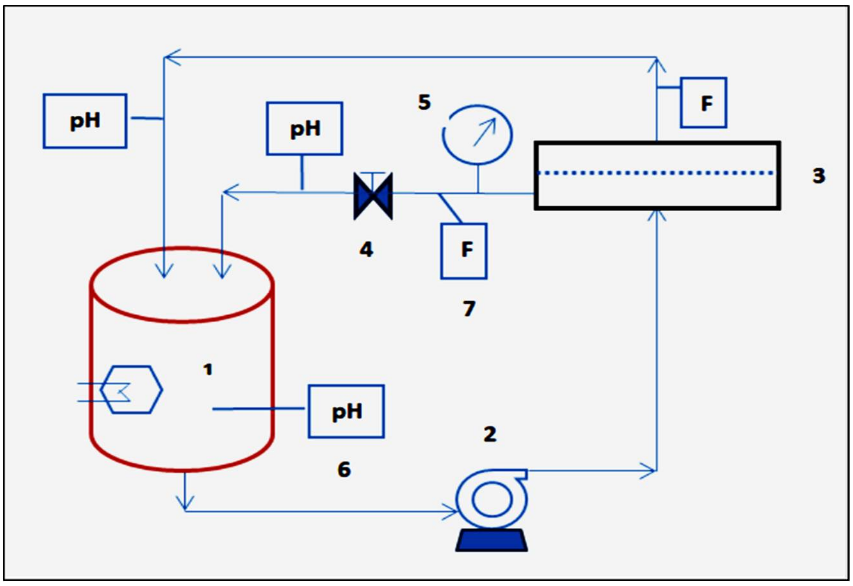

Figure 2: Am/Eu/DTPA filtration system (1: feed tank, 2: pump, 3:cell with NF membrane,4:control valve, 5: pressure gage,6: pH controller,7: flow meter)

At low pH, the proton competition limits the metal ions' complexation by DTPA, and the difference observed can be explained by the chemical speciation in the solution. The membrane selectivity is thus determined by the difference in the stability constants of the 1:1 complexes of ^{241}Am and ^{152}Eu with DTPA. At pH greater than 2.5, the proton competition is limited, and both ^{241}Am and ^{152}Eu are complex. Under this condition, the rejection of both the metal ions reaches a maximum, i.e., 98%, and no difference between ^{241}Am and ^{152}Eu rejection was noticed. Also, a separation factor (SF) superior to 3.5 for (Am/Eu) was obtained at pH 1.7 using DTPA [41]. Other studies employed NF membranes with chemical treatment of feed solution to rid the radioactive ions from contaminated water, summarized in Table 4.

Table 4: Chemical treatment of feed solution followed by NF membranes

Membrane	Additives	Target elements	Rejection (R %)	Ref.
FILMTEC NF 70 (2514) spiral wound membrane (MWCO = 300 $kg.kmol^{-1}$)	Resorcinarene 1	^{137}Cs	85%	[42]
NF membrane (NF45) poly piperazine amide (pore size: 0.48 nm)	Boric acid (BA)	^{60}Co	93%	[43]
NF and RO membranes	$FeSO_4$, $Fe_2(SO_4)_3$, and $Al_2(SO_4)_3$, $FeCl_3$, and alum	UO_2^{2+}, Cu^{2+} and Pb^{2+}	>95%	[44]
NF membrane (selective layer of polyamide on polyethersulfone support)	---	UO_2^{2+}	97%	[45]
NF-membranes (NF270,NF 90,and XN 45), (pore size: 0.84nm, 0.68nm)	Polyacrylic acid or ethylenediamine tetraacetic acid disodium salt	^{90}Sr	99%	[46]

3.2 Ultrafiltration (UF)

The concept of UF membranes used for radioactive effluents in many nuclear facilities was only for the extraction of large particles after extensive studies by researchers who used this type of membrane separation with (precipitation, co-precipitation, complexation, and sorption) reached tremendous results that were able to separate isotopes radioactive dissolved in contaminated water. To enhance the UF membrane for the treatment of dissolved radioactive ions, the Harwell Research Center in the United Kingdom (UK) was able to use UF membranes to separate dissolved radioactive ions from liquid radioactive wastes (LRWs) with (precipitation, complex, and adsorption). This hybrid method was called seeded UF (SUF) [47].In another case, SUF was applied to remove radioactive isotopes at the Institute of Nuclear Chemistry and Technology, Poland. A ceramic UF membrane CéRAM INSIDE® (23-8-1178) 23-channel ceramic module (8 kD cut-off) was used with complexation agents including polyethyleneimine (PEI), polyacrylic acid, sodium polyacrylates (NaPAA), and cobalt cyanoferrates (CoFC). The operation conditions of this hybrid process were conducted in a cross-flow mode, with the pressure in the system being 0.25−0.5 MPa. In that pressure range, the permeate flux of water varies from 26 to 85 L/m^{2}h. The ratio of the polymer concentration to the concentration of the metal ion was selected in the experiments. The ratio changed in the range of 1:1−20:1. A complexing agent is added to the feed solution before filtration. The solution is mixed for 1−2 h and then seasoned for several hours to establish equilibrium (stable activity of permeate). The pH is fixed at the required level at the seasoning time selected in the laboratory experiments. Results showed the ability to remove cesium (137Cs) ions by CoFC followed by UF membrane, the decontamination factor (DF) of this hybrid process reached more than one. Also, the retention of ^{152}Eu was complete, and ^{241}Am was removed after the second portion by NaPAA [48]. Various methods for concentration of radioactive waste have been studied and developed at the Institute of Nuclear Chemistry and Technology (INCT), including MF, chemical precipitation, UF, enhanced UF (SUF), NF and RO were tested at INCT with institutional radioactive wastes collected from the Radioactive Waste Management Plant. The system for treating liquid low and medium-level radioactive waste from nuclear applications (institutional waste) was composed of four stages, like in Figure 3[49].

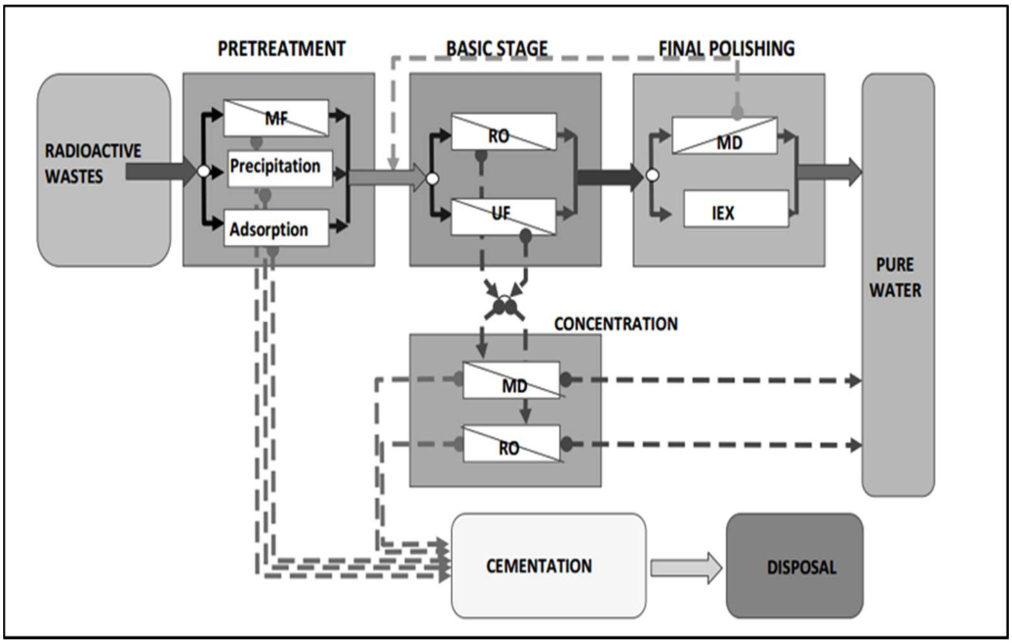

Figure 3: The scheme of the filtration system for low-intermediate level waste treatment [49]

From Figure 3**,** radioactive aqueous waste enters the pretreatment stage, which prepared before the basic proprocessing stage In the pretreatment stage, all suspended matter is removed by depth filters. The organic compounds and oxidizing agents like chlorine are retained by sintered or granulated activated carbon filters. The precipitation process removes The precipitation process removes a large proportion of the radioactivity load roxide or ferrocyanides. Depending on pretreated waste can be cleaned with UF or RO membranes. UF is generally applied for colloidal solutions; however, real waste containing small ions like $^{60}Co^{2+}$ or $^{137}Cs^{+}$ can be treated in the UF process enhanced by soluble polymers or other complexing agents or dispersed sorbents like activated carbon (SUF). After the basic processing stage, the final polishing of permeate in the MD module or standard ion exchange columns occurs. The effluent from this stage is relatively pure water (below discharge limits: 10Bq /L) that can be discharged to the sewage system or used as process water, e.g., for cleaning the membrane modules or other plant components. For more information on using UF membranes to remove radioactive isotopes from liquid radioactive wastes. Table 5 lists the materials used in the treatment of the feed solution followed by UF with the results obtained by researchers in this field.

3.3 Microfiltration (MF)

MF has been widely utilized to treat potable water and residential sewage in the industrial sector to remove particle matter. MF filters particles in wastewater produced by Liquid Radioactive Wastes (LWR) power plants, typically with a concentration factor (CF) of 100. If the precipitated particles are coarse enough, the technique can be utilized in conjunction with precipitation. Depending on the feed stream's parameters, organic and inorganic MF membranes can be utilized. Because of the material's radiation resilience, Ceramic MF has been utilized to remediate highly active wastewater in the nuclear sector [54]. MF membrane pores are too large to remove radioactive ions from water. In nuclear technology, the MF process is used after precipitation, either for pre-treatment purposes or the concentration of the precipitate. The seeded-precipitation method was created using traditional precipitation methods for calcium and barium removal. Because strontium, calcium, and barium have comparable physical and chemical characteristics, their water treatment procedures should be similar, including the seeded-precipitation process, as shown in Figure 4. The impact of precipitation and solid-liquid separation is significantly enhanced in seeded precipitation [56]. In recent years, the seeded-precipitation method has been successfully employed to remove strontium from simulated radioactive wastewater. Flouret et al. [57] used barium sulfate seeds to investigate strontium co-precipitation. At 25 degrees Celsius, the decontamination factor (DF) reached 275. They discovered that a filtering step was required following the co-precipitation procedure to optimize the DF. Luo et al. [58] investigated the strontium removal procedure using co-precipitation and MF technologies. For treating strontium-containing wastewater, they looked at a pellet co-precipitation MF technique. DF was 577, and the concentration factor (CF) was 1958 when the operating temperature was 22 ± 0.8 C. The co-precipitation followed by the MF (CPMF) process is a combination of co-precipitation and MF, combining both advantages. Precipitation alone cannot efficiently separate the solids from the liquid, resulting in a poor decontamination (DF) value. On the other hand, the flux of a membrane rapidly decreases without a precipitation step. The strontium concentration in the raw water used in this study was about 5 mg/L, corresponding to low-level wastes. In two intermittent tests, two dosages of sodium carbonate, 2000 mg/L, and 1000 mg/L, were made to precipitate strontium carbonate from the raw water, which was then subjected to MF. DFs of 237 and 158 and the mean concentration factors (CFs) of 288 and 462 were obtained [59]. More information about these studies and the latest publication on MF applications is included. Table 6 summarizes some examples of hybrid processing to treat liquid radioactive wastes.

