

Review

## Recovery of rare earth elements from waste streams using membrane processes: An overview

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### ARTICLE INFO

#### Keywords:

Rare earth elements  
Resource recovery  
Membrane filtration  
Nanofiltration

### ABSTRACT

Recovery of rare earth elements (REEs) has become an attractive process owing to the high costs and limited availabilities of REEs. In addition, the recovery of REEs has become a necessity because of their high toxicity and negative environmental impacts. Conventional processes for the recovery of REEs from waste streams include chemical precipitation, coagulation, flocculation, flotation, ion exchange, adsorption, and electrochemical processes. These processes have various disadvantages such as high consumption of chemicals, high operational costs, and low purity of the extracted elements. Recently, membrane processes have been used for the recovery of REEs. Compared to conventional methods, membrane processes have higher selectivity for REEs and lower energy requirements; moreover, they offer the possibility of zero liquid discharge. Nevertheless, it is still important to evaluate the economic feasibility of using membrane techniques for the recovery of REEs. This review paper provides an overview of the application of membrane techniques for the recovery of REEs from waste streams. Rejection efficiency of several membrane processes and purity of the extracted REEs were analyzed. Economic feasibility of using membrane processes for the recovery of REEs was examined by comparing the capital and operational costs of the processes with the selling prices of REEs.

### 1. Introduction

Rare earth elements (REEs) are a set of seventeen elements, namely fifteen lanthanides and two d-transition metals (Sc and Y), with similar metallic characteristics (Dushyantha et al., 2020). They are characterized by their metallurgical, optical, and electronic properties, which promote their applications in the manufacture of various sophisticated devices such as smartphones, digital versatile discs, batteries, and computer memories. Recently, the demand for REEs has significantly increased, which has caused an increase in their prices (Pereao et al., 2018). Owing to the increased demand for REEs, adverse environmental impacts associated with the production of REEs have become a global concern. Therefore, several studies have focused on the development of sustainable and economic methods for the recovery of REEs from various sources including discarded electronic devices and diverse waste streams (Li et al., 2019; Pereao et al., 2018).

Hydrometallurgy is a well-developed process for the extraction of REEs from REE-carrying substances such as natural ores and other secondary sources (Jha et al., 2016). It involves two stages: leaching and

separation (Zhang et al., 2020). In the leaching stage, REEs are detached from the REE-carrying substances using chemical reagents in an aqueous solution. Then, different separation processes are employed to recover REEs from the aqueous solution. The recovery of REEs from waste streams or the aqueous solution obtained during hydrometallurgy is generally performed by several conventional techniques including precipitation, electrocoagulation, flotation, solvent extraction (SE), ion exchange, and adsorption (Heckley and Ibana, 2003; Pereao et al., 2018). Precipitation is a common method for the recovery of REEs from wastewater; however, it is considered an unfeasible process, specifically for effluents with low REE concentrations, owing to the high volumes of chemicals required and sludge generated (Hua et al., 2012; Zhou et al., 2018). Electrocoagulation exhibits a high recovery efficiency for REEs; nevertheless, the main drawback of using this process is the high operational cost and the short life cycle of the used anode (Mollah et al., 2001). Flotation is a cost-effective process; however, a post treatment is required to achieve REEs with the desired purity (Van Nguyen et al., 2016). Although SE is also used for the recovery of REEs from waste streams, the primary disadvantage of this process is the low purity of the

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<https://doi.org/10.1016/j.hydromet.2021.105706>

Received 2 February 2021; Received in revised form 3 July 2021; Accepted 4 July 2021

Available online 7 July 2021

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extracted REEs because other metals may be extracted along with the required elements (Su et al., 2020; Tian et al., 2013). Ion exchange using chelating resins can also be employed for the recovery of REEs from waste streams. It is based on the affinity of metal ions for specific resins used to recover REEs (Jain et al., 2007). The effectiveness of ion exchange is limited by the high buffering capacity of this process and the presence of other particles in the influent containing REEs. The existence of clay particles, humic acids, and other metal (such as Fe, Ca, and other heavy metal) ions can adversely affect the efficiency of ion exchange. Adsorption is an economical and suitable technique for the recovery of REEs from waste streams. However, adsorption produces sludge that requires further post-treatment, and the presence of other ions in the waste stream can influence the effectiveness of extraction (Galhoum et al., 2017; Mahmood et al., 2012). To date, numerous state-of-the-art studies have been reported on the development of methods for recycling REEs from waste streams; nevertheless, further development and investigation of most of these techniques are needed before their commercial application (Balaram, 2019; Filippas et al., 2021; Omodara et al., 2019). Algae have also been used for the recovery of REEs from wastewater (Cao et al., 2021). Gueroult et al. (2018) examined the possibility of using plasma separation techniques for recycling REEs. The performances of plasma separation techniques were better than those of the existing recycling methods owing to their lower energy consumption. Yin et al. (2018) prepared a mixture of sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) and hydrogen peroxide for the oxidative leaching of REEs from waste cathode-ray tube screens. Cyanex 272 extractant and a green solvent ionic liquid (1-octyl-3-methylimidazolium [OMIm]) were employed for the separation of REEs. Research related to the recovery of REEs from waste streams using various techniques has attracted extensive attention (Fig. 1). The number of publications on this topic was less than 100 between 2010 and 2013; it rapidly increased and exceeded 300 in 2020.

