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Review

Membrane purification in radioactive waste management: a short review

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ABSTRACT

Radiation hazards of radionuclides arising from nuclear plant facilities are well known. Separation technologies are used to concentrate the radionuclides and prevent the spread of this hazard to the environment. The present review describes the recent advances made in radioactive waste treatment using membrane separation technology. The first part discusses the membrane methods for collective separation of radionuclides and the second part discusses the membrane methods for selective separation of individual radionuclides. For the collection separation of radionulides, methods include reverse osmosis, precipitation followed by ultrafiltration or microfiltration and membrane distillation. Individual elements have been separated using liquid supported membranes, polymer inclusion membranes, solid polymer based electrolysis, nanofiltration, electrochemical salt-splitting process and other advanced separation methods.

1. Introduction

The main sources of radioactive waste are uranium and thorium mining and milling, nuclear fuel cycle operations (uranium conversion and enrichment, fuel fabrication and spent fuel reprocessing), operation of nuclear power plants, decontamination and decommissioning of nuclear facilities and institutional uses of radioisotopes (medicine, industry, agriculture, research reactors and test facilities) (Zakrzewska-Trznadel, 2006). To ensure the safe discharge into the environment, liquid radioactive waste has to fulfill very strict requirements connected with the limits of radioactive substances and other impurities (suspended particulates, biofoulants and organic or inorganic chemicals). To reach the standards described in national regulations, the waste has to be treated, including volume reduction and reduction of radioactive compounds and other solutes in the effluent. There are many methods used for liquid radioactive waste treatment, including chemical precipitation, sedimentation, ion exchange, thermal evaporation, biological methods (Ipek et al., 2002; IAEA TECDOC-1086, 1999; IAEA TECDOC-1336, 2003; IAEA TECDOC-1492, 2006) and membrane permeation. In addition to waste disposal, there is also a requirement for isolation and purification of radionuclides for specified applications.

Starting from the front end of the nuclear fuel cycle, i.e. mining stage, to the back end where radioactive wastes are processed for safe disposal, membrane processes have indicated good potential (Pabby et al., 2008). A membrane is a layer of material which serves as a selective barrier between two phases and remains impermeable to specific particles, molecules, or substances when exposed to the action of a driving force. Some components are allowed passage by the membrane into a permeate stream, whereas others are retained by it and accumulate in the retentate stream. Membranes can be of various thicknesses, with homogeneous or heterogeneous structure. Membranes can also be classified according to their pore diameter. According to International Union of Pure and Applied Chemistry (IUPAC), there are three different pore diameter (dp) size classifications: microporous ($dp < 2$ nm), mesoporous ($2 \text{ nm} < dp < 50$ nm) and macroporous ($dp > 50$ nm). Membranes can be neutral or charged, and particle transport can be active or passive. The latter can be facilitated by pressure, concentration, chemical or electrical gradients across the membrane (Pinnau and Freeman, 1999). The different membrane processes are summarized in Table 1. With advancing membrane technologies, purpose-made membranes to suit a particular separation have come to use. The schemes under which the technology is applied include collective colloid removal (concentration based separation) and specific element purification. The former involves removal of activity as a combination of radionuclides existing in the waste

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Table 1
Types of membrane processes (Zakrzewska-Trznadel et al., 2001).

Process	Reference	Type of membranes	Average pore size	Separation mechanism	Driving force
Reverse Osmosis (RO)	Leiknes, 2009; Zularisam et al., 2006	Thin film composite[a] polyamide (TFCP), cellulose acetate blend (CAB), cellulose triacetate (CTA)	5–10 Å	Preferential sorption-capillary flow	Net applied pressure in excess over the osmotic pressure
Nanofiltration (NF)	Szöke et al., 2005	TFCP	10–30 Å	Preferential sorption-capillary flow	Net applied pressure in excess over the osmotic pressure
Ultrafiltration (UF)	Chmielewski et al., 1999	Composite	30–100 Å	Mainly sieve	Net pressure
Microfiltration (MF)	Yong et al., 2004	Composite	Above 100 Å	Mainly sieve	Net pressure
Electro-dialysis (ED)	Inoue et al., 2004; Inoue, 2003; Inoue and Kagoshima, 2000	Composite	Nonporous	Electrically driven	Applied electromagnetic force (EMF)
Diffusion Dialysis (DD)	Mathur et al., 1998	Composite	Nonporous	Diffusion	Concentration gradient
Liquid Membranes (LM)	Bressot et al., 1996	Selective liquid barrier	Not applicable	Diffusion	Concentration gradient
Bipolar Electrolysis (BE)	Petek et al., 1981	Composite	Nonporous	Electrically driven	Applied EMF

[a] Composite implies membranes made from multi-layered materials.

stream. The latter involves separation of individual elements such as molybdenum, iodine, cesium, strontium, tritium, lanthanides and actinides. The reviews on applications of membranes in radioactive waste treatment are rather limited. The role of reverse osmosis (RO) and membrane distillation (MD) has been extensively discussed for decontamination of low level wastes (Zakrzewska-Trznadel et al., 2001; Potts et al., 1981). The applications of other membrane technologies for example supported membranes have not been reviewed before. The present review gives information on different recent application areas of membrane separation in treatment of radioactive wastes. The application of membranes for collective and selective radionuclide removal is also discussed here.

