

## Review and assessment on the separation of cesium and strontium from the aqueous stream

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### ABSTRACT

Wastewater containing radionuclides of cesium and strontium are considered to be more toxic contaminants. Radioactive wastes have become a great public concern due to the induced radioactivity, difficulty in the disposal, and also the long half-life period. These wastes can affect human health and also have become the biggest challenge for environmental safety. So, the safe management and disposal of radioactive wastes have received considerable attention worldwide. This review paper deals with the critical analysis of conventional separation technologies such as chemical precipitation, adsorption, ion exchange, solvent extraction, and membrane separation processes for the separation of cesium and strontium from wastewater. Also, emerging technologies in membrane separation processes were critically analysed for the separation of these contaminants from wastewater. This review attempts to examine the treatment methods for the effective removal of trace quantities of these contaminants in a huge volume of wastewater. In this study, the virtues as well as limitations of the technologies for the separation of cesium and strontium have been compared and critically analysed. This paper also validates the success of the hybrid technology in the ultrafiltration process known as size enhanced ultrafiltration (SEUF) for the separation of cesium and strontium from the aqueous stream by using biopolymers.

*Keywords:* Cesium; Strontium; Radionuclides; Adsorption; Ion exchange; Membrane processes; Size enhanced ultrafiltration; Polymer enhanced ultrafiltration; Complexation ultrafiltration

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### 1. Introduction

Removal of trace and ultra-trace levels of contaminants from aqueous streams is important and challenging. If not treated properly and disposed of in an isolated environment, the contaminants may find their way to ground water affecting the quality of water. Particularly care is required for very low level radioactive contaminants such as fission products including the long lived radio-isotopes  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ . These isotopes are hazards lasting for more than a few hundreds of years. Being in chemically insignificant levels of concentration, normal chemical methods of removal are not satisfactory. The literature indicates several methods of

treatment and management of these wastes involving physical, physico-chemical and chemical methods [1–6]. The best method of ensuring environmental safety is to recover and reuse. In this context, it is felt appropriate to review the methods either in practice or studied, intending to emphasize the potential of membrane processes towards a possible solution.

Based on the half-life and activity level of the waste, the radioactive wastes have been classified as waste to exempt, low-level waste, intermediate-level waste, and high-level waste. The low level and intermediate level wastes have been further subclassified as short-lived and long-lived waste [7]. These wastes have a high volume of low-level radioactivity.

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The sources of liquid radioactive wastes are nuclear power plants, research laboratories, medical diagnosis, and treatment, etc [7]. These radioactive wastes are hazardous to humans and also to the ecosystem. So, the proper handling, treatment, and disposal need attention for the protection of the environment [8]. The radionuclides present in the radioactive effluent are cobalt, cesium, strontium, tritium, etc [9]. The radioisotopes of cesium and strontium are the chief products of the nuclear fission process and are mostly present in low-level and intermediate-level radioactive wastes [10]. Even though cesium and strontium are at trace level, their radioactivity needs to be concerned [11].

Cesium is one of the natural elements with more isotopes and the only stable form of cesium is  $^{133}\text{Cs}$ . The radioactive isotope of cesium are  $^{134}\text{Cs}$ ,  $^{135}\text{Cs}$ , and  $^{137}\text{Cs}$  having a very long half-life period. The half-life period of  $^{137}\text{Cs}$  is 30 y and it keeps on existing in the environment for a huge period before it becomes a stable element [12,13]. Strontium has four stable forms of isotopes ( $^{84}\text{Sr}$ ,  $^{86}\text{Sr}$ ,  $^{87}\text{Sr}$ ,  $^{88}\text{Sr}$ ) and there are more unstable isotopes of strontium. The radioactive isotope of strontium,  $^{90}\text{Sr}$  has nearly 30 y of half-life period [14]. The radioactive cesium and strontium find their applications in nuclear power plants, medical and research laboratories. However, the growing population and escalating demand for energy have triggered the exploration of various conventional and non-conventional energy sources. Nuclear power is one of the conventional sources of energy that has been intensively employed in recent years. In nuclear power plants, radioactive cesium and strontium are the main products of uranium fission. The handling, disposal, and management of radioactive wastes have become a crucial part of nuclear power plants. Since their half-life period is very long and high yield from the fission process, they remain in the land, and aquatic system [12,14]. Also, these radioactive wastes are highly soluble in water, and their easy interaction and mobile nature with plant and animal tissues make them more diffusible through internal and external exposures [15]. Due to the radioactivity and other possible hazards, radiocesium and strontium ions have fascinated the attention of researchers towards the radioactive wastewater treatment. Exposure of living organisms to radioactive wastes results in severe health hazards such as kidney, liver cancer, and also genetic diseases [15]. The long-time exposure to radioactive strontium results in the replacement of calcium present in the bones of living organisms. The long time exposure to radioactive cesium can damage the liver and renal functions. Due to the long half-life period and toxicity of cesium and strontium, it has become a serious environmental threat that needs to be concerned [11]. The safe management and handling of radioactive wastes are based on the concepts of delay and decay, and concentrate and contain.

The radioactive wastes of cesium and strontium are highly soluble in water and present in more ionic forms at different pH. Cesium can form complexes with organic and some of the inorganic compounds and also acts similar to K and Na. The selective separation of cesium and strontium at a very low concentration (less than ppt level) from wastewater is difficult and does not form an insoluble complex. So, exposure to radioactive cesium can damage the liver and renal functions.

All the levels of liquid radioactive wastes comprise useful products such as cesium, and strontium. These contaminants/value-added products find many medical, research, and industrial applications. The energy contained by these radionuclides could be used for a variety of applications such as preservation of food, blood irradiation, treatment therapy, etc. Hence, the proper treatment and recovery of these valuable products could make the contaminants a material of resource.

This paper deals with the review on the developments in the separation of cesium and strontium from aqueous streams. This paper covers the current technologies adopted for the separation of cesium and strontium with their merits and limitations, assessment of the separation efficiency in terms of percentage removal or decontamination factor, and also the future trends involved.

For the separation of cesium and strontium from aqueous effluent various separation methods have been employed. The separation methods such as precipitation [1,16,17], co-precipitation [2], sedimentation [18], adsorption [4,19,20], ion exchange [5,6,21], evaporation [18], solvent extraction [22], plutonium uranium redox extraction [23], photo adsorption process [24], electrochemical process [25], and membrane separation processes [26–29] have been reported in the literature. Adsorption and ion exchange processes have been reported to be more efficient for handling a high volume of wastewater with trace quantities of cesium and strontium [30,31].