The use of membrane separation processes for the recovery of REEs from waste effluents is promising due to the several advantages afforded by these techniques, for example, high recovery, and selectivity, small space requirements, and minimal sludge volumes. Different types of membranes, including microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) membranes, are employed in these processes. The main drawbacks of membrane techniques are membrane fouling and the high cost of membranes. To the best of our knowledge, the feasibility of using membrane techniques for the recovery of REEs has not been reviewed to date. This review provides an overview of the application of membrane techniques for the recovery of REEs from waste streams. The performances of the membrane processes

were evaluated according to the REE rejection efficiency, membrane fouling, and purity of the extracted REEs. The economic feasibility of using membrane techniques for the recovery of REEs was analyzed by comparing the selling prices of REEs with the total recovery cost. General remarks and future perspectives on the recovery of REEs using membrane processes are provided.

2. separation
3. occurs through a semi-permeable membranes with different pore
4. size that make contact between desirable phases and control
5. the passage of particles by letting solvent go and catching the
6. solute [180, 181]. The membranes that are used in these separate-
7. tion processes have many different aspects in type, fabrication,
8. and structure and will be reviewed in next section

## 2. Recovery of REEs using membrane techniques

The unique characteristics of REEs are their 4f electrons. In general, REEs have the following chemical configuration of the outer shell: 5s<sup>2</sup> 5p<sup>6</sup> 5d<sup>1-10</sup> 6s<sup>2</sup>, which shields the 4f<sup>1-14</sup> electrons. The most stable oxidation state of REEs is M<sup>III</sup>. All REEs exhibit similar chemical and physical properties because of their similar electronic configurations. They are highly localized elements that are not significantly affected by their crystal shapes (Galhoum et al., 2015). REEs are divided into two groups: light REEs (LREEs, La-Eu) and heavy REEs (HREEs, Gd-Lu). Although Y is the lightest element among the REEs, it is considered a HREE because its properties are similar to those of HREEs (Massari and Ruberti, 2013). Separation of REEs is a challenging process because of their same oxidation states (+III). A semipermeable membrane with different pore sizes that rejects solute particles and allows the entry of the solvent can be applied for the separation of REEs. The material retained is accumulated in the reject stream (Azimi et al., 2017). Membranes with different materials, shapes, thicknesses, pore sizes, and permeabilities are available for this purpose. They can be classified according to their pore sizes as follows: microporous, mesoporous, and macroporous with pore sizes of dp < 2 nm, 2 nm < dp < 50 nm, and dp > 50 nm, respectively (Ambashta and Sillanpää, 2012). The differences between the pore sizes and separation mechanisms of various membranes are summarized in Table 1.

### 2.1. Nanofiltration

Nanofiltration (NF) has a high rejection rate for different ions,

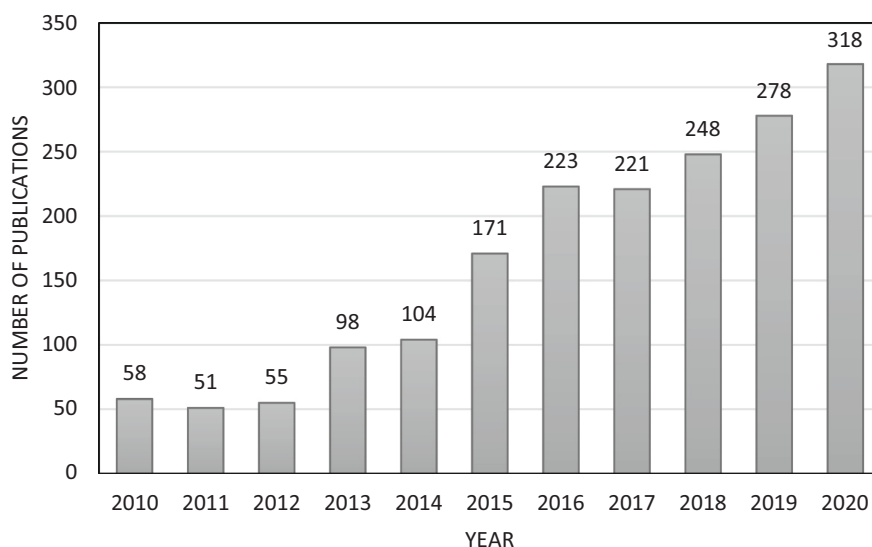


Fig. 1. Number of publications (journal articles, conference papers, and books) on the recovery of REEs from waste streams.