2. Collective ion and colloid removal

In purification strategies for collective radio-colloid removal, RO and MD have been extensively employed. The RO filtration method removes colloids from solutions by applying pressure to the solution when it is on one side of a selective membrane. The result is that the solute is retained on the pressurized side of the membrane and the pure solvent (usually water) is allowed to pass to the other side. A schematic of the RO process is provided in Fig. 1. To be "selective" this membrane should not allow large molecules or ions through the pores, but should allow smaller components of the solution (such as the solvent) to pass freely (Garrett, 1990). The membranes used for reverse osmosis have a dense barrier layer in the polymer matrix where most separation occurs. In most cases, the membrane is designed to allow only water to pass through this dense layer, while preventing the passage of solutes (such as salt ions). This process requires that a high pressure be exerted on the high concentration side of the membrane, usually 2–50 MPa. The principal beta-gamma emitters include elements such as Cs, Sr and Co while the principal alpha emitters include actinides. It is possible to achieve a discharge value of activity concentration for beta and gamma emitters lower than 10 kBq/m^3, while for alpha emitters lower than 1 kBq/m^3. The size variation is the principal reason for the difference in discharge values. Even though they are capable of withstanding a significant amount of radiation dose, synthetic polymeric RO membranes cannot handle acidic or alkaline solutions beyond the 4–9 pH range (Leiknes, 2009; Zularisam et al., 2006)

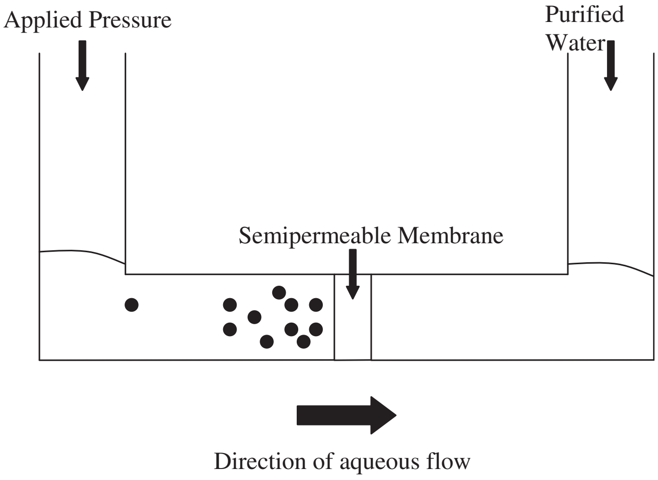

Fig. 1. Schematic of reverse osmosis [In a typical U-tube experiment liquid level is shown by the curved lines. The dotted side indicates impure water. Pressure is applied along the flow of impure water as it passes through semi-permeable membrane. The right side of membrane carries purified water.].

MD is based on the relative volatility of various components in the feed solution. The driving force for transport is the partial pressure difference across the membrane. Separation occurs when solvent vapor (usually water vapor) passes through the membrane pores by a convective or diffusive mechanism. The process can be described by the following steps (refer Fig. 2): water evaporation at the solution-membrane warm interface, transport of the water vapor through the microporous system, and condensation at the cold membrane-solution interface (Curcio and Drioli, 2005). The driving force for the vapor transport in this process is the vapor pressure difference between the two solution-membrane interfaces due to a temperature gradient. MD shares some characteristics with another membrane-based separation known as pervaporation (PV), but there are also some vital differences. Both methods involve direct contact of the membrane with a liquid feed and evaporation of the permeating components. However, while MD uses porous membranes, PV uses nonporous membranes. In case of nonporous membranes, instead of molecular weight or molecular size, the chemical nature and morphology of the polymeric membrane and the extent of interaction between the polymer and the permeants are the important factors to consider. Transport through nonporous membranes occurs by a solution-diffusion mechanism and separation is achieved either by differences in solubility and/or diffusivity. Hence such membranes cannot be characterized by the pore size and pore size distribution in the membranes. MD systems can be classified broadly into two categories: direct-contact distillation and gas-gap distillation. These terms refer to the permeate or condensing side of the membrane; in both cases the feed is in direct contact with the membrane. In direct-contact MD, both sides of the membrane

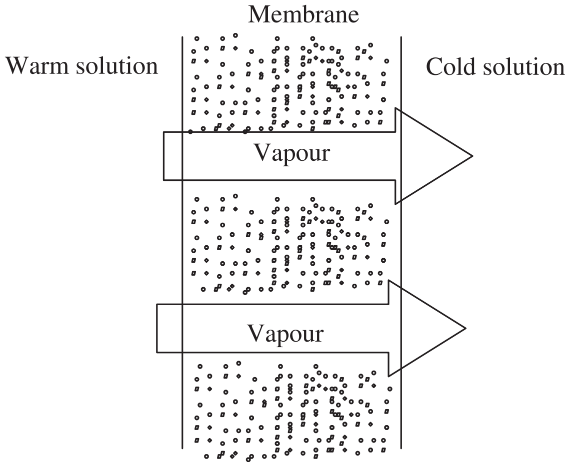

Fig. 2. Schematic of membrane distillation [The dots indicate the pores of the membrane. Vapor from warm solution side passes through pores of membranes and condenses on the cold solution side. For example volatile organic in aqueous medium can move to the cold side leaving the aqueous volume clean.].