Table 5: UF membrane separation processes with the additives in the feed solution

Membrane specifications	Additives	Elements	Feed solution	Result	Ref.
UF (10,000 MWCO)	Metal-binding polymers	zinc, nickel plutonium, americium	Actinide Removal from Los Alamos National Radioactive (LANL) Radioactive Liquid Waste Treatment Facility(TA-50)	After several stages of treatment, the best result was recorded < 100 pCi/L at pH (6.5) for wastewater.	[50]
UF acetate cellulose membrane (20 000 MWCO) by LIKO Bratislava)	Sodium dodecyl sulphate (SDS) and di-2-ethylhexaphosphoric acid(D2EHPA)	^{85} Sr	A solution containing Strontium-85 was obtained by DuPont (USA).	The strontium rejection (R %) is approximately complete under the filtration processes	[51]
Ultra filtration (UF) using a membrane having a pore size of 0.2 µm	Copper ferrocyanides (CuFC) and calcium phosphate.	Cesium and strontium	Low-level wastes (LLW) and intermediate-level wastes (ILW) are generated in various facets of the nuclear fuel cycle and have various chemical composition	The DF for cesium was (200 to 300) binding with CuFC at pH 9, while strontium was (200) with calcium phosphate at pH 11.	[52]
UF unit with polyethersulfone (PES) membranes with MWCO (3, 5, and 10 Kd).	**Sodium dodecyl sulfate (SDS) and non-ionic polyethylene glycol ether (Tergitol 15-S-9) surfactant micelles**	**^{241}Am**	**Simulated solution (HLW)contacted 144Ce,137Cs,95Zr,90 Sr ,95Nb and 241Am**	**Americium (III) has been completely removed using SDS micellar solution at and above pH 3.**	**[53]**
UF membranes (Millipore, USA) were made of regenerated cellulose and had molecular weight cut-offs of 1, 10, and 100 kDa	polyacrylic acid (PAA) and polyethyleneimine (PEI)	Co and Sr	Simulated low-level radioactive waste	Removed >80% of Co ions combined with PEI and less than 40 for Sr.	[53]
UF (MEMBRALOX) with pore size (1-100 nm) and (CeRAM INSIDE) with pore size (15 nm).	Cobalt ferrocyanides (CoFC),polyethylene imine (PEI), polyacrylic acid(PAA)	^{51}Cr,^{124}Sb, ^{125}Sb, ^{60}Co,^{141}Ce,^{137}Cs,^{154}Eu, 152 Eu and^{241}Am,	Real waste with various radioactive elements.	The best result of DF was for 137Cs, 154 Eu, and 241 Am binding with UF/ CoFC at 109.8,3.78 and 4.47, respectively.	[54]

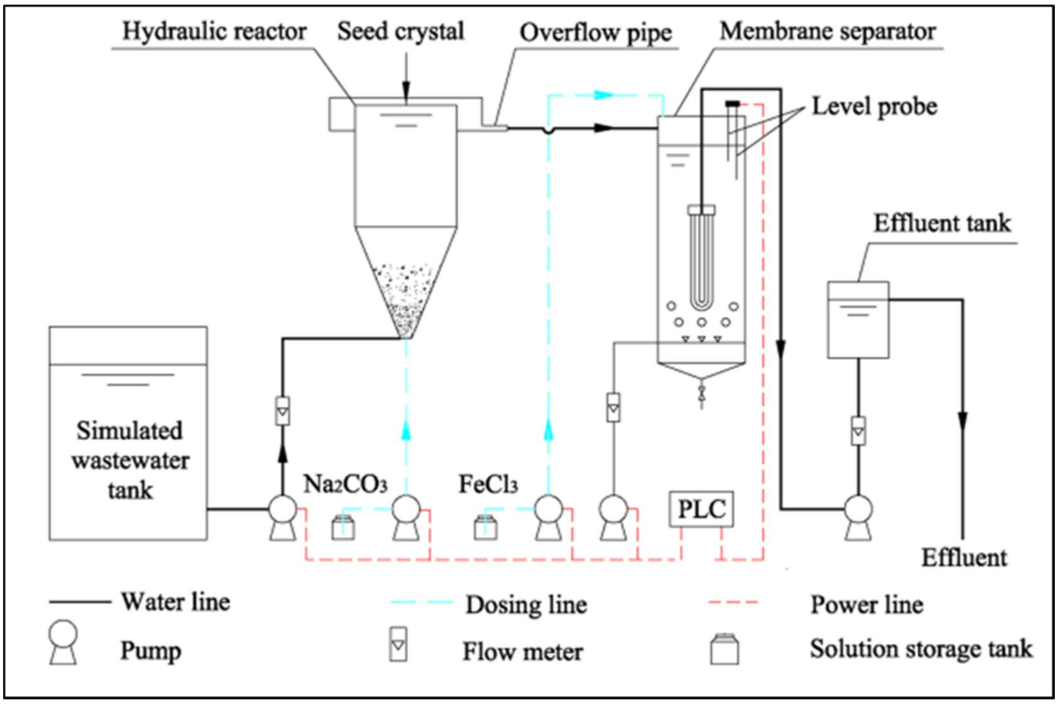

Figure 4: schematic of hydraulic pellet MF (HPC-MF) process for strontium removal [55]

Table 6: The application of MF membrane in wastewater remediation

Hybrid process	Target isotopes	Result	Ref.
Adsorption–microfiltration with polyvinylidene fluoride (PVDF)	^{137}Cs	DF = 208 Strong membrane fouling	[60]
Microfiltration with PVDF hollow fibers and Fe(OH)$_3$ flocculation	^{241}Am, ^{90}Sr	99.9% ^{241}Am rejection, DF =1650	[61]
Microfiltration with sodium carbonate, pore size 0.22 µm	Sr^{2+}	DF = 460	[62]

In the foregoing mentioned in this section, it can be concluded that the great possibilities offered by this method, such as (NF-complexation, seeded UF SUF, precipitation-MF, etc.) in obtaining decontamination and concentration factors with very high values that enable the disposal of radioactive ions, in addition to that reducing the volume of radioactive waste, which is the most important factor in the management of radioactive waste. Despite this, some important factors remain in achieving the best results for the decontamination factor when adding ligands to the feed solution (pH, concentration of radionuclides). Figure 5 depicts the effect of the decontamination factor on the pH of the different ligands added to the feed solution in separating radioactive isotopes.

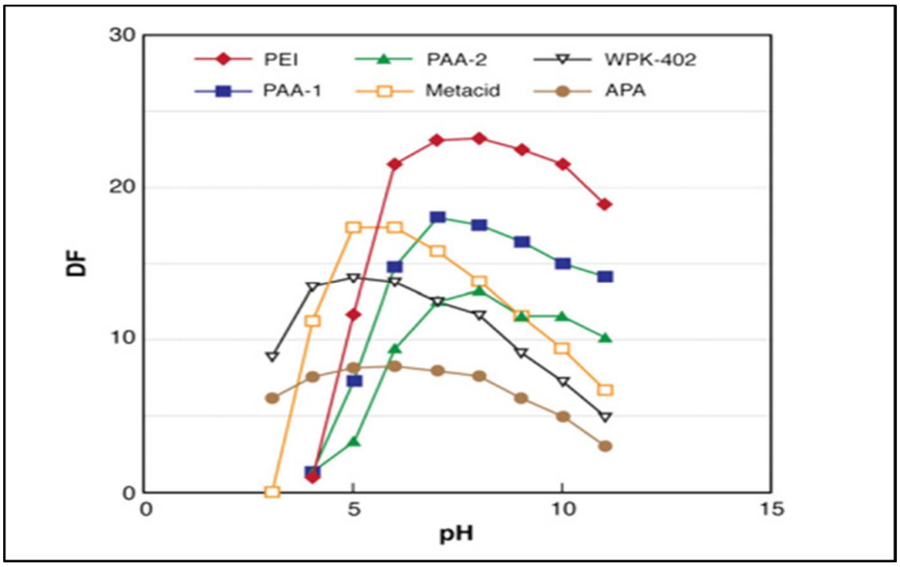

Figure 5: Variation of the DF with pH for different ligands. Reproduced from [63]

From Figure 5, it can be seen how the pH affects the decontamination factor. The best results were within the range (5-8), representing the best binding between the dissolved radioactive ions and the added ligands. As for the low pH ratios, it can be observed that the decontamination factor values decline in acidity media, and the reason is due to the ability of the hydronium ions to bind with the added ligands, which leads to the displacement of the radioactive ions in the feed solution, in addition to that the influence of these ions with a pH of less than 5 leads to the dismantling of the bind for the legend. When the pH is higher than 8, we also notice a decrease in the decontamination factor in alkali media due to these media destroying the structures of legend.

4. Nanomaterials Incorporated Polymeric Membranes

Recently, nanomaterials have been a very popular method for removing heavy metal and radioactive ions from wastewater due to the ability of these materials to bind with ions through the unique stable nanostructure and functional groups that these materials possess. In addition, these materials possess a large surface area and exceptional physicochemical properties. At the nano-scale, these materials show great adsorption capacity and strong chemical affinity with radioactive ions dissolved in contaminated wastewater when compared with larger-scale materials due to high specific surface area exhibit a stronger tendency of various interfacial reactions, which is essential for the high-efficiency selective adsorption of soluble radionuclides in contaminated water. More importantly, the adsorption sites at the surfaces of nanomaterials can be easily functionalized by creating different functional groups for the targeted removal of radionuclides [48,49]. From recent literature, most of the inorganic nanomaterials used to improve polymeric membranes have been metals and metal oxides, for example, titanium dioxide, zinc oxide, silica dioxide, alumina, iron, and zirconia, with some exceptions in some other materials such as NaA-zeolite, NaY-zeolite, magnesium hydroxide, calcium carbonate, hydrated manganese dioxide, multi-wall carbon nanotube, carbon nanotubes, and graphene oxide [13,64,65,66,67,68,69]. The use of nanoparticles in the manufacturing process of polymeric membranes has received much attention during the last decades, particularly as a new step in flux enhancement and fouling reduction. Hybrid membranes comprising inorganic fillers in a polymer matrix are well known. Common fillers are oxides such as SiO_2 and zeolites [64]. The filler concentration can be very high without any loss of the physical properties of a polymeric membrane. The membrane is denoted as a ‘mixed matrix’ membrane, in which both phases are present and have a positive mutual influence [65,66,67]. The filler can be used for separation improvement when, e.g., zeolites [68]; metal oxides have been mainly used for flux enhancement. In the case of mixed matrix membranes, however, the inorganic material is generally used as a bulk material and not as an additive. Using similar materials, but with sizes on the nanoscale is newer. The reasoning behind this approach can be mainly related to the interaction between the two phases: by using well-distributed smaller particles, an enhanced interaction is expected so that the nanoparticle sites are used more effectively for flux enhancement or fouling mitigation.