**Table 1**  
Mechanisms and properties of various membranes.

Parameters	Microfiltration membrane	Ultrafiltration membrane	Nanofiltration membrane	Reverse osmosis membrane
Pore size (Å)	>100	30–100	10–30	2–10
Separation mechanism	Mainly sieve	Primarily sieve	Preferential sorption-capillary flow	Preferential sorption-capillary flow

Zakrzewska-Trznadel et al., 2001.

mainly divalent ions (>95%) (Pereao et al., 2018). The primary limitations of NF are low stability of the membrane material and low economic feasibility of the process. Nanofiltration is typically used for the treatment of municipal wastewater (Dischinger et al., 2017) and desalination (Li et al., 2015; Wu et al., 2004). Utilization of NF for the extraction of REEs from waste streams is a relatively new application. NF membranes are selective and efficient for the removal of REEs; nevertheless, the low stability of NF membranes in acidic media is a major drawback (López et al., 2019b). Table 2 presents the different types of NF membranes used for the recovery of REEs along with their operating conditions and recovery.

Separation of Gd from other REEs was performed using a NF membrane; an organic material was employed to increase the selectivity of the membrane for Gd (Sorin et al., 2005). A commercial Desal G10 NF membrane comprising a polyamide layer with an active layer area of 155 cm<sup>2</sup> and a molecular weight cut-off of 2500 Da was used. Diethylenetriaminepentaacetic acid (DTPA) was used as the organic material as it prefers LREEs over HREEs. This organic material was added to the solution because its chelation ability facilitates the extraction of the desired REE. Fig. 2 depicts the mechanisms of chelation and separation. The Gd rejection efficiency of the membrane was >93% under standard conditions of  $\Delta P = 4$  bar and pH > 2.5. The removal of Ce from a feed solution (FS) containing 10–80 mg/L CeCl<sub>3</sub> was conducted using a NF membrane (Murthy and Choudhary, 2011b). A commercial NF-300 membrane was employed in this experiment; the membrane comprised three layers: a polyamide layer (5–20  $\mu\text{m}$ ), a polysulfone substrate layer (50  $\mu\text{m}$ ), and a strengthening layer of polyester (200  $\mu\text{m}$ ). The effective surface area of this membrane was 150 cm<sup>2</sup>, and the pore size was 300 Da. The concentrations of Ce(III) in the FS and permeate solution were measured using inductively coupled plasma-atomic emission spectrometry. The applied pressure was 2–10 bar, and pH was 2–10. Ethylenediaminetetraacetic acid (EDTA) as a chelating agent was complexed with the NF membrane. The Ce(III) rejection efficiency of the membrane was 94.4% at a FS concentration of 10 mg/L and 90.0% at a FS concentration of 80 mg/L. The rejection efficiency increased with an increase in the applied pressure and a decrease in the concentration of the FS. Using EDTA as a chelating agent, the rejection efficiency increased from 11 to 99% (Murthy and Choudhary, 2011b).

A similar method was used for the recovery of Nd using a NF-300 membrane in a Perma®-pilot scale system, and a rejection efficiency of 86.7% was acquired. The Nd rejection efficiency of the membrane increased to 99.5% when sodium dodecyl sulfate (SDS) was employed as a surfactant; the rejection efficiency was 99.4% when EDTA was used. The overall process performance was enhanced by increasing the pH value (optimum pH value was 6–10 pH) and pressure. In addition, the process performance was further improved by reducing the concentration of Nd in the FS (Murthy and Choudhary, 2011a).

Kose Mutlu et al. (2018) evaluated the performance of NF for the recovery of REEs from coal fly ash leachate. The tested membranes were NP010, NP030, DK, and Duracid (Kose Mutlu et al., 2018). The lowest rejection efficiencies were obtained using NP010 and NP030, and the highest rejection efficiencies were achieved using Duracid and DK. The difference in the rejection efficiencies was caused by the different charges and porosities of these membranes. The permeability of Duracid was lower than those of NP030 and DK. Owing to the highly positive charges on NP010 and NP030 as compared to those on DK and Duracid, NP010 and NP030 were less effective in rejecting the REEs. Therefore, DK was the most suitable membrane for the recovery of REEs. The effects of pressure and pH on the REE rejection efficiencies of membranes were also examined. Results showed that with an increase in pH, the REE rejection efficiency increased because at low pH, the high concentration of ions decreased the Donnan potential and shielding of the counter ions, which resulted in low REE rejection efficiencies. The most suitable pH was found to be 3.5; a permeability of 5.5 L/m<sup>2</sup>/h/bar was obtained at this pH. A high rejection efficiency (99%) was acquired for Y<sup>3+</sup>, Nd<sup>3+</sup>, Dy<sup>3+</sup>, Er<sup>3+</sup>, Eu<sup>3+</sup>, and Tb<sup>3+</sup> along with other elements including Mg, Ca, Fe, Al, Si, and Na. The REE rejection efficiency increased with an increase in pH and applied pressure. Permeate flux was also affected by pH, which decreased with an increase in pH. At high pH, the rejection efficiencies for ions increased, which increased the possible fouling of the membrane.