contact a liquid phase; the liquid on the permeate side is used as the condensing medium for the vapors leaving the hot feed solution. In gas-gap MD, the condensed permeate is not in direct contact with the membrane. Potential advantages of MD over traditional evaporation processes include operation at ambient pressures and lower temperatures as well as ease of process scale-up. MD is a suitable option for liquid low and intermediate level radioactive waste purification. The application to high level liquid waste is limited owing to radiation instability of polymer based membranes. This category of waste is principally strongly acidic in nature and polymer based membranes are chemically unstable in this environment. In contrast to RO it does not involve high pressures; the process is not influenced by high concentrations of the feed solutions and ensures high rejection. The hydrophobic membranes show good resistance to the ionizing radiation and strong chemical environments. The salient features of the two techniques are tabulated in Table 2. The advantage of MD over RO is the possibility of running the process to the high solute concentration required for fossilization in concrete (up to 25% of solute), gaining the high concentration in a one-stage process, avoidance of the sorption of such ions as ^{60}Co^{2+}, ^{137}Cs^{+}, ^{134}Cs^{+} inside the membrane pores and less frequent washing cycles because of the elimination of fouling and sorption phenomena and minimization of secondary waste generation (Zakrzewska-Trznadel et al., 1999).

The membrane processes can also be combined with physical and chemical processes of binding small ions with macromolecular ligands or complexation known as seeded ultrafiltration. Microfiltration (MF) or ultrafiltration (UF) can then be employed as a pre-treatment stage for RO, and this pre-treatment is known as ‘seeded micro- or seeded ultrafiltration (SUF)’ (Chmielewski et al., 1999). As an example of SUF, laboratory simulation experiments were conducted to study the association of ^{239,240}Pu with pseudo-colloids of iron in groundwater samples having dissolved organic carbon (DOC) in the range of 10–60 mg/L (Singhal et al., 2009). Ultrafiltration membranes were used to partition colloidal components into seven different size ranges. Ultrafiltration membranes are rated according to the nominal molecular weight limit (NMWL). The NMWL indicates that most dissolved macromolecules with molecular weights higher than the NMWL will be retained. In the presence of DOC, Fe colloid size increased. In a particular size range of 1.1 nm (500 NMWL) and 1.6 nm (10,000 NMWL) with DOC in the range 10–60 mg/L, seven- to eight-times higher concentrations of Fe were observed. This size was observed as suitable for high sorption of plutonium. The DOC interaction with plutonium is due to combined influence of redox effect, site selectivity effect or coulombic effect. The importance of the study was that it could identify suitable parameters for ultrafiltration studies for removal of plutonium from waste.

Another term used in this category is flocculation microfiltration. The seeded ultrafiltration and flocculation microfiltration differ in size of membrane utilized (refer Table 1 for size cut-off). The low-level radioactive wastewater containing ^{241}Am was treated using a combined flocculation-microfiltration (FMF) process (Yong et al., 2004). The test was operated in 4 runs, and the membrane was cleaned by physical cleaning (discharging sludge and aerating for 20 h with no feed and no effluent) after each run. The pH was adjusted with nitric acid and 20 mg/L potassium permanganate was added to remove surfactants from the feed during pre-treatment. The pretreated wastewater was adjusted to above pH 8 and a dosage of Fe3+ was used at 30 mg/L. When the radioactivity of ^{241}Am in the feed was 809.2 Bq/L, the filtered effluent was below 1.0 Bq/L indicating that the removal was better than 99.9%. The decontamination factor (calculated as the ratio of activity concentration of feed to activity concentration of permeates) was with a high value of 1651.

Table 2
Comparison of reverse osmosis and membrane distillation methods (Potts et al., 1981).

Parameter	Reverse osmosis	Membrane distillation
State of art	Advanced, rapidly developed in many branches of industry	Modest, laboratory and pilot plant units, small industrial facilities. Not applied in nuclear industry.
Rejection rate for exclusion of solutes	Between 50 and 99.7%	≈100%
Retention of radioactive ions	Good, but multistage process is required	Complete
Productivity (values represent a 4 m^2 unit)	High: between 0.05 and 4 m^3/h	Low: 0.01–0.06 m^3/h. Several modules required to reach filtration output of RO
Temperature of operation	Ambient	>50 °C
Pressure	High: 40–50 MPa	Does not involve high pressures
Concentration	Strong influence of the concentration on the permeate flux	Weak influence
Capital and operation costs	Moderate and economical in comparison with conventional methods	High operational cost if cheap thermal energy sources are not available
Membranes	Composite	Hydrophobic, PTFE, PP, PVDF
Resistance to chemicals and temperature	Good for selected membranes	Very good
Resistance to radiation	Good for selected membranes	Good
Lifetime of the membranes	2–3 years in non active solution	>2–3 years
Modules	Commercial, low price	Limited number of manufactures, high price