The conventional separation processes have their limitations such as chemical consumption, sludge handling, and disposal, energy requirement, cost of the adsorbent or resins, non-recoverability of separated ions, etc. In contrary to these limitations, the membrane separation processes possess many advantages and play a vital role in getting the separation of the cesium and strontium from aqueous streams. In the commercial pressure driven membrane separation processes, ultrafiltration holds the advantages such as low-pressure operation, less usage of chemicals, less footprint area, ambient condition operation, etc. However, the size of the contaminants is much smaller than the pores of the UF membrane. In recent years, the advent of size enhanced ultrafiltration has emerged as a potential solution for the use of UF, not only for the separation of cesium and strontium but also for the separation of other trace level contaminants. In this context, we present in this review how UF can be harnessed for recovering the valuables and amicably preserve the environment, by the use of size enhancing biopolymers. Likewise, the potential of other membrane processes such as membrane distillation and hybrid membrane systems have also been highlighted.

## 2. Chemical precipitation

This process has been widely employed in the separation of radioactive wastes from the effluent stream. It's the most versatile process and simple in operation. This process involves the addition of precipitating agents with the wastewater which can form insoluble hydroxides, carbonates, or sulphides with the target ions. This insoluble precipitate can further be separated by sedimentation, filtration, or membrane separation process. However, it involves

a huge volume of chemicals as precipitating agents, and also the removal of sludge becomes the major drawback of this process. Since this process can be operated batch-wise, the effluent can be periodically treated and discharged. This reduces the risk of accumulating radioactive wastes.

The various precipitating agents such as copper ferrocyanides, nickel ferrocyanides, phospho molybdates, tetraphenylborate, sodium titanate, ferric hydroxide, titanium hydroxide, barium sulphate were widely employed for the separation of cesium since they have excellent mechanical strength and stability towards radiation [27,32]. The separation of cesium and strontium by chemical precipitation using  $\text{Cu}_2\text{Fe}(\text{CN})_6$  and  $\text{Ca}_3(\text{PO}_4)_2$  respectively were reported by Rao et al. [27]. The influence of ammonium nitrate on the separation of cesium and strontium has been studied and it was found that at a low concentration of cesium, there was no influence of ammonium nitrate for the separation. The decontamination factor was observed to increase with the process of  $\text{Ca}_3\text{PO}_4$  precipitation of strontium followed by ultrafiltration. The two-stage precipitation for the separation of cesium was carried out using potassium ferrocyanide, nickel nitrate, and ferrum nitrate as precipitating species and successful decontamination factor have been obtained [33]. This process could be fruitfully implemented for the separation of cesium in large-scale applications. The procedure for the two-stage precipitation is shown in Fig. 1.

The precipitating agents calcium phosphate, calcium carbonate, and iron hydroxide were employed for the separation of strontium because of their better ion exchange capability. The separation of cesium by precipitation technique has been studied by Schultz and Bray using phosphotungstic acid [22].

In the past few years, co-precipitation has been studied extensively for the separation of strontium from

wastewater. Many hybrid systems have been reported in the literature for the separation of cesium and strontium [2,34]. One of the hybrid processes is coprecipitation followed by microfiltration/ultrafiltration. Flouret et al. [34] reported the removal of strontium by coprecipitation using barium sulfate seeds. Based on the experimental findings from their studies, it was reported that the coprecipitation followed by membrane filtration such as microfiltration could enhance the decontamination factor. The separation of strontium by hydraulic pellet coprecipitation (HPC) followed by microfiltration (MF) have been reported in the literature with significant decontamination factor [2,35,36]. The experimental findings of Wu et al. [2] show the feasibility and efficiency of the HPC-MF device for the separation of strontium from the simulated wastewater with a significant decontamination factor. The separation of cesium from the aqueous stream and simulated wastewater by nickel hexacyanoferrate(II) as a co-precipitant and sodium lauryl sulfate, cetyltrimethylammonium bromide, or dodecyl amine as a collector has been studied and the experimental findings reveal the excellent percentage removal (99%) and decontamination factor (>110) [37].

Besides the merits such as enhanced percentage removal and decontamination factor, the chemical precipitation process suffers from the limitations such as handling and disposal of a huge volume of sludge, consumption of precipitation chemicals, and requirement of additional separation processes such as settling, sedimentation, or filtration for the separation of contaminants. The currently reported works have also not focused on sludge disposal. An adequate technology is required to handle the huge volume of sludge to upgrade this process at the field level. Recoverability and reuse of chemical precipitating agents could minimize the generation of a huge volume of sludge.

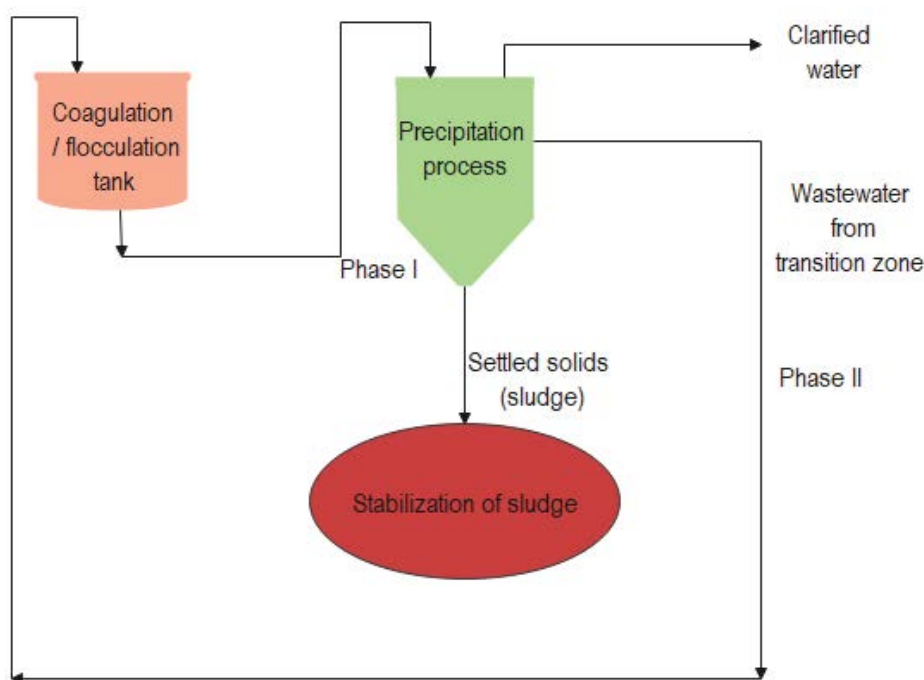


Fig. 1. Two-stage precipitation process for the separation of cesium from wastewater [35].