Another source of REEs is acid mine water (AMW), which mainly contains sulfonic acid, Fe(III), Al(III), other metal ions, such as Zn, Cu, and Ca, and REEs at small concentrations. The NF membranes have been utilized to treat AMW as some types of NF membranes exhibit enhanced affinities for specific ions based on the ionic functional groups, altering

**Table 2**  
Rejection efficiencies and characteristics of various types of nanofiltration membranes.

Membrane	Active layer	MWCO (Da)	Organic chelate	pH	Pressure (bar)	Rejection efficiency %	Rejected REEs	Ref.
NF-300	Polyamide	300	EDTA SDS EDTA	2–10 6–10	2–10 >6	99 99.5 99.4	Ce Nd	(Murthy and Choudhary, 2011a)
Desal G10	Polyamide	2500	DTPA	>2.5	4	>93	Gd	(Sorin et al., 2005)
MPF-34	Polymeric acid-resistance	200	–	1.5	6–13	>80	La, Pr, Nd, S, Dy, and Yb	(López et al., 2018)
NF270	Polyamide on a thin film composite	180 ± 20	–	1	–	>98	La, Pr, Nd, Sm, Dy, and Yb	(López et al., 2019b)
Desal DL	Poly(piperazine amide) and an additional polyamide layer on a thin film composite	–	–	–	–	98	La, Pr, Nd, Sm, Dy, and Yb	(López et al., 2019a)
HydraCoRe 70pHT	Sulfonated polyethersulfones on a thin film composite	–	–	–	–	96–66	–	(López et al., 2019a)
DK	Thin film composite membrane	150–300	–	3.5	12–24	99	Y, Nd, Dy, Er, Eu, and Tb	(Kose Mutlu et al., 2018)
NP010	Polyethersulfone	1000	–	1.5	12	9.5	Y, Nd, Dy, Er, Eu, and Tb	–
NP030	Polyethersulfone	500	–	1.5	12	12.9	–	–
Duracid	Thin film composite membrane	150–200	–	3.5	12	86.4	–	–

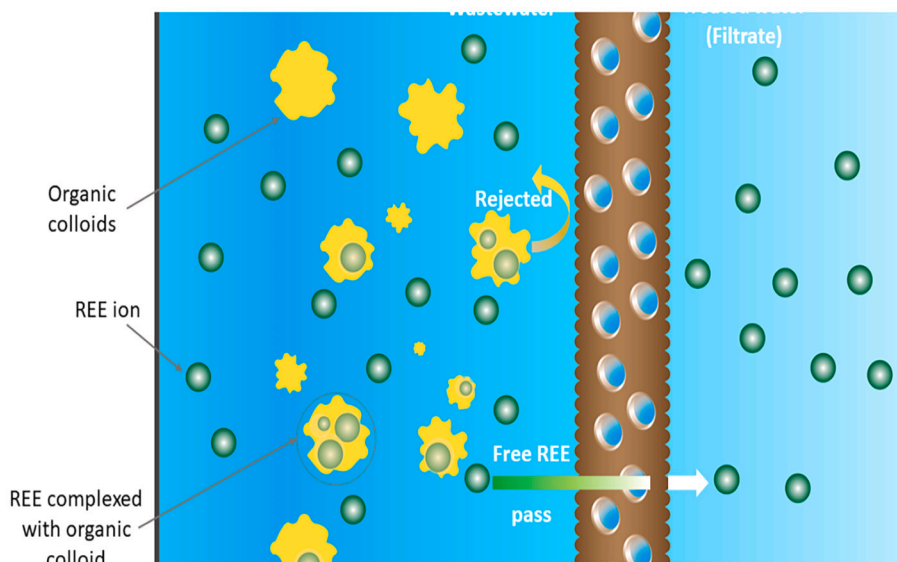


Fig. 2. Rejection of the REEs complexed with organic colloids by a NF membrane.

the selectivities of membranes, present in them. The NF membranes with specific functional groups were employed to recover REEs and other ions from AMW (López et al., 2019a; López et al., 2019b; López et al., 2018).