2.1. Ceramic membranes

It has been discussed before that RO membrane has limitation of applicability in pH range. This statement was made with respect to polymer based membranes that have been extensively used for the application. Ceramic membranes are proposed as advantageous with respect to resistance toward fouling as compared to organic polymer based membranes. In this category, zeolite membranes have received considerable attention because of their superior characteristics of thermal, mechanical and chemical stability. Moreover, zeolite membranes have a great potential to separate mixtures of molecules by both adsorption and molecular sieving because of their unique pore systems (Kiyozumi et al., 2008; Kazemimoghadam and Mohammadi, 2007). Separation by PV and vapor permeation (VP) using ceramic membranes can offer an effective and an energy-saving alternative over conventional distillation processes, particularly for azeotrope or close-boiling liquid mixtures. Various types of supported polycrystalline zeolite membranes were developed on different substrates with minimized inter-crystal pores (Kiyozumi et al., 2008; Kazemimoghadam and Mohammadi, 2007). The general mechanisms of separation through ceramic membranes include molecular sieving and competitive adsorption and diffusion. Molecular dynamic simulation has shown that zeolite membranes are theoretically suitable for ion removal from aqueous solutions by RO. The simulation revealed that 100% Na^+ rejection could be achieved using RO through zeolite membranes (Kiyozumi et al., 2008); MFI being one commercial type. The separation mechanism of the perfect MFI zeolite membranes is the size exclusion of hydrated ions, which have kinetic sizes (0.8–1.0 nm for $(Na(H_2O)_x)^+$) significantly larger than the aperture of the MFI zeolite (diameter 0.51 nm). The sizes of hydrated ions are presented in Table 3. Results of the simulation and experimental RO separation of water/ethanol mixtures using zeolite membranes indicated that it may be possible for zeolite membranes to separate ions and dissolved organic compounds simultaneously from aqueous solutions using RO (Kiyozumi et al., 2008). Another zeolite based membrane; Zeolite NaA membranes (NaA being the commercial name) were prepared by hydrothermal synthesis on a porous α-alumina support with secondary growth crystallization and examined for low-level radioactive solution decontamination by a RO process (Malekpour et al., 2008). For ionic solutions of 0.001 M Cs^+, Sr^{2+} and MoO_4^{2-} more than 99% rejection factors were obtained. Another membrane in this category includes hydroxyl sodalite (Khajavi et al., 2010). This membrane performance was observed as superior to zeolite membranes at elevated temperatures. Sodium salt rejection of ~99.99% was observed in the temperature range 303–473 K and pressure 22 MPa.

3. Selective radionuclide removal

For the separation of specific elements, the membranes are modified for different mechanisms of treatment. These can be primarily classified as ion-selective membranes, liquid membranes and ion-exchanger-based membranes. Liquid membranes can be further classified as supported liquid membranes and polymer inclusion membranes.

Table 3
Size of hydrated ions.

Ion	Hydrated diameter (Å)
H_2O	5.6
Li^+	7.6
Na^+	7.2
K^+	6.6
Mg^{2+}	8.6
Ca^{2+}	8.2
OH^-	6.0
Cl^-	6.6
NO_3^-	6.8

Membrane cells find application in electrochemical methods for treating waste components (Dziewinski et al., 1998). In this method, the salt is electrochemically split and allowed to pass through a cation selective or anion selective membrane so that the final solution is free of the cation or anion (Balagopal et al., 1999). The advantage of electrochemical systems is that the treatment of various wastes, or streams containing selected waste components, can be accomplished in a single treatment unit. A number of ion-selective membranes have been developed for industrial electrochemical processes (Pungor, 1987; Chaudhri and Cheng, 1980).

Supported liquid membranes (SLMs) combine extraction and stripping into one step, these are generally carried out in two separate steps in conventional processes such as solvent extraction (Ho, 2003). Separation of metal ions by means of supported liquid membranes consists of three processes, i.e., solvent extraction of metal ions at the interface between a feed solution and a membrane, transport of the extracted species to the opposite side of the membrane, and back-extraction at the other side of the interface between the membrane and a receiving solution (Jonsson and Mathiasson, 2001). A one-step liquid membrane process provides the maximum driving force for the separation of a target species. In SLMs, the liquid membrane phase is the organic liquid embedded in pores of a microporous support, e.g., microporous polypropylene hollow fibers. When the organic liquid contacts the microporous support, it readily wets the pores of the support, and the SLM is formed. For the extraction of a target species from an aqueous feed solution, the organic-based SLM is placed between two aqueous solutions, the feed solution and the strip solution, where the SLM acts as a semipermeable membrane for the transport of the target species from the feed solution to the strip solution. The organic component in the SLM is immiscible in the aqueous feed and strip streams and contains an extractant, a diluent that is generally an inert organic solvent and sometimes a modifier. For SLMs, facilitated transport is the mass transfer mechanism for the target species to go from the feed solution to the strip solution.

Polymer inclusion membranes (PIM) contain macrocyclic units with the ability to entrap specific metal ion (Fontàsa et al., 2007; Scamporrino and Vitalini, 1992). These are cellulose triacetate based membranes with inclusion carriers that have specificity toward radionuclide. The membrane type, PIM or polymeric plasticizer membrane (PPM) (Sugiura et al., 1989) is claimed to combine the virtue of rapid transport with high selectivity and ease of set up and operation, exhibiting at the same time excellent durability. Because this type of membrane is independent of organic solvents due to the fact that the carrier molecules are trapped within the cellulose triacetate matrix, they do not suffer from loss of organic solvents nor as much leaching of carrier into the aqueous phases. The PIM entraps the macrocyclic carriers in the membrane matrix during the casting process, effectively inhibiting carrier loss to adjacent aqueous phases.