### 3. Adsorption and ion-exchange

The adsorptive capability and selectivity of the adsorbents or ion exchange resins for the separation of radioactive wastes may change on the exposure to the effluent. Inorganic resins are more resistant to radiation as compared to organic resins [38,39]. The radioactive wastes of cesium and strontium have been reported to be effectively separated from wastewater by using the well-known ion exchange resin, zeolite. The inimitable characteristics of zeolite resin such as good mechanical and thermal strength, resistance to radiation, and ion exchange capability, make it suitable for the separation of radioactive cesium and strontium from wastewater. However, the contaminant gets adsorbed over the surface of the adsorbent and creates the secondary pollutant. The treatment of this secondary pollutant becomes the hurdle in the adsorption or ion exchange process for the separation of radioactive wastes. Also, the recoverability of adsorbent or resin is very poor.

#### 3.1. Adsorbents

Adsorbents from natural materials show less efficiency compared to synthetic adsorbents which are expensive. Adsorbents such as zeolite, crystalline silicotitanate, ammonium phosphomolybdate have been employed for the separation of radioactive cesium [40]. A wide range of inorganic adsorbents has been employed for the treatment of a large volume of effluents to remove and concentrate radionuclides. Synthetic adsorbents find applications due to their better selectivity, adsorptive capacity, and stability towards radiation, stability to withstand acid, and slightly alkalic medium [41]. A wide range of ion exchange resins such as crystalline silicotitanate has been reported in the literature for the separation of radioactive wastes [42]. The sorption of  $\text{Cs}^+$  and  $\text{Sr}^{2+}$  by various resins has been reported in the literature. The sorption technique is the desired process for handling a huge volume of effluent and can treat the effluent to have minimum contaminants which can be disposed of or sent for landfill disposal. Studies have been carried out to analyse the natural adsorbents such as activated carbon, hydrargillite, montmorillonite clay, palygorskite, AMP-calcium alginate composite [43], PAN-based adsorbents [44–47], crystalline silicotitanate (CST) [48–50], metal-organic frameworks (MOF) [51], zirconium selenomolybdate [52], ammonium molybdophosphate (AMP) and ammonium tungstophosphate (ATP) [53–55] for the separation of cesium and strontium from aqueous streams [56,57]. There are a variety of chitosan-based adsorbents employed for the separation of radionuclide from wastewater. In literature, magnetic chitosan has been reported for the separation of strontium from wastewater [57,58].

Various adsorbents such as organic and inorganic reported in the literature have shown better stability towards radiation and heat [59,60]. The spent inorganic adsorbents can be utilized for the production of ceramics and glasswares. However, these adsorbents are still under the development stage to handle a huge volume of effluent due to their insufficient ionic selectivity or complex formation. On the other hand, adsorbents synthesized from natural materials which are cheap and abundantly available, such as functionalized

montmorillonite clay has shown better ionic selectivity [61], especially cation ion exchange capacity [62,63]. The functionalized porous montmorillonite has been synthesized and applied for the separation of cesium and strontium from the effluent [64].

The conventional adsorbents due to their low adsorption capacity and recoverability have attracted attention towards the synthesis of nanostructured adsorbents such as magnetic zeolite composites, and dual functional micro-adsorbents for the separation of radioactive wastes. In recent years, the synthesis of low cost, hydrophilic, multi-functional, and highly porous biopolymeric hydrogel-based adsorbents is finding much attention.

#### 3.2. Separation studies on the removal of cesium and strontium by adsorption and ion exchange

Abdel Monem et al. [65] synthesized a magnetic nano-zeolite for the separation of cesium and strontium and performed batch scale binary ion-exchange experiments at different temperatures, pH of the solution, contact time, and concentration. The experimental results were found to be more satisfactory for the separation of cesium and strontium from wastewater using magnetic nano-zeolite. However, there is a drawback in the separation of nano-zeolite from the spent stream. The earlier studies reported the centrifuging of the spent stream to separate the nano-material which is expensive. Recent studies reported the magnetic separation of nano-adsorbent, which is economical and efficient. The thermodynamic studies revealed that the separation is an endothermic process with a positive change of entropy.

Brown et al. [66] examined the separation of cesium using activated carbon and zeolite as an adsorbent and found 10% and 70% adsorption respectively. Liang et al. [67] investigated the separation of cesium and strontium from the aqueous stream by adsorption using mordenite as an adsorbent. Mordenite is one of the abundantly available forms of zeolite and exhibits excellent ion exchange ability, thermal stability which makes it suitable for the separation of cesium and strontium from aqueous streams. The studies show that the separation mechanism is by ion exchange and also slightly by physical adsorption. Jianlong Wang and Shuting Zhuang [68] have reported the separation of cesium by using crown ethers and calixarenes derivatives as adsorbents which are selective to cesium separation. The mechanism of adsorption involves the ion-dipole interaction or cation –  $\pi$  interaction. The separation efficiency depends upon the coordinating structure and the adsorption can be by physical and chemical grafting [69–71]. Compared to chemical grafting the physical adsorption is simpler and by intermolecular attraction but the interaction is weak [68].

Xudong Zhao et al. [72] have reported the selective separation of cesium and strontium from wastewater by magnetic Nb-Substituted Crystalline Silicotitanate. The adsorbent showed excellent stability towards radiation and acidic condition. The studies show the equilibrium time of 8 hr for the separation of cesium and strontium with adsorption capacity of 14.38 mg/g at basic pH and 11.18 mg/g at acidic pH for strontium and cesium respectively. As the separation involved two different extreme pH conditions, the selective

separation of cesium and strontium could be possible by altering the pH.

Sayed S. Metwally et al. [73] have examined Hydroxyapatite-cobalt(II) ferrocyanide for the separation of cesium and strontium since it has excellent ion exchange capability. The process involves the ionic interchange of  $\text{Sr}^{2+}$  with  $\text{Ca}^{2+}$  since these two are having an identical ionic radius. The percentage sorption of  $\text{Cs}^+$  and  $\text{Sr}^{2+}$  was observed to be 99% and 85% respectively at temperature 318K. Asgari et al. [74] studied the synthesis of neodymium (Nd) as a basic metal and benzene tricarboxylic acid (BTC) as a ligand forming Nd-BTC metal-organic framework and its application for the separation of cesium and strontium. The mechanism of the process is illustrated in Fig. 2. The maximum adsorption capacity of 86 mg/g of cesium and 58 g/g of strontium at pH 8 have been reported.