4.1 Individual Nanoparticles Incorporating with Membranes

In terms of the practical application of metals, metal oxides such as (TiO_2, SiO_2, ZnO, Al_2O_3, and ZrO_2), and other types of powder nanoparticles, for instance, (zeolite, graphene oxide, Prussian blue, carbon nanotubes, and metallic ferrocyanides), the particle size of these materials nanoparticles is the most important part to be taken care of in terms of the availability of sufficient information when combined with the membranes. The powder often consists of particles of different sizes in nanoparticle synthesis processes; Therefore, the average particle size and size distribution must be calculated when included with the membranes. The reason is due to the ability of these particles to agglomerate and form large groups, which causes defects in the membrane surfaces or blockage of their pores. In recent years, tremendous progress has been made in particle size analysis, particularly in analytical approaches that allow rapid responses, high levels of reproducibility, and the inclusion of many different particle sizes, particularly with diffraction and laser scattering, such as dynamic light scattering (DLS). Recent uses included incorporating nanoparticles (NPs) into polymeric membranes to improve their hydrophilicity to reduce fouling, cost-effectiveness, easier and well-developed methods of fabrication, reinforcing the membranes to increase their resistance to chemical corrosion, temperatures, and radiation tolerance, antibacterial and increasing the selectivity and permeability of membranes to rid dissolved radioactive ions and heavy metals from contaminated wastewater. The fundamental mechanism of these NPs compounds, such as inorganic materials and metal oxide, is adsorption. It is characterized, however, by its capacity to exchange ions in its peculiar structure with radionuclides dissolved and heavy metals in contaminated water. Furthermore, their utilization gives polymeric membranes additional advantages. Consequently, these materials may be employed as single nanomaterials or modified nano-composite materials incorporated with polymeric membranes. For example, when silica nanoparticles are incorporated into polyvinylidene fluoride membranes, the membrane has better temperature resistance as well as higher levels of selectivity and diffusivity [69]. Zinc oxide nanoparticles in a chitosan membrane showed strong mechanical properties and a high antibacterial activity [70]. When silica nanoparticles are incorporated into polysulfone membranes, it increased gas permeability results [71]. Polyethersulfone membranes incorporating aluminum oxide nanoparticles demonstrated increased porosity, a low flux decline, and pseudo-steady-state permeability [72]. Silica nanoparticles incorporated into polybenzimidazole membranes resulted in increased selectivity during gas separation and enhanced permeability [73]. Adding particles to a polymer matrix has also been shown to stabilize the polymeric membrane regarding variations in perm-selectivity when temperatures vary [74]. Metal oxide nanoparticles may have several disadvantages. For example, reduced permeability of PSF membrane due to high TiO_2 content (more than 2%) [75]. Furthermore, an excess of metal oxide nanoparticles increases pore size, negatively influencing membrane performance due to an imbalance between permeability and selectivity and the environmental risks associated with nanomaterials loadings. Table 7 illustrates how particles in the liquid phase interact with one another on the surface [76]. Surface contact happens when nanoparticles in a polymeric solution meet the parameters listed. While scientists understand surface contact theory, it is still unclear what elements might promote or induce agglomeration. When making a membrane, this makes dispersing the nanoparticles challenging. According to Yu et al.[74], raising the concentration of nanoparticles may also enhance their agglomeration. Gilbert et al. [77] have claimed that changing the pH and ionic strength of the solution might produce agglomeration between nanoparticles. From the foregoing, a comprehensive understanding is required when adding metals or their oxides and other nanomaterials to avoid these defects when using them in manufacturing polymeric nanocomposite membranes.

Table 7: Examples of surface interaction between particles in the liquid phase [76]

Surface interaction	Generation mechanism
Van der Waals interaction	Short-range electromagnetic force between atoms and/or molecules
Electric double-layer overlap	Electrical interaction via overlapping of an electric double layer around a particle in solution
Steric interaction of adsorbed polymer	Short-ranged interaction via the overlap of an adsorbed polymer layer on particles
Bridge force	Bridge of surfactant formation and/or intra-particle polymer binding
Hydration force	Overlapping hydrogen-bonded water molecules on the particle's hydrophilic surface
Depletion	Negative absorption of solute and polymer due to lower attraction to the surface than solvent

The cost-effective treatment of radioactive waste resulting from nuclear accidents and the overconsumption of radioisotopes has become an important issue, and much effort has been dedicated to protecting the environment from such toxic radioactive elements. Amongst which, radioactive iodine in liquid is deemed one of the most real threats to humans, as it is produced post-use in medical and industrial applications. Mushtaq et al.[78], prepared the hybrid membrane, citrate stabilized Au (NPs) (mean diameter: 13 nm) were added to a syringe filter a unit containing a cellulose acetate membrane (pore size: 0.22µm, diameter: 4.91 cm^2) was used to eliminate iodine (^{125}I) from the aqueous solution. Scanning electron microscopy (SEM) analysis of the surface of the (Au-CAM) and cross-sectional images showed that the nanomaterials were incorporated stably on the cellulose nano-fibrils, as shown in Figure 6. Elemental analysis of the membrane using energy-dispersive X-ray spectroscopy (EDX) exhibited a set of peaks representing gold, along with carbon and oxygen atoms, which were observed from the carbohydrates in the cellulose polymer.

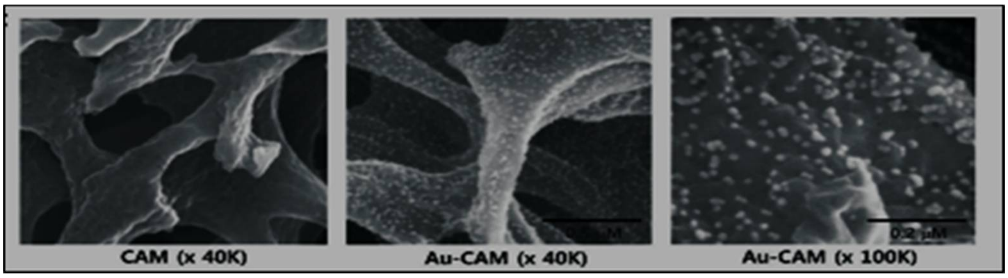

Figure 6: SEM images of the CAM (left) and Au-CAM (center: ×40 K magnified, right: ×100 K magnified). White particles in the images show AuNPs on the cellulose fibers[78]

To investigate the desalination performance of the composite membrane, the Au-CAM (100 pmol of AuNPs incorporated on each side) prepared by using a vacuum filter method was immersed into three aqueous solutions (pure water, 1.0 M NaCl, and 10 mM NaI) containing 10 nM [^{125}I] NaI. As a result, more than 95% of the radioactivity in pure water was rapidly adsorbed onto the Au-CAM membrane. Figure 7 illustrates the preparation steps to remove iodine by Au-CAM membrane.

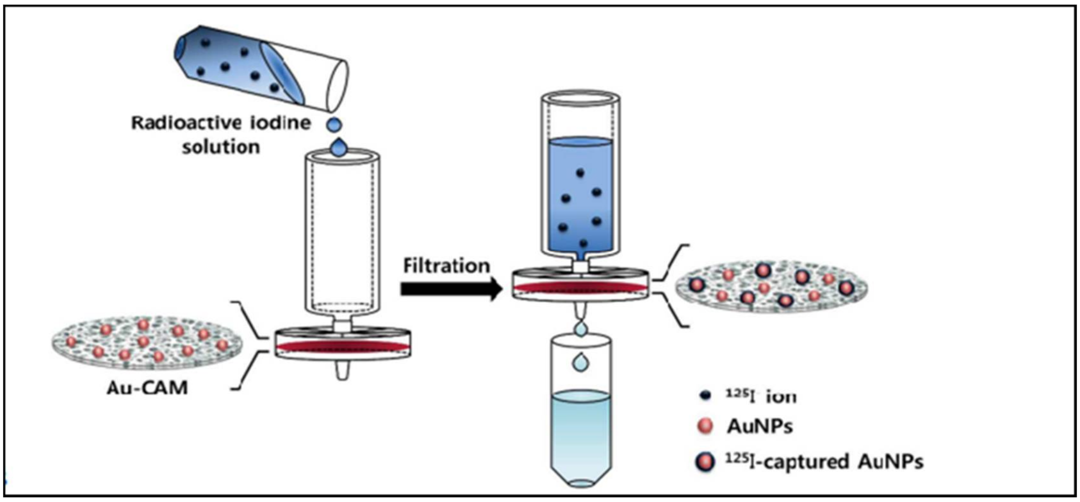

Figure 7: Schematic illustrates the desalination procedure for iodien using AuNPs immobilized on a cellulose acetate membrane (Au-CAM) [78]

In another study to remove heavy metal and radioactive ions from drinking water, He et al.[79] fabricated adsorptive hollow fiber nanocomposite membranes for As (V) removal by incorporating zirconia nanoparticles into the PSf membrane. Figure 8 (a-d) demonstrates the morphologies of hollow fiber adsorptive membranes derived from PSf/zirconia at different content of zirconia nanoparticles. The addition of zirconia nanoparticles increased the overall porosity and enlarged the formation of microvoids in the cross-section of the membranes. This is related to the hydrophilicity of the zirconia nanoparticles, which drew more water during the phase inversion technique. The batch maximum adsorption capacity of As(V) as high as 131.78 mg/g was achieved, as illustrated in Figure 8e. Figure 8f showed that the concentration of As in the first 106 L permeate obtained from PSf/zirconia membranes incorporated with 1.5 wt% of zirconia was below 10 mg/L, below the EPA WHO drinking water standards. In continuous filtration mode, PSf/zirconia membranes demonstrated an adsorption capacity of 25.83 mg/g, and no leakage of zirconia ions was detected during long-run filtration.