As with supported liquid membranes, membrane-based ion exchangers also find wide range of applicability (Oji et al., 2009). Titanate based ion-exchangers have been studied extensively in this regard. The unique properties of titanates as ion exchange materials lies in the ability of this class of materials to function in a wide range of pH conditions without degradation. Titanate ion-exchange materials also perform well in the presence of competing ions such as sodium, calcium and potassium, which are always present in caustic nuclear waste streams. Thus titanate based ion-exchange materials are able to enhance the decontamination of majority radionuclides present in nuclear wastes at part per million levels. In a membrane-based ion-exchange titanate sorbent, the leaching of titanate components would be minimized because the solution would not be in direct contact with the ion-exchange particles alone, but rather with both the ion-exchange (IX) particles and the membrane support fibrils. The silcotitanate based membrane sheets were used to evaluate the removal of surrogate radioactive materials for ^{137}Cs and ^{90}Sr from high caustic nuclear waste stimulants (Oji et al., 2009). The membrane supports met the nominal requirement for non-chemical interaction with the embedded ion-exchange materials and were porous enough to allow sufficient liquid flow. The titanium impregnated ion-exchange membrane discs removed more than 96% of dissolved ^{137}Cs and ^{90}Sr from caustic nuclear waste salt stimulants. Ceramic membranes combined with complexation were applied for hazardous wastes containing radioactive substance processing (Zakrzewska-Trznadel and Harasimowicz, 2004). The membrane method combined with such complexing agents such as soluble chelating polymers and cyanoferrates of transient metals were used to bind radioactive ions and to enlarge the separated molecule size (Yong et al., 2004).

4. Separation of specific elements

The radioactive wastes generated in the nuclear fuel cycle activities are of various types. Each type of waste requires specific treatment depending upon the type and level of the radioactivity as well as the chemical nature and the physical state of the waste. High Level Waste (HLW) is one of the most hazardous wastes produced in the nuclear industry, as it accounts for nearly 99% of total radioactivity released in various waste streams in a nuclear fuel cycle. Spent fuel contains valuable nuclear materials such as uranium and plutonium (formed by neutron irradiation of ^{238}U), which are recovered through reprocessing. The PUREX process is the most common aqueous reprocessing method and is based on a solvent extraction technique (Benedict et al., 1981). It uses tri-n-butyl phosphate (TBP) dissolved in a hydrocarbon diluent such as n-dodecane as extractant. The spent fuel is dissolved in nitric acid and the uranium and plutonium are then extracted into TBP and are stripped using suitable strippants. The highly radioactive waste raffinate remaining after the recovery of uranium and plutonium contains various fission products as well as significant quantities of actinides such as neptunium, americium and curium which are poorly extracted in TBP. In addition, small fractions of uranium and plutonium, which remain unextracted during the processing, are also present in this waste solution. The raffinate containing all these radioactive nuclides is concentrated by evaporation into highly radioactive liquid waste (HLW). This waste is a rich source of valuable radioisotopes which can be isolated and purified for specified applications. Membrane technology has been successfully demonstrated for the isolation and purification of specific radionuclides, finding scope in wide range of applications.

4.1. Molybdenum

Molybdenum-99 is a parent radioisotope to the daughter radioisotope ^{99m}Tc, which is used in many medical procedures, especially for imaging. Technetium can easily be separated from Mo using an ion exchange method.

Aliquat 336 was used as a carrier, diluent and emulsifying agent for the concentration of Mo on a liquid emulsion membrane (refer Table 1). Stripping was followed with NaOH and dodecane (Kulkarni and Mahajani, 2002).

4.2. Cesium

Cesium finds application as an irradiation source for various medical and non-medical applications. An example of membrane usage is the accident in June 1998 when a ^{137}Cs source was accidentally melted in the furnace of a stainless steel production company in Spain (Arnal et al., 2003a, b). As a result of this incident, the furnace and its cooling circuit were radioactively contaminated. Cs from contaminated ashes was removed to the extent of 98% using the RO method.

The removal of ^{137}Cs from nitric acid feed was investigated using plasticized polymer inclusion membrane (PIM) containing several crown ether carriers: di-benzo-18-crown-6 (DB18C6), di-benzo-21-crown-7 (DB21C7) and di-*tert*-butylbenzo-18-crown-6 (DTBB18C6) (Mohapatra et al., 2009). The PIM was prepared from cellulose triacetate (CTA) with various crown ethers and plasticizers. Both DTBB18C6 and tri-*n*-butyl phosphate (TBP) were found to give higher transport rates for ^{137}Cs as compared to other carriers and plasticizers. Although TBP plasticized membranes showed good transport efficiency, it displayed poor selectivities. On the other hand, an entirely opposite separation behavior was observed with 2-nitrophenyloctylether (NPOE) plasticized membranes suggesting the possible application of the latter membranes for the removal of bulk ^{137}Cs from the nuclear waste. The plasticized polymeric membrane made from CTA, DTBB18C6 and TBP was efficient in transporting Cs from acidic solutions suggesting the applicability of this method as a viable option for radioactive waste remediation. The stability of the membrane was good only for the feed at 1 M HNO_3; suggesting that the application of this membrane can be done for pre-concentration of radiocesium from waste solutions of low acidity.