Kim et al. [64] investigated the ionic exchange capacity of Montmorillonite clay for the separation of cesium and strontium from the aqueous stream and it was enhanced by the facile solid-state NaOH treatment to create micropores through desilication. The results show nearly 300 mg/g adsorption capacity for cesium and 200 mg/g for strontium at an equilibrium concentration of cesium and strontium.

Chitosan coated on Fuller's earth in the form of beads has been studied for the separation of strontium by Shameem Hasan et al. and maximum adsorption capacity as 30 mg/g at 298 K has been reported. The functionalized

adsorbents for the separation of cesium and strontium have been adopted in recent years.  $\text{Cs}^+$  and  $\text{Sr}^{2+}$ , coexist in trace amounts in a concentrated solution of competitive cations including  $\text{Na}^+$ ,  $\text{K}^+$ , and  $\text{Ca}^{2+}$ , the sorption method is often desired to process a high volume of wastewater leaving minimal waste for packaging and landfill disposal.

There are a lot of scopes for the development of technology for increasing the sorption capability and selectivity [14]. In recent years, there are so many developments in the synthesis of nano-scale functionalized inorganic resins with better sorption capacity, selectivity, and mechanical strength for the separation of cesium and strontium. A brief literature review on the separation of cesium and strontium by adsorption and ion exchange processes is given in Table 1 [20,75–85].

There are certain limitations of adsorption and ion exchange processes for the separation of cesium and strontium. When the polymeric adsorbents or resins are exposed to radioactive wastes, it may affect the cross-linking. There might be the formation of low molecular weight complexes which can increase the solubility and transferability of contaminants thereby decreasing the retention of contaminants in the effluent streams [39]. Also, there is no possibility for recovery and reuse of adsorbents and ion exchange resins. However, recent developments in silica-based hybrid materials make them stable to withstand radioactive wastes.

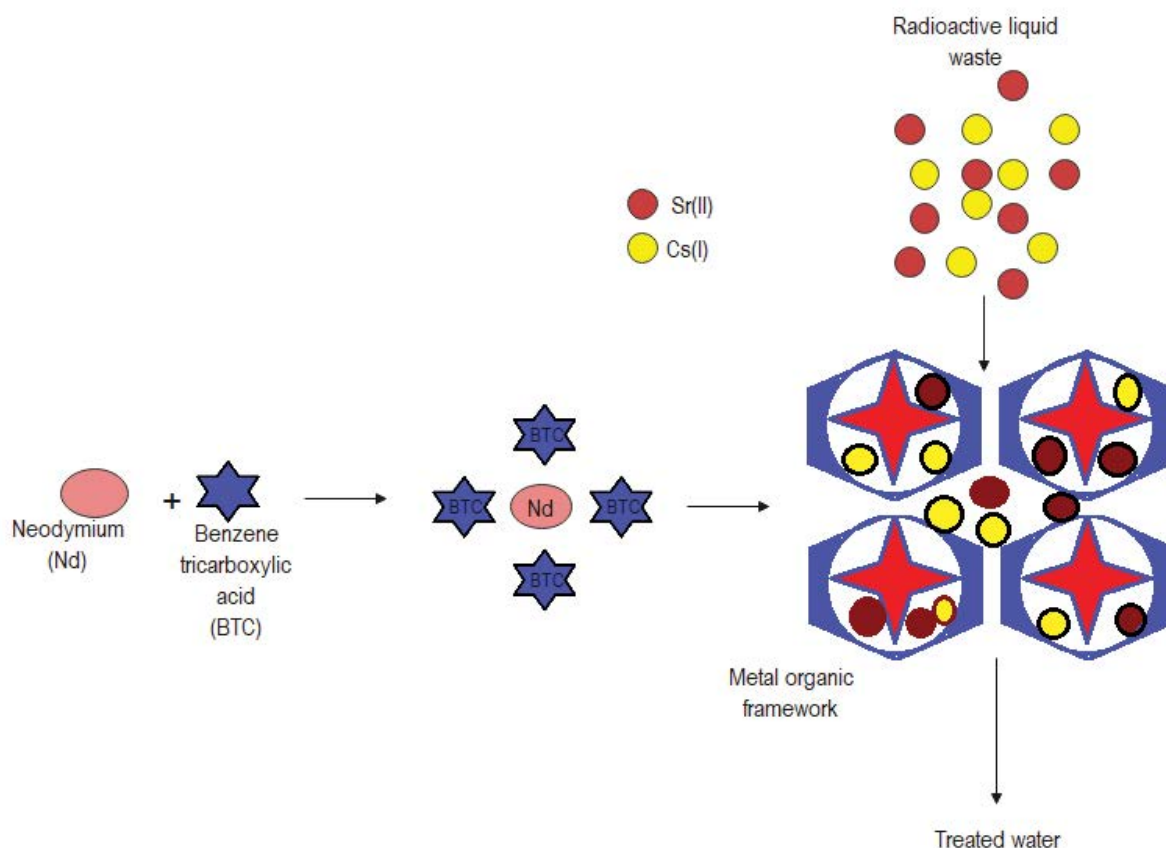


Fig. 2. Schematic representation of the sorptive process for the separation of  $\text{Cs}(\text{I})$  and  $\text{Sr}(\text{II})$  [74].

Table 1

A brief literature review on the separation of cesium and strontium from the aqueous stream by adsorption and ion exchange processes