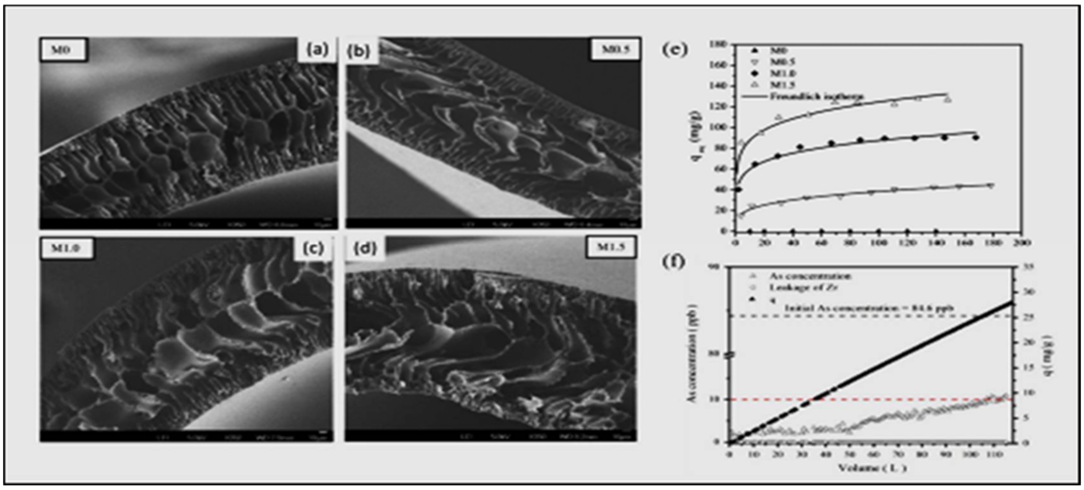

Figure 8: (a-d) SEM images of PSf/zirconia hollow fiber membranes at different loading of (a) 0 wt%, (b) 6.5 wt%, (c) 13.0 wt%, (d) 19.50 wt%, (e) adsorption isotherm study, (f) continuous filtration of M1.5 PSf/zirconia adsorptive nanocomposite membrane [79]

To enhance the performance of adsorptive membranes, modification of GO nanoparticles before the dispersion in the polymeric membranes has been performed [80]. Modified GO nanoparticles using polyaniline (PANI) by in-situ polymerization technique forming PANI@GO. Figure 9 (a) shows a digital image of GO and PANI@GO nanoparticles. The modified GO was then impregnated into the PES membrane to remove Pb(II) from the water. The adsorptive membranes with PANI@GO loading of 0.25 wt% (PPG1) exhibited lower pure water flux, porosity, and hydrophilicity compared to pristine PES and PES incorporated with 0.125 wt% of PANI@GO (PPG2) as demonstrated in Figure 9(b). This is due to forming a thicker membrane skin layer upon the addition of PANI@GO, as illustrated in Figure 9(c). However, this membrane exhibited a very high Pb(II) adsorption capacity of 202 mg/g according to the Langmuir isotherm model. It could be regenerated up to five cycles with 80% removal, as illustrated in Figures 9(d) and (e) respectively.

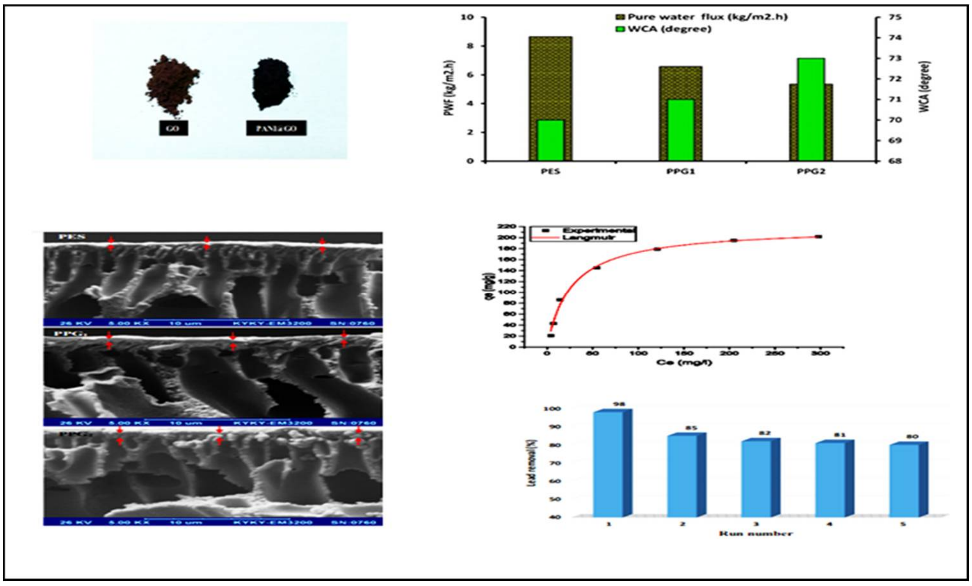

Figure 9: (a) Digital images of pristine GO and PANI@GO, (b) pure water flux, the porosity contact angle of Pristine PES, PPG1, and PPG2 membranes, (c)SEM images of PES, PPG1, and PPG2 membranes, (d) Langmuir isotherm analysis of PPG2, (e) regeneration of PPG2 membrane [80]

Although carbon-based adsorptive membranes show promising results in adsorption capacity, the hydrophobicity of carbon-based materials could reduce the water permeability of the adsorptive membranes. This condition is not favorable for water treatment applications. Hence, pre-treatment of fabricated carbon-based nano-adsorbent is crucial to increase the hydrophilicity and compatibility before introducing the membranes. Incorporating zeolite into the polymeric membrane to form polymer-zeolite membranes allows a higher degree of flexibility and cost reduction than alumina-based ones. Zeolites have well-defined pores and a framework that allows the replacement of alkali and alkaline earth metals to counteract the total negative charge between Si^{4+} and Al^{3+} ions, making them suitable for the remediation of heavy metal and radioactive ions in contaminated water. Yurekli et al. [81] successfully embedded zeolite nanoparticles into the PSf membrane in a flat sheet configuration to treat Ni(II) and Pb(II) in an aqueous medium. Impregnated faujasite NaX zeolite nanoparticles synthesized by hydrothermal method into PSf membrane. The synthesized faujasite NaX zeolite nanoparticles were ultrafine and showed consistency in particle size of 170 nm. They found that PSf/zeolite membrane comprised of 10 wt% zeolites and zero evaporation time (exposure of casting solution into the air) exhibited the highest water permeability of 57 L/m^2.h.bar. This result indicated that neglecting the evaporation stage could determine the highest water permeability due to instantaneous solidification, which altered the membrane structure. The sufficiently high loading of zeolite in the membrane has favorably enlarged the pore size to increase the water permeability of the membranes. Ni(II) and Pb(II) adsorption capacities were recorded as 122.0 and 682.0 mg/g, respectively. In addition to the aforementioned possibility of using nanomaterials embedded with polymeric membranes, hybridization between the chemically treated feed solution methods referred to in section 2.2 and nanocomposite membranes in this section is possible. Abbas et al. [13] studied a new type of nanocomposite membranes made by adding NaY zeolite with a carrying amount ranging from 0 wt% to 0.3 wt% with (nano diameter 59 nm) to polyethersulfone (PES). These membranes were employed to remove the cesium-137 isotope from a real sample collected from the Tuwaitha nuclear site. The addition of nano-zeolite to polyethersulfone was verified through characterization techniques. The results of this characterization described the interaction mechanism between NaY zeolite and polyethersulfone for the first time, as shown in Figure 10.

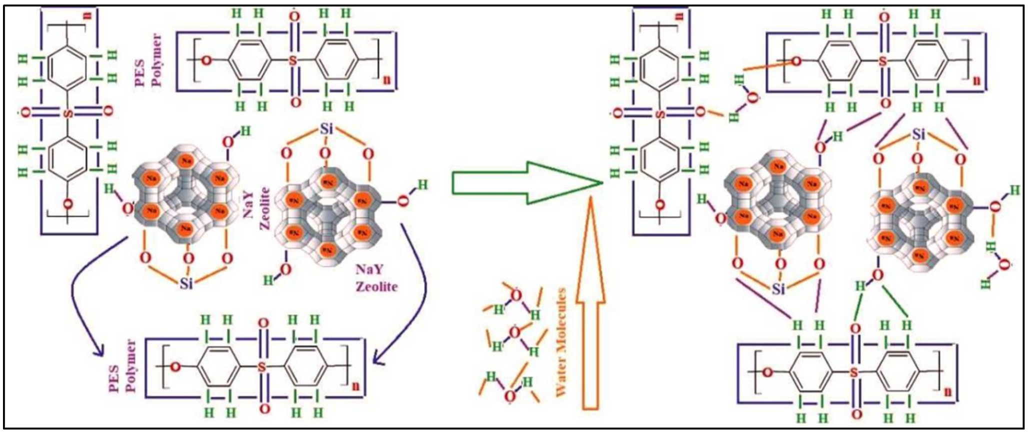

Figure 10: The proposed interaction model between the components of the PES/Zeolite membrane

This mechanism indicated in Figure 10 illustrates the bonding between the active groups of zeolite with the hydrogen group on the surface of the membrane through hydrogen bonding forces. This connection led to the membrane's wettability will increase compared with the pure PES polymer. This increase is due to a shift from the hydrophobic to the hydrophilic state. In addition, the swelling capacity will increase for the same reason, as shown in Figure 11.

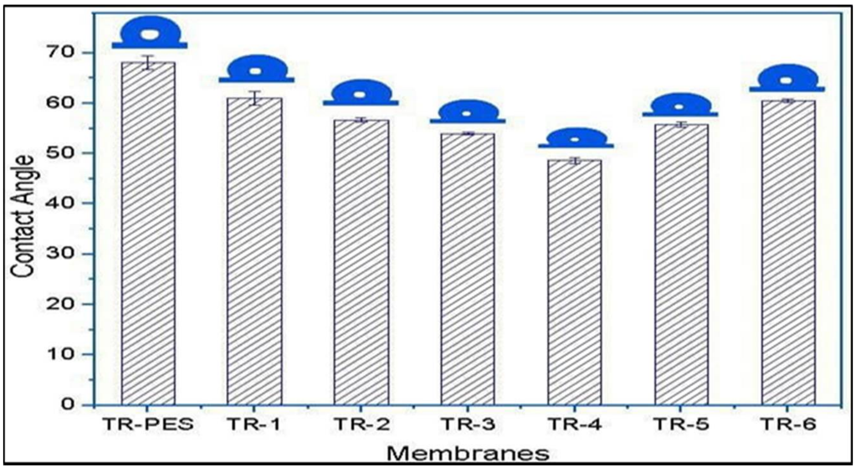

Figure 11: Static pure water contact angle of pure PES and PES membrane with various content of NaY zeolite wt.(%)

The purified water flux (PWF), porosity, and overall membrane efficiency were raised. As a result, a change in the harshness of the surface of the membrane will occur compared with the roughness of the pure PES polymer, and the pore diameters will be enlarged. This means that the surface and cross-section of membrane morphology will noticeably change, as shown in Figure12. Also, the ion exchange capacity of the current membrane will be enhanced. The PES/zeolite membrane carries a negative charge derived from Si-O and Al-O. The negative charge can efficiently attract cations, such as Cs^+, which will be attached to the surface and membrane pores. Furthermore, the ion exchange interaction between the Na+ ion in the zeolite structure and the cation present in the liquid waste improves the ion exchange efficiency for nanocomposite membranes. Results disclosed that the optimum removal rate (90.2%) was obtained by the membrane prepared using 0.15% NaY, while the decontamination factor (DF) was 10.2 at pH 7.5. Therefore, legend agent copper ferrocyanides (CuFC) have been added to the feed solution to promote the removal efficiency of ^{137}Cs and enhance the decontamination factor. As a result, about 99.2% Cesium retention and 121.2 decontamination factors were achieved.