Facilitated transport of Cs from aqueous nitrate feed solutions to a receiver solution containing distilled water through a supported liquid membrane containing calix(4)-bis-2,3-naphtho-crown-6 (CNC) in 0.45 μm PTFE (polytetrafluoroethylene) membranes was investigated (Raut et al., 2008). The carrier solution usually consisted of CNC dissolved in a mixture of 80% 2-nitrophenyl octyl ether and 20% *n*-dodecane. The transport rates were found to be influenced by the mobile carrier concentration, cesium concentration as well as the feed acidity. The extracted species confirmed to a stoichiometry of 1:1 metal to ligand ratio suggesting the extraction of an organophilic mono-Cs-crown complex.

4.3. Tritium

Tritium is released by power reactors and is needed in fusion reactors.

Solid-polymer-electrolyte (SPE) water electrolysis using poly-perfluorosulfonic acid (PFSA) membrane such as Nafions (du Pont de Nemours & Co., Inc.) has been attractive in the electrolytic processing of tritiated water, since the SPE is a promising method for large-scale and high efficiency hydrogen generation (Iwai et al., 2010). The PFSA membrane consists of a polytetrafluoroethylene (PTFE) backbone with perfluoroalkylether (PFAE) side-chains that terminate in a -SO_3—M^+ group, where M^+ is an exchangeable cation.

A 2-stage Pd membrane reactor (PMR) is also capable of recovering tritium m (Birdsell and Willms, 1998; Le Digabel et al., 2002). The catalyst releases tritium in gaseous phase. This method of use of gas separation membranes in tritium removal systems was demonstrated to recover gram levels of tritium from liter-scale volumes of aqueous medium.

Using multiple bipolar electrolytic separation of hydrogen isotopes with Pd-25%Ag electrodes, the feasibility of tritium separation from light and heavy water has been demonstrated (Petek et al., 1981).

4.4. Cobalt

^{60}Co is used in the sterilization of medical supplies and medical waste, radiation treatment of foods for sterilization (cold pasteurization), industrial radiography (e.g., weld integrity radiographs), density measurements (e.g., concrete density measurements), and tank fill height switches. ^{57}Co is used as a source in Mössbauer spectroscopy and is one of several possible sources in XRF.

The selective removal of Co species from simulated nuclear liquid waste was investigated with different nanofiltration (NF) membranes (refer Table 1) at various solution pH levels, initial Co concentrations, and background ion concentrations (Ho Choo et al., 2002). Substantial Co rejection by NF was achieved along with partial separation of monovalent ionic species, although it depended on the level of liquid pH and the presence of background species. Greater Co rejection at increased pH was attributed to the precipitation of $CoCO_3(s)$ associated with natural carbonates originating from atmospheric CO_2 gas. The decrease of Co rejection with the addition of boric acid was found to occur due to the formation of complexes between Co and boric acid. Certain nuclear wastes carry ^{60}Co in an EDTA complex (Szöke et al., 2005). Nanofiltration again helped in the removal of Co^{3+} EDTA complex from a drain wastewater model solution, which contained mainly $NaBO_3$ at an alkaline pH. The NF experiments were performed at a constant temperature (25 °C) and pressure range 1–10 MPa. The Co complex and the borate ion rejection (R) as well as the permeate flux of the membrane was investigated as a function of pH. The rejection of the Co^{3+} EDTA complex ion and especially the borate were strongly pH dependent. The rejection of the complex ion and the borate was increased at alkaline pH (at pH 8, $R = 73\%$; at pH 11.5, $R = 96\%$ for the Co complex; at pH 8, $R = 7\%$; at pH 11.5, $R = 59\%$ for borate). The removal of the Co complex ion from an alkaline borate solution by NF is possible in two ways: at slightly alkaline pH by a two-step separation, or at a more alkaline pH (pH $>$ 9) by a one step separation.5.

4.5. Sodium

Sodium has been used in different processing stages in the nuclear industry. It does not have radiological concerns, however; the recycling of sodium is important owing to discharge constraints. As per United States Environmental Protection Agency Guidelines, Federal Water Pollution Control Act, a typical limit value is 200 mg/L.

Sodium Super Ion Conductor (NaSICON) was studied at Pacific Northwest National Laboratory to investigate its ability to separate Na from radioactively contaminated sodium salt solutions (Fountain et al., 2008). Ceramatec Inc. developed and fabricated a membrane disk containing a proprietary NASGY material formulation. A 19 M NaOH feedstock product for recycle into waste treatment processes such as sludge leaching, regenerating ion exchange resins, inhibiting corrosion in carbon-steel tanks, or retrieving tank wastes was obtained using this membrane. In actual waste tests, average Na transport rates of 10.3 kg/day/m^2 were achieved at average Na transport efficiencies of 99%. The membrane was found to be highly selective to Na ions resulting in no detectable cation transport except Na and a small quantity (0.04%–0.06%) of ^{137}Cs. An average decontamination factor of 2000 was observed with respect to ^{137}Cs.