A solution-electro-diffusion model was used to describe the transport of ions through the membrane. Typically, three different membranes were studied: NF270 (López et al., 2019a; López et al., 2019b), Desal DL (López et al., 2019a; López et al., 2019b), and HydraCoRe 70pHT (López et al., 2019a). The NF270 membrane consists of a thin film composite comprising three layers: an active upper layer built of piperazine semi-aromatic-based polyamide, a support layer of microporous polysulfone, and a backing layer composed of non-woven polyester. The primary functional groups in NF270 are amine (R-NH<sub>2</sub>) and carboxylic (R-COOH) groups. Although Desal DL has the same active layer as NF270, it contains an additional layer that impacts the hydrophilicity, roughness,

and acid–base properties of the membrane. The active layer of HydraCoRe 70pHT is built of sulfonated polyethersulfones incorporated into a thin film composite comprising a polysulfone layer and a polyester backing layer. Each membrane has different types of functional groups; thus, they exhibit different isoelectric points, which endow them with different affinities for ions and compounds. For instance, NF270 and Desal DL have both R-NH<sub>2</sub> and R-COOH groups, and their isoelectric points are 2.5 and 4.0, respectively, as Desal DL has a double active layer. HydraCoRe 70pHT comprises sulfonic groups (R-SO<sub>3</sub>H). The structures of the functional groups of NF270 and HydraCoRe 70pHT along with their rejection abilities for different ions are shown in Fig. 3.b and a, respectively. The NF270 and Desal DL membranes are positively charged, whereas HydraCoRe 70pHT is negatively charged.

Effects of the pH of the FS on the rejection efficiencies of NF

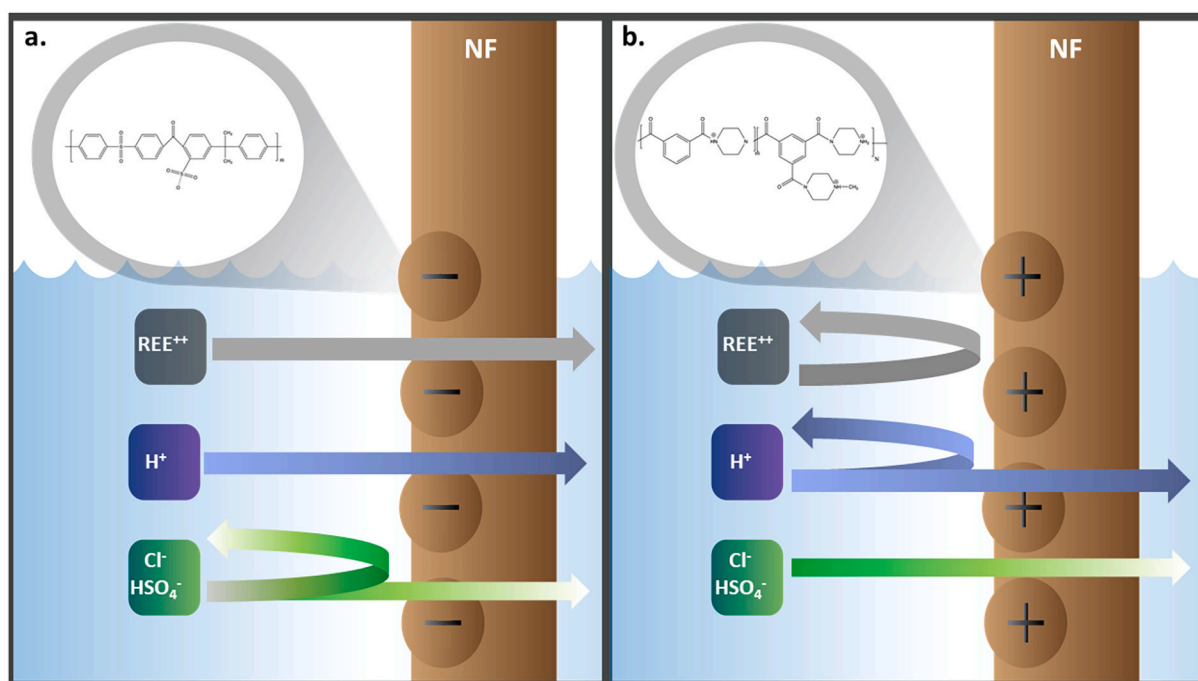


Fig. 3. Schematic showing the ion selectivities of NF membranes according to the charge of the functional group in the active material of the membrane: a. HydraCoRe 70pHT and b. NF270.