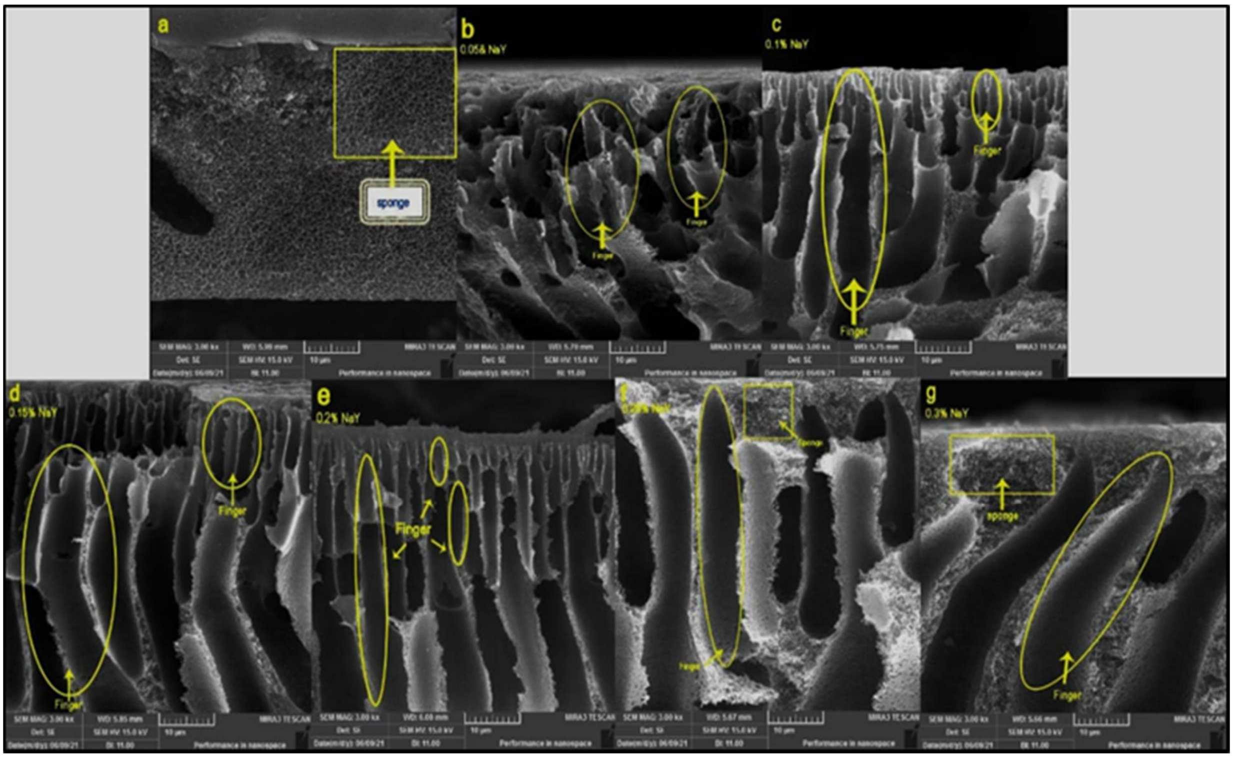

Figure 12: SEM cross-section for (a) pure PES membrane, (b) PES membrane with 0.05% zeolite, (c) PES membrane with 0.1% zeolite, (d)PES membrane with 0.15% zeolite, (e) PES membranewith 0.2% zeolite, (f) PES membrane with 0.25% zeolite, (g) PES membrane with 0.3% zeolite

In another study, PB/PAN nanofibers were created using electrostatic spinning and removed ^{137}Cs from seawater (1000 Bq/L) [70]. The mesoporous structure and enhanced surface area of PB/PAN significantly improved adsorption kinetics, achieving 70% removal efficiency in less than 1 hour. Furthermore, a new class of composite membranes made of UF - embedded with nanoparticles was used to remove the dissolved radioactive ions from wastewater. Long et al. [71] investigated a PVDF/ZIF-8 nanocomposite adsorption membrane by the contra-diffusion approach to effectively remove iodine from radioactive wastewater. According to the findings of this work, the combination of surface graft of poly (4-vinyl pyridine) brushes and optimum addition of sodium formate into ligand helps produce a well-intergroup and continuous ZIF-8 layer on the metal ions side surface of PVDF-g-P4VP membrane. In dynamic adsorption, the PVDF/ZIF-8 nanocomposite membrane showed a high flux of 66.19 (L/ m2.h.MPa) and an iodine removal efficiency of 73% till 180 minutes. Also, the pH of the feed solution shows a significant effect on the iodine removal efficiency can be maintained at around 92% in a weakly alkaline medium of pH=8. In other applications of nanomaterials embedded with membranes. Wen et al. [72] fabricated a multifunctional flexible free-standing sodium titanate-nanobelt (Na-TNB) membrane assembled as advanced radiation-tainted water treatment and oil uptake. The adsorption behavior of $^{137}Cs^+$ and $^{90}Sr^{2+}$ on Na-TNB membranes under various environmental conditions and the maximum adsorption coefficient value (Kd) for Sr^{2+} reaches $107 mL.g^{-1}$. In this study, the adsorption mechanism of the Na-TNB membrane is clarified by forming a stable solid with the radioactive cations permanently trapped inside. Figure 13 (a) shows the membrane without Na-TNBs. Figure 13(b) and (c) show the membrane with Na-TNBs in which ion exchange with Sr2+ and Cs+ ions took place and the phase transition to the rutile in concentrate acid solution. Besides, the engineered multilayer membrane is exceptionally capable of selectively and rapidly adsorbing oils up to 23 times the adsorbent weight when coated with a thin layer of hydrophobic molecules.

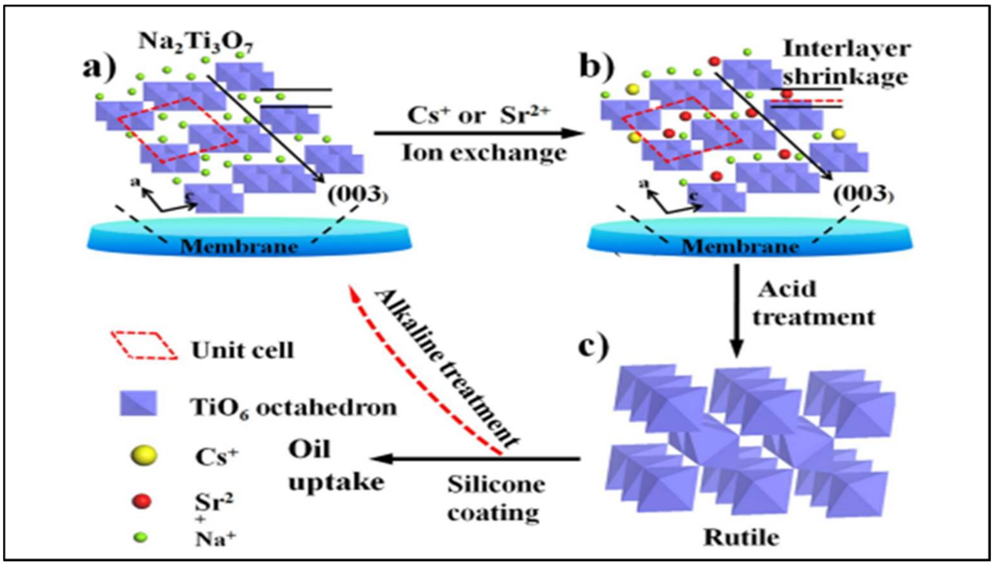

Figure 13: Schematic structural features of Na-TNBs before (a) and after ion exchange with Sr^{2+} and Cs^+ ions (b),(c) The phase transition to the rutile in concentrate acid solution. And the rutile can be further used to regenerate the titanates through alkaline treatment and silicone coating for oil uptake [82]

4.2 Modified Nanomaterials

In addition to the above characteristic properties of nanomaterials, more than one nanomaterial can be combined to improve the properties of these additives and then incorporated with polymeric membranes, especially UF and MF membranes. These materials add new functions to the membrane (absorption, ion exchange, and magnetization). By incorporating more than one nanomaterial, this modification adds stability and non-dispersion of the active nanomaterial by another nanomaterial on the membrane surface through interaction mechanisms and surface chemistry. Improving nanoparticles by merging more than one substance is one of the latest methods to remove radioactive ions. The reason is that the ions are characterized by their small radii size and ability to dissolve in water. Most importantly, these radioactive ions significantly impact human health and the environment in the long term, where they can be within ecological chains such as (cesium-137). Figure 14 depicts the cesium-134 and cesium-137 cycles as they transferred through the ecological chain.

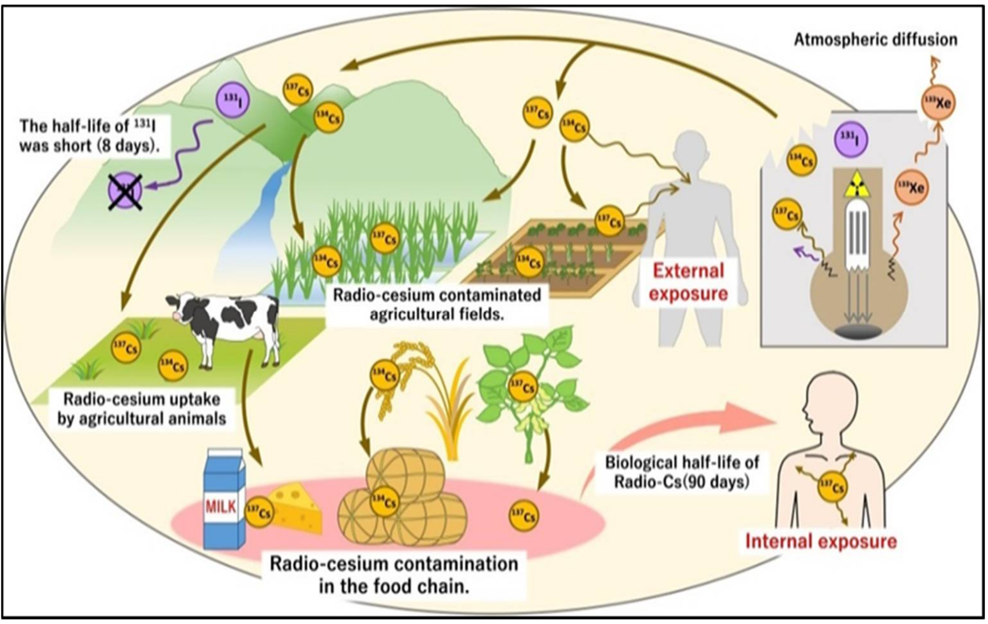

Figure 14: Main pathways of fission products involved ^{137}Cs and ^{134}Cs [83]

From Figure 14, it can be estimated how dangerous this isotope is to humans and the environment, as it can reach humans through drinking water or food products and air. After my accidents at the Chornobyl nuclear power plant in Ukraine and the Fukushima nuclear power plant in Japan, the concentrations of this isotope certificate increased, especially after the leaks of this isotope in the sea that took place in Fukushima. Hence, the researchers turned to get rid of this isotope's danger. The research relied on the technology of nanomaterials and transition metal materials and their inclusion in membranes as a method to get rid of this isotope. Liu et al. [16] developed surface-modified polyvinylidene fluoride (PVDF) membrane consisting of

Prussianblue (PB) and aminating silica (A-SiO_2) nanoparticles was fabricated for the selective removal of trace radionuclide cesium (Cs) from water. Also, they explained the interaction mechanism between the surface of the membrane and the functional groups of the added nanomaterials, as shown in Figure 15.