Metal ion	Adsorbent/Ionic resin	Initial concentration	Percentage adsorption/adsorption capacity	References
Cs <sup>+</sup> Sr <sup>2+</sup>	Nanostructured chitosan/molecular sieve-4A hybrid	10 mg/L	92%–94%	[75]
Cs <sup>+</sup> Sr <sup>2+</sup>	Natural zeolite	10 mg/L	67.8% 93.5%	[76]
Sr <sup>2+</sup>	Activated carbon	100 mg/L	5.07 × 10 <sup>-4</sup> mol/g	[77]
Sr <sup>2+</sup>	Almond green hull	45 mg/L	79.7%	[20]
Cs <sup>+</sup> Sr <sup>2+</sup>	Fe(II)-modified geopolymer	102 mg/L 100 mg/L	67.2% –	[78]
Cs <sup>+</sup> Sr <sup>2+</sup>	Highly dispersed silica	–	53.9 mg/g 12.9 mg/g	[79]
Cs <sup>+</sup> Sr <sup>2+</sup>	Sulfur-modified chabazite (CHA)	5.87 ppt 9.20 ppt	95% 97%	[80]
Cs <sup>+</sup> Sr <sup>2+</sup>	Ammonium molybdophosphate–polyacrylonitrile	10 mM 10 mM	0.61 mmol/g 0.18 mmol/g	[81]
Cs <sup>+</sup>	NH <sub>4</sub> PMA/SBA-15		70.92 mg/g	[82]
Cs <sup>+</sup>	NH <sub>4</sub> PMA/SiO <sub>2</sub>		21.52 mg/g	[82]
Cs <sup>+</sup>	Poly(AAc-co-B18C6Am) hydrogels	2 mM	74.6 mg/g	[83]
Cs <sup>+</sup> Sr <sup>2+</sup>	MIOC	20 mg/L	41.6 mg/g 47.6 mg/g	[84]
Cs <sup>+</sup>	NaNCl (natural clinoptilolite) NaNCh (natural chabazite) NaNM (natural mordenite) NaSM (synthetic mordenite)	97.59% 98.10% 98.07% 97.98%		[85]

Recent progress in the design and preparation of nanostructured inorganic materials has attracted growing interest based on the potential for improving the retention performance when coupling such functionalities as ion exchange capacity, structural flexibility that may result in steric retention effects, as well as the propensity to interact specifically with the target metal cations. The competent technology and cost-effective adsorbent for the separation of strontium with high salinity have been reported in the literature [86]. The study utilized zeolite (Z4A) from the Bayer process as an adsorbent for the selective separation of strontium from seawater. This study explored the influence of cations Na, Mg, Ca, and K in the selective separation of strontium from seawater. The results show that the separation of strontium was not affected by the presence of other cations. The separation of cesium and strontium by ion exchange process using clinoptilolite has been reported recently. Clinoptilolite is a naturally occurring component with fewer impurities and it can be processed to increase its efficiency. It has excellent stability to withstand radiation [29,87–89]. Clinoptilolite, pre-activated with con. NaCl and HCl has been studied for the separation of cesium and strontium from wastewater from simulated salinity water. The results reveal nearly complete removal of cesium using pre-activated clinoptilolite

and moderate removal of strontium [90]. Also, the study shows the moderate separation of cesium and strontium from simulated seawater due to the interaction of potassium ions. Various natural and synthetic zeolite have been investigated for the past few years, and the studies prove the efficiency of natural chabazite (NaNCh) for the removal of cesium [85]. In a recent study, sodium iron titanate has been examined for the selective separation of cesium and strontium from alkaline solution and exhibited promising results [91]. However, the ion exchange process suffers from certain limitations such as the regeneration of resins and the huge quantity of spent resins. The innovation of low-cost resins and recoverability of resins could make this process suitable for the required applications.

#### 4. Solvent extraction

Due to the better efficiency, selectivity, and ease of operation, a lot of research studies are going on in the field of solvent extraction for the separation of cesium and strontium [92]. However, the feasibility of the process for the separation of Cs<sup>+</sup> has not yet been proved [93]. Various solvent extraction techniques such as Transuranic extraction, strontium extraction, chlorinated cobalt dicarbollide/polyethylene glycol, phosphine oxide process, and universal

solvent extraction process have been reported in the literature [94]. The separation of cesium ions by the combined mechanism of solvent extraction and adsorption by crown ethers and calixarenes has been reported recently [13]. Since the cost of the solvent and equipment involved are expensive, solvent extraction suffers from its large application for the treatment of cesium and strontium [95]. In recent years, membrane solvent extraction has become more popular for the separation of organic contaminants. By proper process modification and selection of solvent, this process could offer a noteworthy solution for the separation of cesium and strontium.

## 5. Membrane separation processes

In the past few years, the applications of membrane separation processes have been broadening due to their merits such as (i) ambient temperature operation, (ii) no phase change, (iii) minimum chemicals usage, (iv) less footprint area, (v) simple in operation, (vi) can be coupled with other conventional operations and (vi) high selectivity [96,97]. The recent developments in the membrane separation processes, especially pressure-driven membrane separation processes such as ultrafiltration [98,99], nanofiltration [100] and reverse osmosis [87,88], concentration driven processes such as membrane distillation [89,101], and membrane solvent extraction, and some hybrid membrane separation processes made them more compatible, feasible and also economical for the separation of radioactive wastes in aqueous streams. Table 2 shows the applications of membrane processes for the separation of cesium and strontium from the aqueous stream [29,95,99,100,102–104].

### 5.1. Forward osmosis

One of the membrane separation processes, compared to RO, has equally proven to be an efficient technology for

the removal of radionuclides from aqueous streams. This process is driven by the osmotic pressure difference across the membrane, without the intervention of any external energy source [105–107]. Unlike RO, it is not a high-pressure operation. So, this process could be an effective alternative for RO for the separation of radioactive wastes.

Wang et al. [95] studied the feasibility of commercial forward osmosis membranes cellulose triacetate with a cast nonwoven support, cellulose triacetate with embedded polyester screen support, and polyamide-based thin-film composite with embedded polyester screen support for the separation of cesium from aqueous stream. The draw solution NaCl at different concentrations and velocity were applied for the studies. The two different membrane orientations, active layer facing feed solution and active layer facing draw solution were employed for the studies. The studies show that the cellulose triacetate with a cast nonwoven support membrane with active layer feed solution configuration could provide significant retention (nearly 97%) of cesium. Also, it was observed that the percentage retention of cesium and its flux were not affected by the concentration of draw solution NaCl.

### 5.2. Micellar enhanced ultrafiltration

Among the pressure driven membrane separation processes, UF has many advantages and finds wide applications in wastewater treatment. However, the heavy metal ions could not be retained by the UF membrane because of its pore size. Micellar enhanced ultrafiltration is an upgraded technique of the UF process, which utilizes the addition of surfactants to trap the metal ions [108]. This process involves the addition of surfactants to trap and create a large structure with metal ions which could be easily retained by the UF membrane. The surfactant is added more than the critical micellar concentration to achieve the separation of metal ions. When the concentration of surfactant exceeds

Table 2  
Applications of membrane separation processes for the separation of cesium and strontium from the aqueous stream