An electrochemical salt-splitting process, based on Na^+ selective ceramic membranes was also developed to recover and recycle NaOH from high-salt radioactive tank wastes (Kurath et al., 1997). The ceramic membranes chosen were Na-selective super-ionic conductors (NaSICON) based on the rare-earth elements Nd and Dy. The membranes were incorporated into a polyethylene scaffold for implementation into commercially available plate-and-frame electrochemical cells. A purified caustic product with a NaOH in excess of 3 M was produced from waste stimulants with the Dy- and Nd-NaSICON membranes. Membrane fouling was not observed, even though gibbsite was precipitated in large amounts during some of the runs. Preliminary testing of the NAS-D material indicated that a Na current density of 38 mA/cm^2 with a Na current efficiency of approximately 90% was achievable over 1000 h of operation with an applied potential of 4.5 V. Of the two membranes, Dy-NaSICON showed limited lifetime in the very alkaline solutions. NafionA Type 324 or 350 and an inorganic-based membrane, CeramatecA NASD were also reported to produce NaOH depleted waste stream (Hobbs, 1999).

4.6. Iodine

Iodine is used in nuclear medicine therapeutically and can also been seen with diagnostic scanners if it has been used therapeutically. Major uses of ^{131}I include the treatment of thyrotoxicosis and some types of thyroid cancer that absorb iodine. The ^{131}I isotope is also used as a radioactive label for certain radiopharmaceuticals that can be used for therapy. The more purely gamma-emitting radioiodine ^{123}I is used in diagnostic testing (nuclear medicine scan of the thyroid). The longer half-lived ^{125}I is also occasionally used when a longer half-life radioiodine is needed for diagnosis, and in brachytherapy treatment.

Paper membranes bearing trimethyl hydroxyl propyl ammonium anion exchange groups or methyl iodide-hexane with higher permselectivity for iodine were used for the separation of Na^{125}I and Na^{36}Cl (Inoue et al., 2004; Inoue, 2003; Inoue and Kagoshima, 2000). Removal of iodine from radio-immune analysis waste has also been carried using membranes (Arnal et al., 2000).

4.7. Strontium

^{90}Sr is extensively used in medicine and industry, as a radioactive source for thickness gauges and for superficial radiotherapy of some cancers. Controlled amounts of ^{90}Sr and ^{89}Sr can be used in treatment of bone cancer. As the radioactive decay of ^{90}Sr generates significant amount of heat, and is cheaper than the alternative ^{238}Pu, it is used as a heat source in many Russian/Soviet radioisotope thermoelectric generators, usually in the form of SrF_2. It is also used as a radioactive tracer in medicine and agriculture.

For the removal of ^{90}Sr from strong alkaline solutions in the presence of 1 M NaOH and 3 M $NaNO_3$, a method based on Di-2-ethyl hexyl phosphoric acid (D2EHPA) acting as a carrier in liquid membrane for simultaneous extraction-reextraction was adopted by Kocherginsky et al. (2002). After removal, Sr was precipitated as $SrSO_4$, using H_2SO_4 as the stripping phase, thus giving the possibility to concentrate radioactive Sr in a small volume of solid phase within one technological step. Neither work showed that the SLMs removed radioactive ^{90}Sr to the target of 8 pCi/L or lower from feed solutions of 300–1000 pCi/L using D2EHPA (Ho and Wang, 2002).

A combination of biomass treatment, fluidized bed/membrane reactor, and a minimum-suspension fluidized bed reactor was proposed to remove Sr cations from aqueous solutions (Chaalal and Islam, 2001). The adsorbents were used in series in a packed bed (with algae), fluidized bed/membrane reactor, and a minimum suspension fluidized bed reactor. The concentration was reduced from 100 mg/L to less than 10 mg/L. In order to reduce the low-level of Sr concentration even further, bio-encapsulation using thermophilic bacteria and membrane separation was used.

4.8. Lanthanides

Nuclear transmutation is the conversion of one chemical element or isotope into another, which occurs through nuclear reactions. It has been proposed that instead of shifting radioactive waste to underground repository, the long lived radionuclides can be transmuted into short lived ones. For example actinides can be fissioned to lighter elements using slow neutrons. In the category of mixed wastes, lanthanides serve as radioactive poisons because they absorb neutrons and slow the fission chain. It is therefore desirable to remove them before sending waste for transmutation.

In order to examine the feasibility of treating LLW wastewater by SLMs, experiments were performed using a stirred permeation cell (Teramoto et al., 1999). Aqueous solutions of HNO_3 and/or $NaNO_3$ containing about 550 mg/L Ce^{3+}, 490 mg/L Fe^{3+}, 320 mg/L Cr^{3+} and 330 mg/L Ca^{2+} were used as a simulated low-level radioactive wastewater. The liquid membrane consisted of octyl (phenyl)-*N*,*N*-diisobutyl carbamoyl methyl phosphine oxide as a carrier of Ce, tributyl phosphate (TBP) as a modifier and dodecane as a solvent. The strip solutions were water and aqueous sodium citrate solutions. Rapid transport and enrichment of Ce were realized by adding sodium citrate to the strip solution as the masking agent of Ce and also using a feed solution of low nitric acid concentration and high sodium nitrate concentration. Increasing the temperature from 298 to 318 K was very effective in enhancing the permeation rate of Ce. The membrane area required for removing 99.9% of Ce from the feed solution containing about 550 ppm Ce was estimated as 3.3 m^2 at 1 m^3 day^{-1} of the treatment rate.