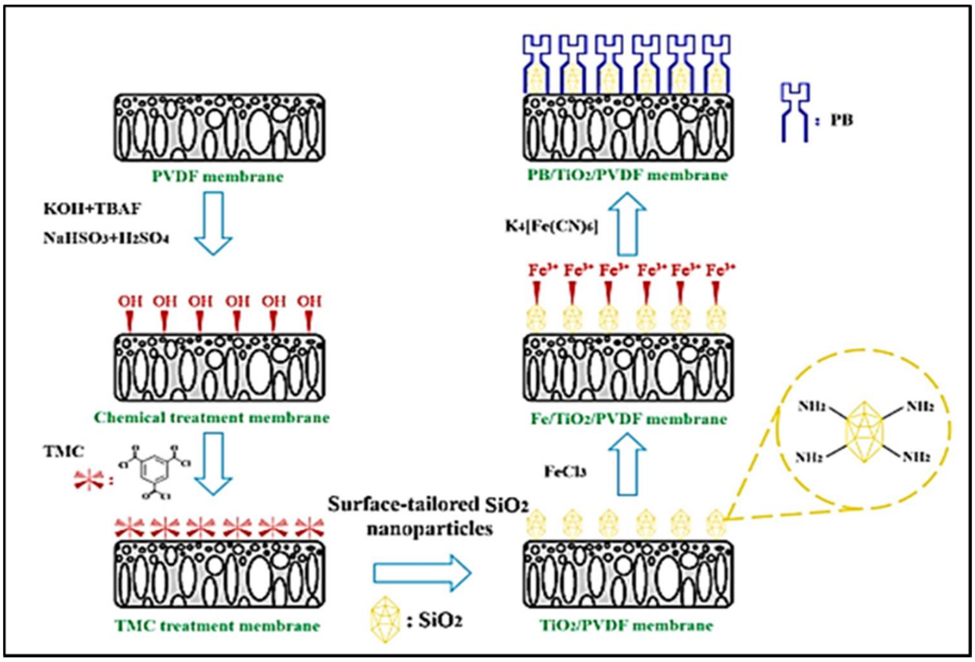

Figure 15: Schematic diagram of PB/A-SiO2/PVDF membrane modification process [16]

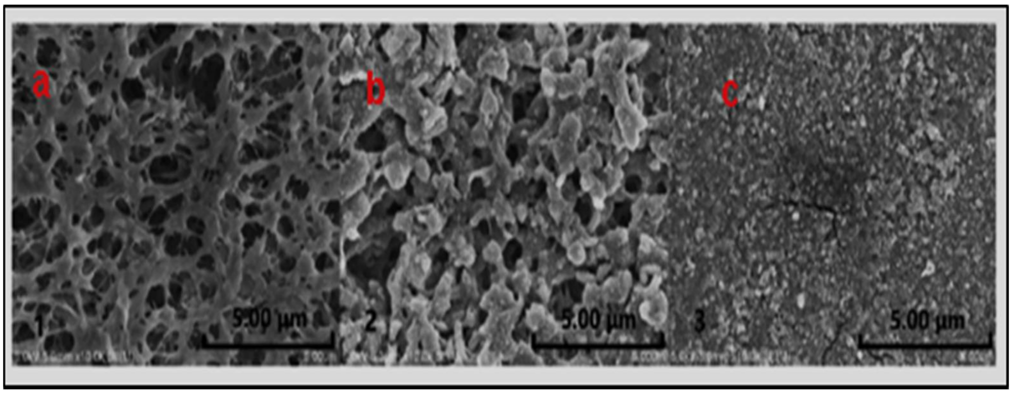

Figure 16: SEM images of membranes with different PB mass fractions. (a = 0%; b = 5.06% ; and c = 21.9%)[16]

Through the interaction mechanism shown in Figure 15, the membrane gained new properties diagnosed through characterization techniques. The SEM images indicated that the PB crystals appeared in the form of a cubic lattice, which was added to the membrane by (0 to 21.9%) that the surface of the membrane became more compact, while the pores were covered and gradually disappeared with the increase of the mass fraction of PB as observed in Figure 16.

The result of this membrane surface enhancement by nanoparticles, through the permeation flux and Cs removal rate of modified membranes with different mass fractions of Prussian blue, was investigated after 10 h filtration. It could be observed that the membrane flux decreased as the loading amount of PB increased from 0 to 21.9% on the PVDF support layer, while the Cs removal rate improved from 78% to more than 99%. Among these different PB mass fractions (X) deposited on the membrane surface, 8.0–15.0% loading fraction X was highlighted as the best result. It shows a relatively higher Cs removal rate (> 97%) and permeates flux (> 800 L/h·bar·m^2) in comparison with NF and RO membranes (< 50 L/h·bar·m^2). Prussian blue (PB) is known to be an effective cesium adsorbent, but the direct application of PB is limited by the difficulty of its recovery from the solution. For this reason, loaded in support media, poly(vinyl alcohol) (PVA) sponge, for use as a selective material for cesium adsorption [84]. The commercially available PVA sponge was functionalized by the addition of poly (acrylic acid) (PAA) (i.e., PAA-PVA) to enhance the PB immobilization, which increased both PB loading and binding strength. AS CONFIRMED BY FOURIER-TRANSFORM INFRARED SPECTROSCOPY, the AA functionalization changed the major functional groups from hydroxyl to carboxylic. PB was further synthesized in the PAA-PVA using layer-by-layer (LBL) assembly, which contributed to more stable PB formation and reduced detachment of PB during washing. The overall scheme of preparation is presented in Figure 17 (a) shows a schematic of PVA surface modification by AA, while Figure 17 (b) illustrates the LBL assembly for stable PB growth by adding a precursor as a final step. The PVA modification by AA is a type of radical polymerization. Free radicals on the PVA backbone can be formed by the chemical initiator.

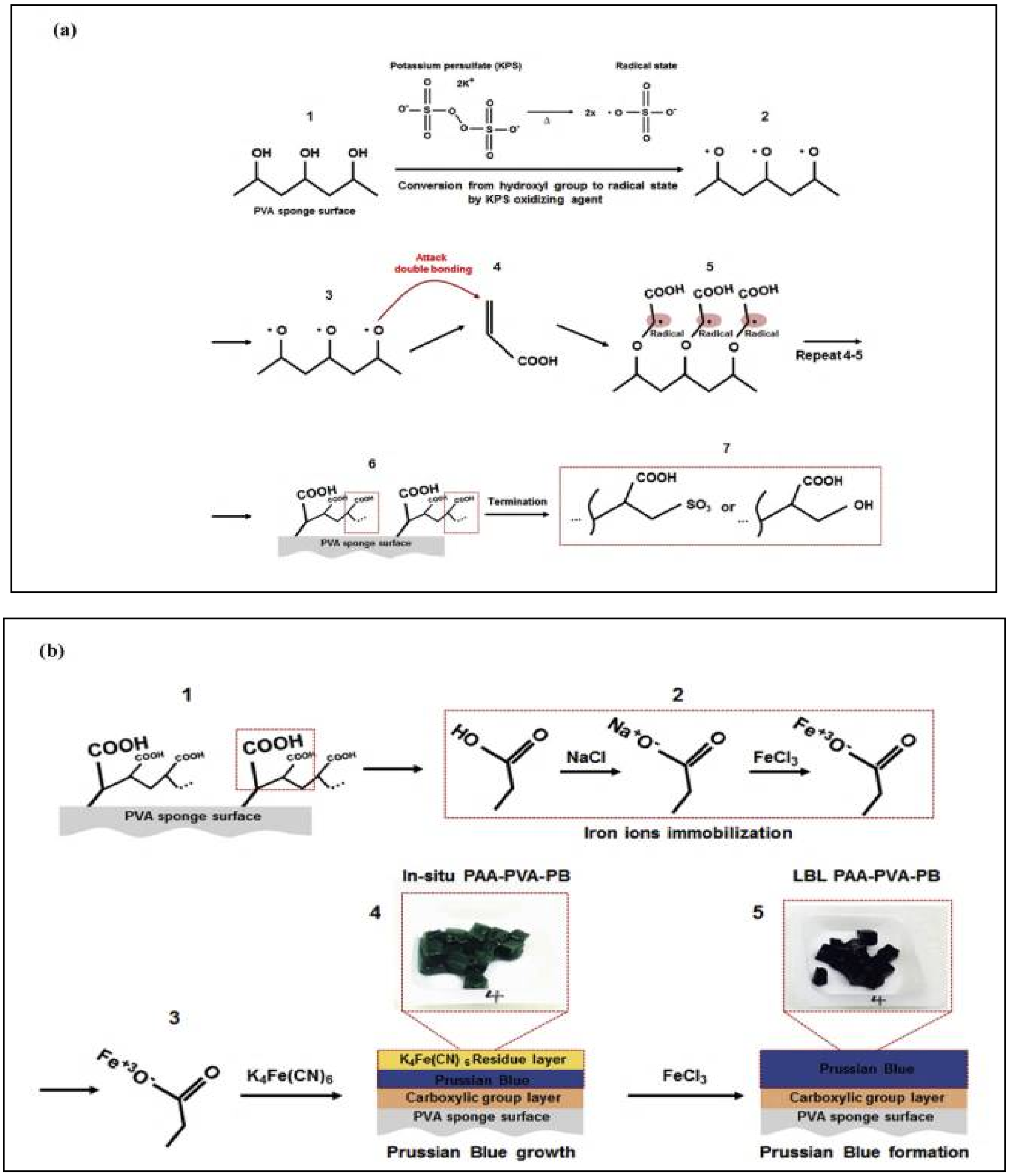

Figure 17: (a) Schematic of PVA surface modification by AA, (b) LBL assembly for stable PB growth by adding precursor as a final step [84]

Potassium persulfate (KPS) is an initiator. The hydroxyl group of PVA then reacted with the initiator to form a radical state. The acrylic acid monomer is then grafted on the radical site of PVA. The prepared adsorbent, PAA-L@PVA-PB, was tested for cesium adsorption capability. Cesium adsorption was equilibrated within three hours, and the maximum cesium adsorption capacity was 4.082 mg/g, which was 5.7 times higher than Pure-L@PVA-PB. Saberi et al. [85] fabricated poly(ε-caprolactone) electrospun nanofibers-copper hexacyanoferrate composite ion exchanger (PCL nanofibers–CuHCF) was prepared and used for the cesium ion removal from aqueous solution. The cesium ion removal experiments showed that the obtained PCL nanofibers–CuHCF composite exhibited excellent performance in removing cesium ions. The influences of several variables, such as temperature, pH, and contact time, were evaluated in batch experiments. PCL nanofibers–CuHCF composite ion exchanger showed excellent selectivity for Cs^+ in the presence of competing cations, mainly because of the excellent features of CuHCF.