Target element	Source	Membrane separation process	Percentage retention/decontamination factor	References
$^{60}\text{Co}$ , $^{137}\text{Cs}$ , and $^{85}\text{Sr}$	Non-active and radioactive model solutions	Direct contact membrane distillation	1,400 800 600	[102]
Cs(I) Sr(II)	Simulated radioactive wastewater	Direct contact membrane distillation	100% 100%	[29]
Cs(I) Sr(II)	Simulated radioactive waste water	Nanofiltration	75.5% 99.2%	[100]
Cs(I)	Simulated radioactive wastewater	Forward osmosis	96.24%	[95]
Cs(I) Sr(II) Sr(II)	Simulated radioactive wastewater Nuclear laundry wastewater	Forward osmosis Metal oxide enhanced microfiltration	– >90%	[103] [104]
Cs(I) Sr(II)	Simulated non-active wastewater	Size enhanced ultrafiltration	99% 99%	[99]

the critical micellar concentration, it leads to the formation of large micelles. The metal ions bind with the micelle's surface and get trapped by it [109]. This enhances the separation of heavy metal ions by the UF membrane. For the separation of cationic heavy metal ions, anionic surfactants are mostly preferred because of the opposite charge. Some of the surfactants are expensive and it generates a huge volume of sludge. This process could be adopted for the separation of cesium and strontium from aqueous streams with suitable surfactants. The recovery of the surfactants could be possible by acidification, chelation, and precipitation techniques [110]. Significant results have been reported in the literature for the recovery of surfactants by acidification using sulphuric acid, nitric acid, and hydrochloric acid, and by chelation using iminodiacetic acid, ethylenediamine tetraacetic acid, citric acid, potassium ferricyanide, and potassium ferrocyanide, by precipitation techniques using potassium iodide, ferric and ferrocyanide [111–113]. However, it suffers from the limitations of sludge handling, treatment, and disposal problems. The volume of sludge generation could be minimized by the recovery of surfactants.

### 5.3. Size enhanced ultrafiltration

It is an emerging technology in membrane science and finds applications in the separation of heavy metal ions and radioactive wastes from the aqueous stream. This involves the addition of water-soluble size enhancing species such as polymer with the aqueous stream containing the contaminants, followed by the separation by ultrafiltration. The mechanism of separation by size enhanced ultrafiltration involves the complex formation which follows the principle of coordination chemistry. The schematic representation of complex formation is illustrated in Fig. 3. The polymer ligands make a complex with the target ions which becomes a macromolecular structure and it could be easily retained by the ultrafiltration membrane. Since, this process is highly dependent on the pH of the solution, by reversing the pH of the solution we could recover the value-added components. Unlike, other conventional separation processes, which suffer from the limitations such as non-recoverability, addition of a huge quantity of

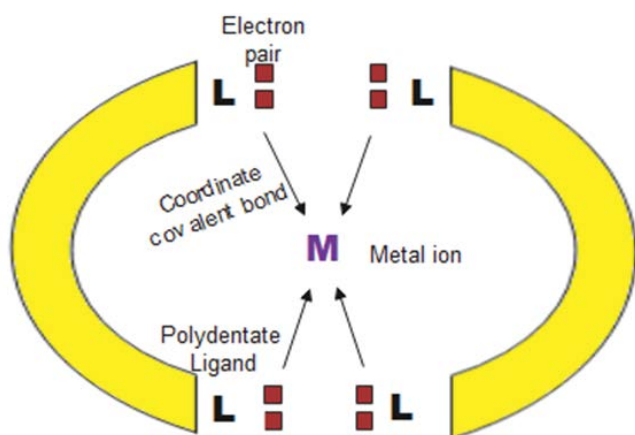


Fig. 3. Schematic representation of metal polyligand complex formation.

chemicals, sludge disposal problem, size enhanced ultrafiltration could remove and recover the target ions.

Our previous work was focused on the study of the removal of cesium and strontium from simulated wastewater by size enhanced ultrafiltration [99]. Plenty of synthetic polymers has been reported in the literature for the separation of radioactive wastes. However, those polymers are expensive, complex, and non-biodegradable. The study utilized the chitosan derivative, carboxymethyl chitosan as a size-enhancing species. Chitosan is a biopolymer that is cheap, abundantly available, non-toxic, and bio-degradable. There are only a few papers on the separation of cesium and strontium using chitosan derivatives as a size-enhancing species. The process parameters such as initial pH, initial concentration of cesium and strontium, and loading ratio (polymer/metal ion concentration) were analyzed and optimized using response surface methodology to yield maximum percentage retention and binding capacity. The interactive effect of process parameters on the percentage retention and binding capacity was analyzed through the ANOVA table. The study shows more than 99% retention of Cs(I) and Sr(II). The results show that the initial pH and initial concentration of Sr(II) have a significant impact on the percentage retention of Sr(II). Also, the initial pH, initial concentration, and loading ratio have a significant influence on the percentage retention of Cs(I). The significant results were observed at base pH for the separation of both Cs(I) and Sr(II). However, considerable percentage retention was obtained even at neutral pH, which in turn shows the viability of separation without much altering the pH of the effluent [99]. The promising results are shown in the literature for the separation of Cs(I) and Sr(II) by size enhanced ultrafiltration using chitosan derivative confirms the success of the process for the treatment of radioactive wastes.

### 5.4. Membrane distillation

Membrane distillation is a thermally driven process, the vapour pressure difference across the membrane acts as the driving force for the transfer of water vapour through the hydrophobic membrane [29,114–116]. The evaporation method was used in the nuclear power industries to treat the radioactive wastes. Since the process requires high energy and expensive cost of operation, it has been replaced by recently developed technologies. As an efficient alternative to traditional methods adopted for the treatment of radioactive wastes, membrane distillation could provide better retention without consuming much energy, and significantly economic operation for the treatment of low-level radioactive wastes. Several studies have been reported in the literature for the treatment of saline water and other industrial effluents by membrane distillation [101,102]. Various membrane configurations such as direct contact membrane distillation, air gap membrane distillation, sweep gas membrane distillation and vacuum membrane distillation have been adopted for the effluent treatment [117].

#### 5.4.1. Direct contact membrane distillation (DCMD)

In the DCMD process, there is a direct contact between the hot solution side and the membrane. Vapour from the



hot solution moves through the pores of the hydrophobic membrane due to the difference in the vapour pressure across the membrane. Then the vapour condenses on the cold solution side. DCMD process finds many applications such as desalination, separation of heavy metal ions, volatile organic compounds, the concentration of fruit juices in the food process industry, and also the treatment of radioactive wastes. Wen et al. [101] investigated the effect of the presence of inorganic salts in the separation of cobalt, cesium, strontium, and boron by using direct contact membrane distillation. It was observed from the results that the higher concentration of boron (5,000 mg/L) does not have any impact on the permeate flux, decontamination factor of nuclides. Also, the study showed the transportation of adsorbed boron through the membrane by the mechanism of adsorption–desorption [118]. The study proves the success of direct contact membrane distillation for the treatment of nuclides in the presence of boron from highly saline wastewater. Also, it could be concluded from the study that the volume of saline radioactive wastewater could be reduced and the cost of solidification and storage could also be minimized. Fig. 4 shows the schematic representation of direct contact membrane distillation for the separation of radioactive waste.