membranes have been investigated (López et al., 2019a). Results demonstrated that with a decrease in the pH of the FS, the rejection efficiencies of the membranes for metals increased. This was owing to high repulsion between the membrane and the cations at higher pH. However, for  $H^+$  ions, the rejection efficiencies of NF270 and Desal DL were lower because  $H^+$  easily crossed the membrane due to its small size relative to those of other cations. When HydraCoRe 70pHT was used, the presence of negative charge enhanced the rejection of anions and decreased the rejection of metal cations as compared to the cases of positively charged membranes (the rejection efficiency was  $\sim 80\%$  for HydraCoRe 70pHT, whereas it was  $>98\%$  for NF270 and Desal DL). The concentrations of Al(III) and Fe(III) did not affect the rejection of metal ions; however, the removal of anions, including sulfate and chloride, increased because of the change in the type of sulfate species present in water with an increase in the concentrations of Al(III) and Fe(III). Furthermore, the composition of the membrane influences the membrane flux; for example, NF270 reaches a flux of up to  $40 \mu\text{m/s}$ , whereas Desal DL reaches a flux of  $15 \mu\text{m/s}$  because of the thinner structure of NF270 than that of double-layered Desal DL (López et al., 2019a). The effect of aging was also studied, and the rejection efficiency of a 4-week-old membrane was found to be lower (80–70%) as compared to  $>98\%$  of a new membrane (López et al., 2018).

Ceramic nanomembranes are another type of NF membranes that are chemically and mechanically more stable than polymeric nanomembranes. Nevertheless, ceramic nanomembranes have low selectivities and high costs when compared with polymeric membranes (López et al., 2020). Fig. 4 shows a schematic of the comparison between ceramic and polymeric nanomembranes.

Ceramic membranes were used for the removal of metal ions such as As(III), and rejection efficiencies of 70–100% were obtained depending on the type of membrane and the quality of FS (Aosai et al., 2014; Dupré et al., 1999; Markus et al., 2018; Pédröt et al., 2009). Acid-resistant ceramic membranes were employed for the treatment of AMW (López et al., 2020). Typically, two types of membranes were analyzed: a ceramic membrane composed of  $TiO_2 + Al_2O_3$  and a polymeric acid-resistance sheet from Koch Membrane System (MPF-34). The size of the nanopores, active area, membrane diameter, and membrane

thickness in the ceramic membrane was 1 nm,  $44.9 \text{ cm}^2$ , 6.5 mm, and 2.0 mm, respectively. The targeted REEs were La, Pr, Nd, S, Dy, and Yb. Rejection efficiencies of MPF-34 and the ceramic membrane ( $TiO_2$ ) were approximately 80% and  $< 60\%$ , respectively. The selectivity of ceramic membrane for REEs was low because this membrane also rejected other metals including Al, Zn, and Cu.

Despite all the promising perspectives of using NF for the recovery of REEs from waste streams, there are still some challenges, such as high tendency of membrane fouling, low selectivity between ions, low chemical resistance, and limited lifetime of membranes, that impede large-scale applications of this technique. Membrane fouling in NF is more complex and severe as compared to that in other membrane techniques (that is, ultrafiltration and microfiltration) because the high negative charge of the membrane leads to a strong attractive force between the positively charged foulants and the membrane surface (Hafiz et al., 2021; Jarusutthirak et al., 2007; Kramer et al., 2020). This might result in a shorter membrane lifetime and inhibit the recovery performance of the process. NF demonstrates a high rejection efficiency (that is, 90%) for divalent ions and a low rejection efficiency (namely, 20–80%) for monovalent ions depending on the charges and molecular sizes of ions (Parlar et al., 2019). The use of NF alone is often insufficient to obtain the desired separation because it is difficult to completely retain one solute and allow the entry of the second component simultaneously, particularly when the solute molecules have similar sizes or charges. Polymeric NF membranes have low resistances to organic solvents (Lim et al., 2017). This is due to the low stability and deterioration of the active layer of the membrane. The aforementioned limitations can be overcome using a pretreatment process; nevertheless, the development of a NF membrane with higher resistance to fouling and higher selectivity for specific ions depending on the application is highly recommended.

## 2.2. Ultrafiltration

Pore sizes of ultrafiltration (UF) membranes are relatively large compared to those of NF membranes; thus, UF membranes cannot reject REEs dissolved in water. However, UF membranes can be used for the