The concentrations of Cs^+ and K^+ ions after treatment with the PCL-CuHCF composite were determined to explore the cesium removal mechanism. According to the results, the total K^+ concentration in the aqueous solution increased as the Cs+ concentration decreased, suggesting the ion exchange between K^+ and Cs^+ as the main adsorption mechanism. To get more insights into the adsorption mechanism of cesium ions, XRD analysis was performed on PCLCuHCF composite materials before and after cesium adsorption, as shown in Figure 18.

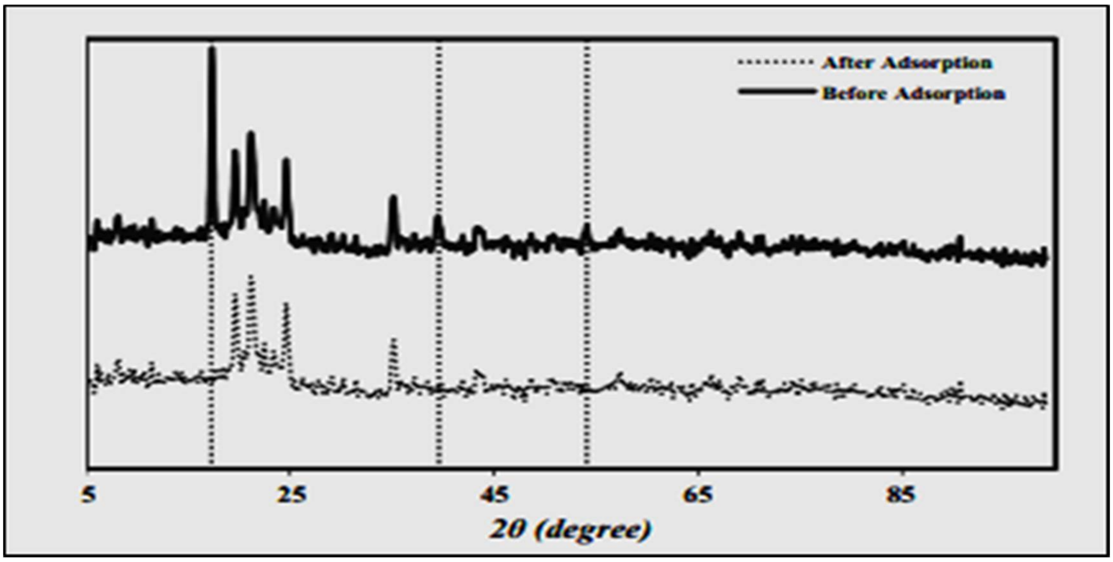

Figure 18: XRD patterns of PCL-CuHCF composite nanofiber before and after Cs^+ adsorption [85]

Following Cs^+ adsorption, the characteristic diffraction peaks are around 17.3°, 39.6°, and 54.1°, shown through the dashed line (disappeared, and the remaining peaks correspond well to those of CsCuHCF. From these results, the removal mechanism of Cs^+ by the PCL-CuHCF composite is the ion exchange between Cs^+ in solution and K^+ interleaved in the crystal structure of KCuHCF. Ion-exchange adsorption effectively separates cesium and rubidium from other alkaline metal ions in aqueous solutions. Jia et al. [86] developed a novel and facile method for in-situ preparation of Prussian blue (PB) nano-layer on porous polyacrylonitrile (PAN) membranes by heterogeneous nucleation in potassium ferrocyanide solution and then growth in FeCl3 solution, or potassium ferrocyanide solution as a single precursor. The effects of reactant concentrations and reaction time on the PB layer, the static adsorption, dynamic adsorption, desorption, and reuse of PB membranes were investigated. The micropores (about 50-200 nm in size) can be seen on the surface of the pristine PAN membrane, and the cross-section displays an asymmetric structure. For the PB/PAN membrane, with the increased $FeCl_3$ concentrations, the membrane micropores become smaller in size or even diminished. At the same time, the thickness of the PB layer increases from 62 nm to 195 nm, and the PB nanoparticle size decreases from 40 nm to 22 nm due to the large nucleation rate at high $FeCl_3$ concentration, Figure 19 shows SEM images of the membranes. Figure 19 (a) shows the surface of the pristine PAN membrane, while Figure 19 (b) shows the PB/PAN membrane prepared with 0.005 mol L-1 of FeCl3. Figure 19 (c) PB/PAN membrane prepared with 0.125 mol L-1 of FeCl3. The cross-section of the pristine PAN membrane in Figure 19 (d) and the PB/PAN membrane prepared with 0.005 mol L-1 of FeCl3 is shown in Figure 19 (e). Finally, the PB/PAN membrane prepared with 0.125 mol L-1 of FeCl3 is shown in Figure 19 (f).

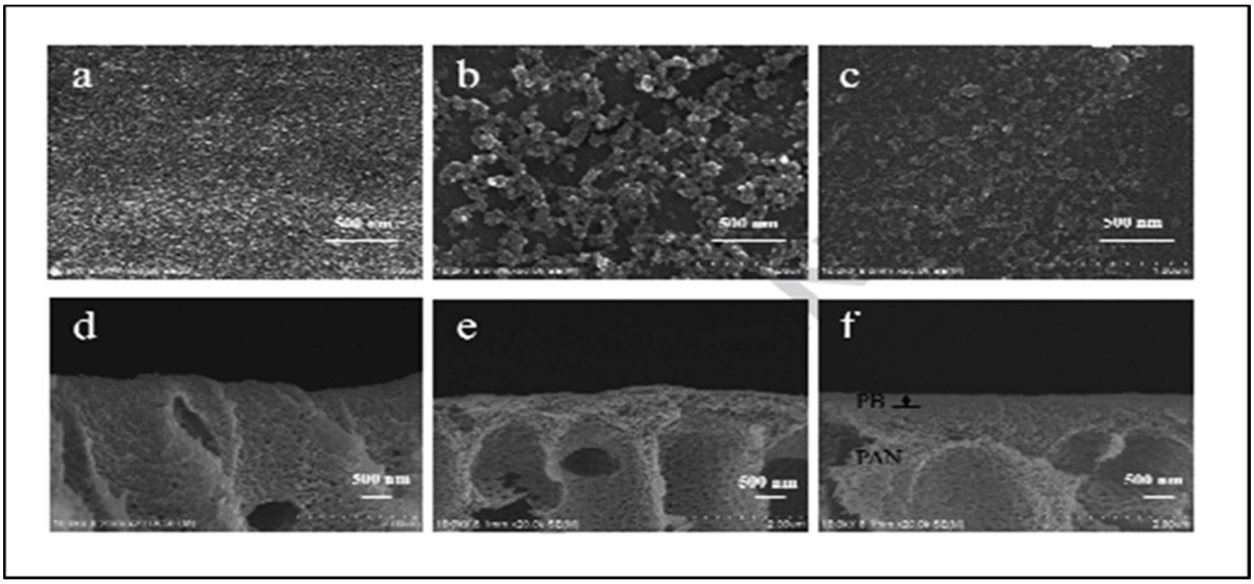

Figure 19: SEM images of membranes. (a)The surface of pristine PAN membrane, (b) PB/PAN membrane prepared with 0.005 mol L^{-1} of FeCl3, (c) PB/PAN membrane prepared with 0.125 mol L^{-1} of FeCl3, (d) Cross-section of pristine PAN membrane, (e) PB/PAN membrane prepared with 0.005 mol L^{-1} of FeCl3, (f) PB/PAN membrane prepared with 0.125 mol L^{-1} of FeCl3 [86]

The reason is that using $FeCl_3$ at a rate of (0.005) mol / L prevents the membrane's pores from completely blocking after covering it with a layer of PB. It can be visualized from Figure 19 that an increase in the amount of added PB led to an increase in the thickness of the layer loaded with it on the membrane. This leads to a loss of membrane properties such as (hydrophilicity) and thus affects the performance of membranes. The results showed that the maximum adsorption capacity of the PB/PAN membrane attains 0.714 mmol g^{-1}(25 °C). The ideal selectivity factor of Cs^+ vs. Li^+, K^+, and Na^+ is 41.76, 35.50, and 23.67, respectively. Compared with the PB powder, the adsorption performance of PB immobilizing on the membrane surface does not deteriorate. The PB/PAN membranes display a practical perspective in separating cesium and rubidium from other alkaline metal ions. Another study examined the ability of carbon nanotube (CNT) membranes in the selective removal of strontium (Sr^{2+}) and cesium (Cs^+) from water and wastewater [87]. This study demonstrates that Ar/O2 plasma treatment is a greener technology for producing CNT functionalized membranes for selective removal of contaminants from water to achieve “fit-for-purpose” treatment. Both pristine and functionalized CNT membranes showed excellent selectivity to Sr^{2+} over Mg^{2+} and $Ca^{2+,}$ while their selectivity to Cs^+ over K^+ and Na^+ diminished. Moreover, despite the complex water composition, the CNT membranes' selectivity for Sr^{2+} in wastewater effluent. Overall, the selectivity sequence of the CNT membranes for divalent cations supports the occurrence of inner-sphere complexation on CNT surfaces. In contrast, the monovalent cations were probably determined by electrostatic interactions. From the foregoing about incorporating nanomaterials with membranes in this section, it is the most suitable method with dissolved radioactive ions. It does not require complex equipment in its application to reduce the risks of these materials. In addition, these membranes can be used multiple times after the currency of reactivation and in the long term.