In another study on SLM, aqueous Ce^{3+} solutions containing sodium nitrate and nitric acid were used as the simulated low-level radioactive wastewater (Teramoto et al., 2000). The SLM consisted of octyl(phenyl)-*N*,*N*-diisobutyl carbamoyl methyl phosphine oxide (CMPO) as a carrier of Ce^{3+}, TBP as a modifier and dodecane as a solvent. The strip solutions were aqueous solutions of chelating agents such as trisodium citrate and disodium hydrogen citrate. A small plate-and-frame type SLM module was used and circulation mode for both the feed and the strip solutions was adopted. The SLM was stabilized by adding an organic membrane solution to the reservoir of the strip solution and by circulating the membrane solution through the strip side of the SLM module so that the SLM could frequently contact the membrane solution dispersed as droplets in the strip solution. By this method, the SLM was not degraded by repeated replacements of both the feed and the strip solutions.

In-situ electrooxidation of Ce^{3+} to extractable Ce^{4+} and its transport across bulk liquid membrane (BLM) composed of TBP/dodecane mixtures was studied under varied hydrodynamical and chemical conditions (Kedari et al., 1999). More than 90% permeation of Ce with a maximum flux of 8.63 E-5 mol/m^2/s could be accomplished under the experimental conditions: stirring rates at feed and strip solutions were 380 and 300 rpm, respectively; feed was 1 M of HNO_3 containing 5 mM $Ce(NO_3)_3$; LM contained 30% TBP/dodecane; and the receiving phase was distilled water. Radiochemically pure ^{144}Ce was partitioned from the Ce-Am mixture obtained by extraction chromatographic fractioning of high level radioactive waste. This also resulted in the purification of ^{241}Am in the feed solution with a decontamination factor of 112 from Ce.

Polymer Inclusion Membranes (PIM) consisting of cellulose triacetate (CTA) plasticized by an organic solvent, 2-nitrophenyl *n*-octyl ether (NPOE) containing carriers such as CMPO or *N*,*N*,*N*,*N*-tetraoctyl-3-oxapentane diamide (TODGA) were found as very effective to transport Ce^{3+} ions from the feed phase to the stripping phase (Kusumocahyo et al., 2006).

A study was carried out on the transport of Nd from nitric acid solution, using a flat sheet contained liquid membrane technique, in order to investigate the feasibility of treating low-level radioactive wastewaters using hollow fiber membrane contactors (Soldenhoff and McCulloch, 2002). The solvent consisted of CMPO as the active carrier in n-dodecane. The effect of various modifiers was explored and a mixture of TBP and isotridecanol was identified as the most effective modifier, minimizing acid coextraction and preventing third phase formation.

The permeation of rare earth elements through a bis(2-ethylhexyl) hydrogen phosphate-decalin membrane supported on a microporous polytetrafluoroethylene sheet was studied using a multitracer containing radioactive nuclides of Sc, Zr, Nb, Hf, Ce, Pm, Gd, Yb, and Lu in 0.5 N HCl (Ambe et al., 1998). The feed solution at pH 1.4 gave the highest permeation rate for Ce, Pm, and Gd, amounting to about 95% of permeation for Ce and Pm, 80% for Gd, and 10% for Yb in 21 h. Sc, Zr, Nb, Hf and Lu were not transported at all from the feed solution. Permeation rates of Yb and Lu from the feed solution at pH 1.4 to receiving solutions of 0.75, 1.0, 2.3, and 4.0 M HC1 increased with the concentration. The results obtained indicate that the light rare earth elements can also be selectively separated from the heavy ones using membrane technology in the absence of any chelating agents or salts in the feed and receiving solutions.

The method of using liquid emulsion membrane (LEM) featuring the cation carrier bis-(2-ethylhexyl) phosphoric acid in cyclohexane for the recovery of Sm^{3+} from nitrate medium was investigated (El Reefy et al., 2009). Span-80 was used as a surfactant and HNO_3 as a stripping solution. Permeation of Nd^{3+} from nitrate medium was investigated using liquid emulsion membrane (LEM) containing, thenoyltrifluoroacetone (HTTA) and trioctylphosphine oxide (TOPO) in cyclohexane as synergistic mobile carriers, span 80 as a surfactant and HNO_3 as a stripping solution.

4.9. Actinides

Actinides are the sources of fertile and fissile fuels for nuclear reactors. Their separation is also therefore for a significant application.

The extraction and stripping of uranium ions from nitrate media using a hollow fiber liquid membrane contactor was studied using TBP diluted in kerosene as extractant and NaOH as a stripping solution (Ramakul et al., 2007).

Water soluble chelating polymers such as polyethyleneimine polymer that incorporates N-methylhydroxamic acid chelating groups for the selective separation of trivalent cations such as Fe^{3+} and tetravalent actinide ions such as Th^{4+} or Pu^{4+} from aqueous, radioactive waste streams were developed (Bisset et al., 2003).

5. Conclusions and scope for future

The data summarized in this paper suggest that membranes in radioactive waste management practices have a wide scope. The collective removal of elements has been adopted on industrial scale; however the majority of data on individual element separation is from laboratory scale experiments. This suggests that more work is required to mode to operational-scale adaptation. The application of nanoparticle embedded membranes is emerging as a new application in water purification (Kim and Bruggen, 2010). The use of nanoparticles in the manufacturing of membranes allows for both a high degree of control over membrane fouling and the ability to produce desired structure as well as their functionalities. These have been applied for both sorption and catalytic degradation applications. There are no reports on these for radioactive waste purification and hence this is an important emerging area for scope of study.

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