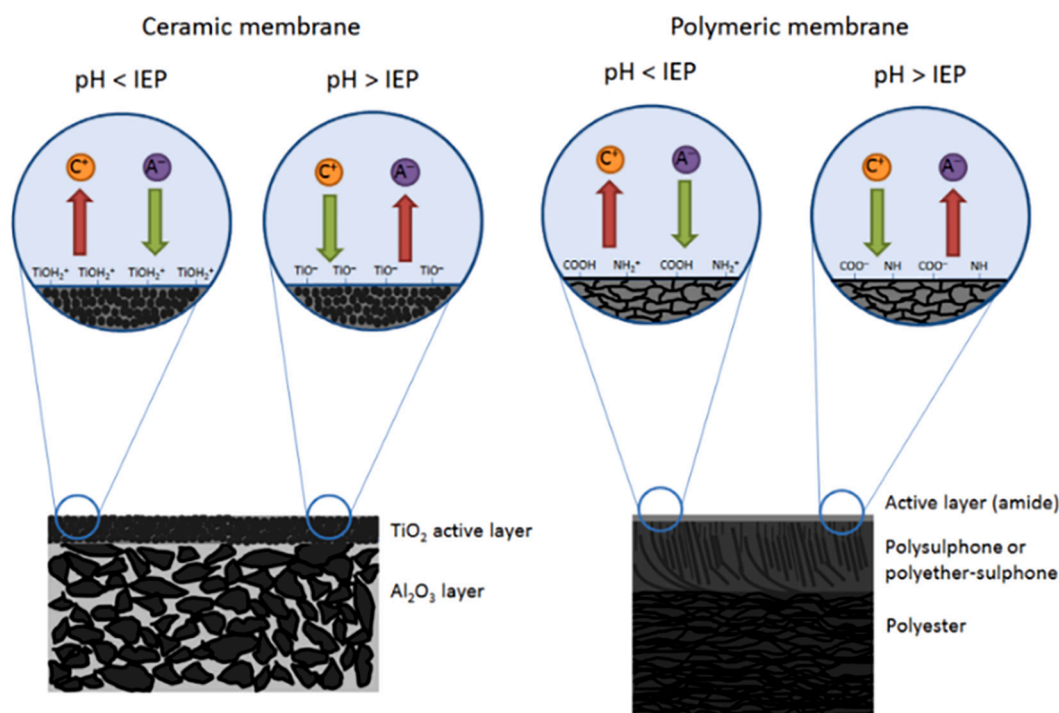


Fig. 4. Schematic of the comparison between ceramic and polymeric NF membranes (Benfer et al., 2001).

separation of dissolved organic matter (DOM) as their sizes range from 1  $\mu\text{m}$  to 1 nm. The DOM can retain contaminants via sorption or by forming complexes with metals including REEs (Chaufer and Deratani, 1988; Kose Mutlu et al., 2018; Marty et al., 1997; Masse et al., 1988; Pourret et al., 2007a; Pourret et al., 2008; Pourret et al., 2007b). Ultrafiltration has been employed in several studies to investigate the influences of DOM on the amount and speciation of REEs in different natural waters (Pourret et al., 2007b). The UF membranes were used to determine the amounts of contaminants carried and complexed by DOM (Pourret et al., 2007a). These membranes with pore sizes of 30, 10, and 5 kDa were utilized to filter the DOM. The REEs are present in organic or Al/Fe-complexed colloids. Different thermodynamic models were used to predict the complexation of metals with DOM using 0.2  $\mu\text{m}$  cellulose acetate filters with pore sizes of 30, 10, and 5 Da and a pH of 3–9 (Pourret et al., 2007b). Various studies have reported that a large fraction of REEs forms complexes with DOM. The ability of organic colloids to form complexes with metals and sorb metals can be utilized to separate metals at low concentrations from water using UF membranes; this method is called complexation–UF (Chaufer and Deratani, 1988; Juang and Chiou, 2000). In other studies, complexation–UF was used to separate REEs from waste streams, using a concept similar to the one used in the case of NF (Fig. 2). Herein, three different types of DOMs were employed as complexation agents in UF: polymers (Stern et al., 2007), humic substances (HS) (Innocenzi et al., 2018b), and micelles (Juang and Chiou, 2000). Table 3 presents the different complexation agents used in UF. Polymer-complexation–UF (PCUF) involves the complexation of polymers with metals to obtain large-sized substances in water that can be filtered by an UF membrane. Several water-soluble polymers, such as polyacrylic acid (PAA), polyethylenimine (PEI), and sodium salt of PAA (PAASS), were used for the recovery of metals by UF membranes (Duan et al., 2015). A PCUF was utilized to recover REEs via PAA complexation, and Amicon Ultra-15 centrifugal units were employed to filter the REE–polymer complexes. The used membrane and PAA had molecular weight cut-off values of 30 and 4000 kDa, respectively. The rejection efficiency of the UF membrane for REEs exceeded 90% when 30 mg/L PAA and a pH of 8–9 were used. Another method to modify UF membranes is the use of a micellar to form a micellar-enhanced UF membrane (MEUF). Ceramic UF tubular membranes with molecular weight cut-off values of 210 and 1 kDa were used along with SDS (Innocenzi et al., 2018a). The recovery of both Y and Zn exceeded 99%. The recovery decreased with an increase in pore size and a decrease in the SDS concentration. However, water flux decreased after the addition of SDS, and the influences of pressure and time were negligible.