4.3 Nonradioactive Treatment

Membrane separations embedded with nanomaterials can be employed in other applications, including removing heavy metal ions, dyes, oily water in wastewater, and water treatment in petroleum industries. Alsalhy et al. [88] evaluated the performance of the PPSU membranes of poly(phenyl sulfone) (PPSU)membranes using different concentrations of ZnO nanoparticles as additives via the induced phase inversion method; a result of this study, the membrane hydrophilicity increases due to the addition of ZnO nanoparticles. The flux of the PPSU membranes with 0.025 wt. % ZnO as additives enhanced from 76 to 107 (L m-^{2} h^{-1} bar-1) with no significant change in solute rejection. Al-Ani et al. [89] investigated the impact of implanting TiO_2 nanoparticles on PVC for long-term UF membrane performance to treat refinery wastewater; these results clearly identified the impact of the TiO_2 nanoparticles content on the long-term PVC/TiO_2-NPs performance and confirmed the hypothesis that it is possible to use TiO_2 nanoparticles to effectively enhance the lifetime of membranes during their long-term operation through the improvements that have occurred from adding nanoparticles to polymeric membranes properties. Zeolite nanoparticles possess regular spherical morphology, non-oriented pore distribution, hydrophilic nature, high surface area, and thermal and mechanical stability. These properties make it widely used in various membrane separation processes, for instance, UF, desalination, and gas separation. Rezakazemi et al. [90] studied the transport properties of gases in polydimethylsiloxane (PDMS)/zeolite A mixed matrix membranes (MMMs) were determined based on pure gas permeation experiments, the MMMs exhibited both higher selectivity of H_2/CH_4 and H_2 permeability as compared with the neat PDMS membrane, suggesting that these membranes are very promising for gas separations such as H_2/CH_4 separation. Kadhim et al. [91] used graphene oxide nanoparticles (GO-NPs) were utilized to modify the polyethersulfone (PES) membrane and prepare mixed matrix membranes (MMMs) through the phase inversion method. The results of this work explained by adding GO to the PES casting solution resulted in longer lifetimes of the membranes due to enhancement in the fouling resistance and flux recovery efficiency (FRE) after backwashing. The dye removal was higher than 99% for all the membranes studied and both dyes (AB-210 and RB), at dye concentrations of 10, 50, 80, and 100 ppm and operating pressure of 3 bar.

5. Membrane Fouling

In pressure-driven membrane processes, membrane fouling is a complex problem due to several factors, as mentioned in the first section of this review. Interestingly, fouling of the membrane reduces flux but also permeates quality. In addition, membrane fouling affects membrane lifetime and increases feeding pressure and maintenance costs. Membrane contamination can also result from focus polarization, characteristic of RO and NF membranes [92]. Membrane fouling accumulates on the outer surface, as in RO membranes, or it can be inside the membrane's pores, as in UF and MF membranes. Membrane fouling can be classified according to the content of the feed solution, including colloids, inorganic and organic matter, large particles, and biological particles. Traditional methods of membrane fouling can be reduced by cleaning the feed solution and removing impurities from it by UF or MF membranes, followed by the main separation process by NF or RO membranes. It is possible through this treatment to reduce the layers accumulated on the surfaces of the dense membranes, which reduces the cost of cleaning them and increases the life of the membrane [93]. The removals of cesium (Cs) and strontium (Sr), two hazardous and abundant radionuclides in the aquatic environment were assessed with their isotopes in synthetic water containing Suwannee River natural organic matter (SRNOM), natural surface water (SW), and a wastewater effluent (WW) by two different types of ultra-low pressure RO membranes (M1 and M2) [94]. The rejections of Sr by membranes M1 and M2 were higher than 97.5% and 96.0%, respectively, and the rejections of Cs exceeded 90.0 % and 85.0 %, respectively, in the filtration of real water. The membrane M1 exhibited a more significant flux decline in the filtration of the SRNOM solution, while more severe flux declines were observed with the membrane M2 in the filtration of SW and WW. Protein-like materials with relatively high molecular weight were the main contributors to the flux decline, and humic-acid-like compounds had little effect on the flux decline. Donnan exclusion and size exclusion by humic-acid-like compounds improved the rejections by the membrane M2 with weaker hydrophilicity, while the cake-enhanced concentration polarization reduced the rejections of Cs and Sr by the membrane M1 with stronger hydrophilicity. The ionic strength in the real water resulted in the mitigation of membrane fouling. This study provided important insights into foulant characterization and the mechanisms of organic-fouling-enhanced rejections of Cr and Sr by ultra-low pressure RO membranes. In recent decades, nanomaterials embedded with pressure-driven membranes have been very popular in treating membrane fouling due to their unique properties in modifying the membrane surface and improving its performance, as mentioned in section 3. Alsalhy et al. [95] studied of embedding ZnO-NPs on PVC membrane performance of an anti-biofouling polyvinyl chloride/zinc oxide (PVC/ZnO) membrane was prepared using the phase precipitation method for application in a University of Cape Town membrane bioreactor-submerged membrane bioreactor (UCT-MBR) for treatment of actual hospital wastewater, resulted of addition of ZnO nanoparticles, up to 0.3 g, had a positive effect on the hydrophilicity of the PVC/ZnO membrane with decreasing the contact angle (CA) value by 17.775°. The pure water permeability (PWP) of the membrane improved by 315% with the addition of 0.1 g of ZnO. The cake layer build-up on the membrane surface was reduced from 52.8 to 10.42 μm with an increase of ZnO nanoparticles up to 0.3 g, as 0.4 g ZnO had no further effect on the cake layer thickness. The long-term PVC- 0.3 g nanoparticles were improved up to 70 days before membrane cleaning compared with 29 days for neat PVC membrane. The chemical oxygen demand (COD) removal efficiency of the UCT-MBR process was approximately similar, around 73.5% for all membranes. In other studies, hydrophilicity particles can be used to enhance the polymeric membranes to reduce fouling in contaminated wastewater. Yu et al. [96] investigated poly (vinylidene fluoride) (PVDF) UF membranes that were modified with poly(vinyl pyrrolidone) (PVP) through a two-step surface grafting method to enhance the antifouling properties. PVP of small molecules was used to modify the membrane pores, and cross-linked PVP modified the membrane surface. The pore modification enhanced the membrane flux from 130.0 $L\,m^{-2}\,h^{-1}$ to 170.7 $L\,m^{-2}\,h^{-1}$, and the surface modification increased the membrane hydrophilicity (i.e., the contact angle decreases from 93° to 83°). During the fouling tests with bovine serum albumin (BSA) solution and laboratory simulation low-level radioactive wastewater (LLRW), the modified membrane had a much slower flux reduction compared with the raw membrane. After water wash, the flux recovery rates of the BSA fouled membranes were 98% for the modified membrane and 46% for the raw membrane, respectively. Additionally, the modified membrane had the same rejection on nuclide and surfactant as the raw membrane. Based on these results, the two-step modified membrane is suitable to be used in LLRW treatment with the advantages of less frequent membrane cleaning and a longer membrane lifetime.

6. Cleanup Process of Membranes

During the treatment and removal of radioactive ions and organic and inorganic materials from radioactive liquid waste resulting from various activities of nuclear applications by separation by pressure-driven membranes, these membranes are exposed to membrane fouling.. it requires cleaning the membrane to perpetuate the lifetime of the membrane as one of the methods used to remove membrane fouling. To apply the membrane cleaning process, it must be subject to a procedure within specific items, including that the number of cleaning materials used should be small to avoid the generation of secondary waste and that the cleaning materials should be compatible with the membrane fouling materials to avoid the occurrence of secondary reactions with each other that may be more severe and cause an increase in fouling, and that the materials are proportional to Additive in cleaning operations with the materials from which the membrane is made to avoid damage. These are items that should be considered when initiating membrane cleaning operations. One of these procedures applied in cleaning membranes in pilot projects is using acidic or alkaline materials interspersed with rinsing operations with deionized water [97,98]. There are different common materials used as membrane cleaning materials, including (sodium hydroxide, citric acid, sodium tetra-polyphosphate, dilute sulfuric acid, sodium ethylenediaminetetraacetate, and EDTA) [7,99,100]. These compounds are widely applied and relatively inexpensive. Some reagents, however, are very good centers of nucleation and initialization of fouling and scaling. In such a case, the subsequent fouling can be even more severe after restoring the membrane. Hence, specially prepared membrane manufacturer formulations can be superior and worthy of consideration. Various cleaning agents are commercially available in the market and can be used for this purpose.

7. Conclusions and Future Outlook

Among the various membrane separation processes used for radioactive water decontamination, pressure-driven processes such as RO, NF, UF, and micro-filtration (MF) have already proven to be the most effective. UF and MF membrane pores are too large to remove radioactive ions from water. However, UF and MF membranes can be used with enhanced fluxes and rejected to remove dissolved radioactive ions when the size of the radioactive ions is enlarged by sorption, precipitation, or complexation with some additives to the feed. Additionally, the main effect of nanoparticles on membrane properties was increased porosity, pore size, and contact angle, resulting in improved membrane performance. Furthermore, the hydrophilicity of nanoparticles contributed to the improved hydrophilicity and wettability of the membrane's surface. Also, nanoparticles, when embedded with polymeric membranes, give new functions such as ion -exchange with radioactive cations. This review demonstrates that by adding nanoparticles to the polymeric membrane flexibly and cost-effectively, it is possible to avoid using RO, which requires high-pressure, high-energy consumption, and is expensive to remove radioactive waste. Nanomaterials have been demonstrated to be an effective adsorbent or ion exchange for selectively removing various radioactive species present in contaminated water. Moving forward, functionalized nanomaterials with high selectivity and radiation resistance are expected to play an increasingly important role in isolating and confining radioactive waste toward appropriate post-disposal. Moreover, as evidenced by their high adsorption or exchange capacity and desirable selectivity, nanomaterials can remove various types of radionuclides from contaminated water. Furthermore, radionuclide adsorption is affected by various factors, including the surface properties and functionality of the adsorbent. Surface complexation, electrostatic interactions, ion exchange, and surface precipitation have previously been identified as primary mechanisms between radionuclides and nanomaterials. Despite the superior performance of membrane technology, adopting innovative strategies is highly dependent on cost-effectiveness. The cost of radioactive effluents and the treatment strategies may differ significantly. For example, the adsorption or ion exchange method of embedded nanoparticles with membranes is usually considered one of the most cost-effective strategies due to the lack of widely used equipment and no secondary waste generation. However, using advanced materials at high costs combined with the membranes can be offset. In general, it can be treated using recyclable and environmentally friendly nanomaterials to remarkably solve the ion exchange/adsorption cost problem in nanocomposite films. The proposed approach is to use low-cost or renewable materials. Alternatively, cost-effectiveness can be improved by refurbishing consumables. The latter is particularly encouraged because low-cost materials usually possess low adsorption capacities. Renovating the developed consumables can cost less when they are included with the membranes to remove radioactive isotopes from contaminated water.

Author contribution

All authors contributed equally to this work.

Funding

This research received no specific grant from any funding agency in the public, commercial, or not-for-profit sectors.

Data availability statement

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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