5.4.2. Vacuum membrane distillation (VMD)

In the VMD process, a vacuum is created in the permeate side of the membrane module. Vapour that permeates through the pores of the membrane is condensed outside the membrane module. This process finds applications in the separation of aqueous volatile solutions, separation of radionuclides from wastewater. Wang et al. [119] investigated the separation of cesium by vacuum distillation.

The percentage retention of Cs(I) was found to be more than 99%. The mass transfer rate was conquered by the collision of water vapour molecules and there was no significant impact of counter ions such as chlorides and nitrates on the retention of Cs(I). It was also observed from the results that the high salinity of water could affect the retention of Cs(I). Fig. 5 illustrates the schematic representation of the vacuum distillation process for the separation of radioactive waste.

The membrane distillation in the field of treatment of radioactive wastes is an emerging technology and successful results were obtained in lab-scale and pilot scale. It could be concluded that the further enhancement of the process by the development of advanced functional membranes such as superhydrophobic and oleophobic, and eradicating the fouling of membrane, could lead to the implementation of the process at the field level.

5.5. Reverse osmosis (RO)

RO is a well-known and well-proven technology in the field of desalination and water treatment. It also finds applications in the separation of organic and inorganic compounds [120–122]. It is an efficient technology since it offers high retention, minimum usage of chemicals, and less footprint area. The separation of radioactive wastes from the aqueous stream has been reported in the literature [7,18,123]. In the past few decades, many attempts have been made to separate cesium and strontium from low-level radioactive effluents [18,124]. Literature shows that there

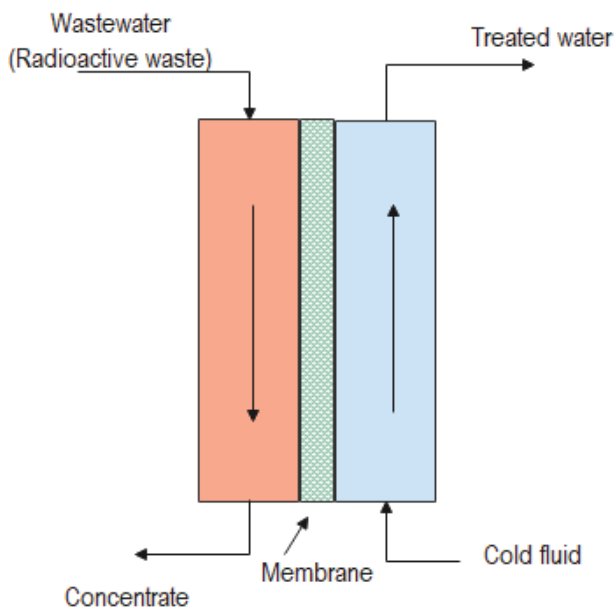


Fig. 4. Schematic representation of direct contact membrane distillation process for the separation of radioactive waste.

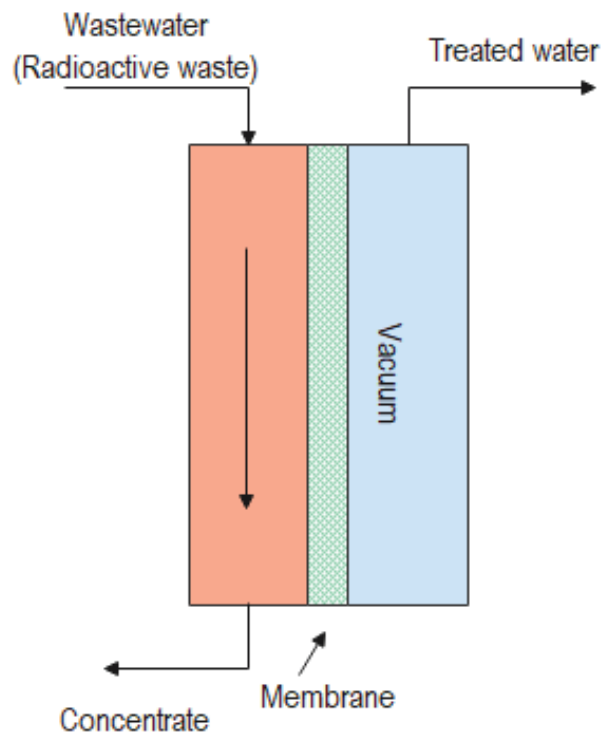


Fig. 5. Schematic representation of vacuum distillation process for the separation of radioactive waste.

is no influence of background compounds such as boron on the retention of cesium and strontium by RO [125–127].

Arnal et al. [126] investigated the performance of RO for the treatment of  $\text{Cs}^+$  from the aqueous waste. The plant consisted of prefiltration of different pore sizes such as 5 and 0.45  $\mu\text{m}$ . After pretreatment, the effluent was passed through the RO module of three stages operating at low pressure and high pressure. The percentage retention was found to be 98%. However, the permeate flux was not significant, and this could have been rectified by incorporating pretreatment stages such as microfiltration and/or ultrafiltration. Sasaki et al. [128] studied the separation of radioactive cesium and strontium from seawater by advanced RO technology. They investigated the percentage retention of cesium and strontium by advanced polyamide RO membrane and it was found to be nearly 100%.

Ding et al. [87] studied the separation of cesium and strontium of concentration in the range of 0.1–1 ppm from simulated wastewater at low pressure (<1 MPa) by polyamide RO membrane. The effect of pH, coexisting ions, and time on the percentage retention was studied. The percentage retention of cesium and strontium was almost 100% at pH 3 and 9 respectively. Due to the difference in the permeation of  $\text{NH}_4^+$ ,  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$ , the percentage retention of cesium was found to increase with the presence of  $\text{NH}_4^+$ , and decrease in the presence of  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$ . However, the percentage retention of strontium was found to increase in the presence of both monovalent and divalent ions. Also, the effect of the presence of anions on the percentage retention of cesium and strontium was studied. Since the hydrated ionic radius of cesium and strontium are different, it was observed from the results that the retention of cesium was controlled by the mechanism of Donnan's effect and strontium was controlled by the mechanism of electrostatic interaction.

Chen et al. [88] studied the separation of cobalt, cesium, and strontium from low-level radioactive wastes in the presence of silica and boron by RO, with the possible recovery of water with a higher concentration of boron and lower concentration of silica within the acceptable limit for reclaim. The studies were conducted at varying temperatures, pH, and pressure. The increase in temperature from 15°C to 35°C does not show any influence on the percentage retention of cesium and strontium. The percentage retention of cesium and strontium was found to be in the range of 96%–99% and 86%–95%, respectively. At pH 9.5, the percentage retention of cesium and strontium was found to be nearly 100%. At a pressure of 1.4 MPa, the percentage retention of cesium and strontium were found to be 92% and 100%, respectively and with an increase in the pressure, there was no significant increase in the percentage retention.

Labeled et al. [129] studied the separation of cesium and strontium from contaminated groundwater and seawater. The percentage retention was found to be more than 95% for both cesium and strontium from contaminated groundwater. There was no significant difference in the percentage retention of inactive cesium and radioactive cesium at trace level concentration. However, it was observed from the results that there was a slight drop in the retention of cesium with the scale-up in the system.

The application of RO could be an excellent choice for the treatment of radioactive wastes, which is also confirmed

by the studies reported in the literature. However, RO suffers certain limitations such as high-pressure operation, not being possible for the absolute separation of contaminants, and also fouling of the membrane.

#### 5.5.1. RO-MD hybrid membrane separation process

When we employ hybrid processes, we can make use of the benefit of both the processes and also the limitations of one process could be overcome by the other process. The hybrid processes have proven to be a promising technology for the treatment of low and intermediate-level radioactive wastes. The combination of RO and MD has been employed by Zakrzewska-Trznadel et al. [18] for the treatment of low and intermediate-level wastes. The hybrid process RO with two MD units is shown in Fig. 6 [18]. The radioactive waste is sent to the RO module. The permeate stream that comes out of the RO, which contains treated water with trace contaminants is sent to the MD unit where the trace level contaminants are further separated and pure water is obtained as distillate. The retentate from the RO, which contains the concentrated stream of contaminants is sent to the MD unit for further recovery of residual water in the concentrate. The flow diagram of RO-MD hybrid system is shown in Fig. 6. The combination of RO with MD could increase the process efficiency and also decontamination factor. Their study involved the three-stage RO module with two different configurations. The load capacity was calculated based on the operation for 200 d/y with the permeate flow rate of 20 and 4  $\text{m}^3/\text{d}$  respectively. Based on the lab-scale experiments with the MD unit, the pilot-scale was demonstrated with the capacity of 300–1,500  $\text{dm}^3/\text{h}$  of radioactive wastes and distillate flux of 10–6  $\text{dm}^3/\text{h}$ .

The separation of cesium and strontium by membrane separation processes greatly depends on the proper choice of the membrane material, membrane module, concentration of the radioactive wastes, and their activity. The separation studies reported in the literature exhibit the success of the technology to treat radioactive wastes. However, some of the studies are still on the pilot scale and full-fledged application for the industrial-scale needs to be implemented for the separation of radioactive wastes.

## 6. Conclusion

The existing separation processes in the treatment and management of radioactive contaminants have many challenges and are not a perfect solution to ensure environmental safety. Radioactive species have beneficial effects as well for society in medical treatment, food irradiation and long-term storage of food, sterilization of medical products, and domestic waste. All these are possible if at least from low-level wastes the long-lived fission products are separated and recovered so that they can be put to practical applications: a win-win situation both from resource recovery and environmental safety point of view. SEUF with suitable biopolymers such as chitosan derivatives could be an excellent technology for the separation of cesium and strontium. Membrane distillation is an emerging technology with vast scope in the separation processes. The recent developments in membrane technology have made them a more viable

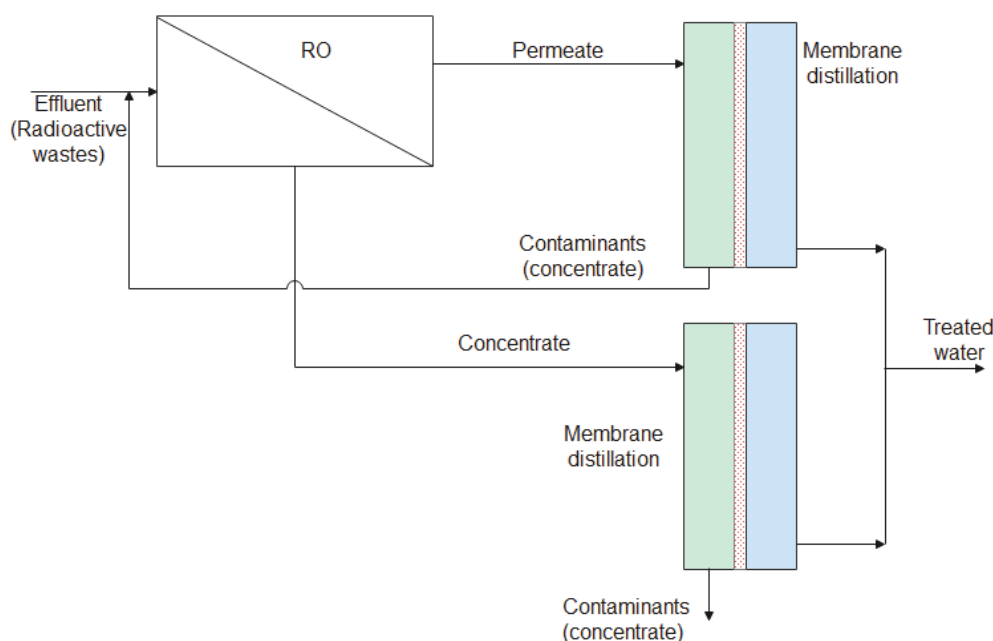


Fig. 6. Schematic representation of RO-MD hybrid system.

and also economical approach for the treatment of radioactive wastes. The advent of hybrid membrane systems and advanced functional membranes have eradicated the drawbacks of the membrane separation process and made them suitable for the application in the field of radioactive wastes. The possibility of coupling the membrane separation processes with other conventional processes could produce a substantial outcome and could be a better alternative to overcome the drawbacks of the conventional processes. In recent years, the application of nanocomposite membranes for separation studies has attracted attention, since it could overcome the fouling of membranes and enhance their functionality. This is an emerging field and there are a lot of scopes in the future. The process intensification with membrane processes could make it viable for large-scale applications.

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