### 2.3. Microfiltration

Microfiltration (MF) membranes have large pore sizes, and therefore, these membranes cannot reject free REE ions from waste streams. Nevertheless, MF can be coupled with other techniques to enhance the overall rejection efficiency of the membrane. Liquid–liquid extraction is a suitable technique for the removal of dissolved materials; however, it is less efficient for the removal of materials at low concentrations owing to its several limitations including easy emulsification, loss of extractant, high energy consumption, and long process time (Hou et al., 2013). Membrane dispersion microextraction (MDME) is a method in which the

extraction efficiency is enhanced using MF membranes as a dispersion medium. Microdroplets of one phase are made through the small pores of a MF membrane and are forced to cross the membrane to interact with the desired materials in the other phase and thus extract these materials (Fig. 5). Various studies have analyzed the rejection efficiencies of stainless-steel micromembranes for different REEs using 2-ethylhexyl phosphoric acid-2-ethylhexyl ester (EHEHPA) as an extractant (Chen et al., 2018b; Hou et al., 2015a; Hou et al., 2013; Hou et al., 2015b).

Removal of different types of REEs using MDME, EHEHPA, and stainless-steel MF membranes was investigated (Hou et al., 2013). Removal of La(III) from a hydrochloride acid–EHEHPA kerosene system was conducted using MDME, and the effects of bulk and organic phase flow rates on the extraction efficiency were analyzed. An extraction efficiency of up to 95.9% was achieved when the flow rate of the organic dispersion phase was 60 mL/min. Influence of pH during La(III) stripping was also studied, and a recovery of 82% was acquired at a pH of 2.0. Recovery of Ce(III) using MDME was also studied (Hou et al., 2015a). Effects of the flow rates of the aforementioned two phases, temperature, pH, and the initial concentration of EHEHPA on the extraction efficiency were investigated. Results showed that when the initial concentration of EHEHPA was increased from 0.01 to 0.04 mol/L, the extraction efficiency increased from 25 to 99.7%. With a decrease in temperature and an increase in the concentration of Ce(III), the extraction efficiency decreased until an equilibrium state was reached. The optimum pH for the process was determined to be 3.63, and generally, the extraction efficiency increased with an increase in pH. The impacts of the flow rates of both phases on the extraction efficiency were examined, and with an increase in the flow rates of both phases, the extraction efficiency increased (Hou et al., 2013). At high flow rates, the interaction area between the two phases increased due to the smaller diameters of microdroplets. Effects of the process operating conditions on the diameters of droplets were also investigated (Hou et al., 2015b). Extraction of Pr(III) using the same MDME system was examined by imaging the droplets of the membranes to visualize changes in the diameters of droplets with respect to pH, flow rates, and initial concentration of EHEHPA (Hou et al., 2015b). With an increase in the initial pH, EHEHPA concentration, and flow rates, the diameters of the droplets decreased. Inspired by Hailong's study, the extraction of three different types of REEs (namely, Nd(III) (light), Eu(III) (medium), and Er(III) (heavy)) using MDME and the same extractant and MF membrane was analyzed (Chen et al., 2018a; Chen et al., 2018b). When a phase ratio of 25:1 was employed, an extraction efficiency of >98% was obtained at an extraction time of 30 s. Furthermore, stripping efficiencies of 0.99, 0.96, and 0.91 were achieved for Nd(III), Eu(III), and Er(III), respectively. Table 4 shows the results and the highest rejection efficiencies acquired using MDME for the separation of REEs. All results demonstrated that the extraction of REEs via MDME was enhanced from 5 to 30% to up to >90% when MF membranes were used, which indicate that the combination of MF and MDME is a promising technique for the recovery of REEs.

Microfiltration suffers from membrane fouling and its inability to remove dissolved ions. MF suffers from membrane fouling because of the accumulation of suspended solids on the membrane surface and inside the pores. The fouling on the membrane surface can be reversed by backwashing; nevertheless, the accumulation of foulants inside the membrane pores is irreversible and shortens membrane lifetime.

**Table 3**  
Removal of rare earth elements (REEs) by ultrafiltration membranes.

Membrane	MWCO	Organic material	Organic material conc.	pH	Rejected REEs	Initial conc.	Rejection efficiency %	Ref.
Amicon Ultra-15 centrifugal filter units	30 kDa	Polymer: polyacrylic acid (PAA)	1000 mg/L	7.5	La Sm	–	89.2 95.8	(Duan et al., 2015)
Single tube Membralox <sup>®</sup> TI-70 ceramic membrane	1 kDa	Surfactant: sodium dodecyl sulfate	10 mM	5–6	Y	30 mg/L	99	(Innocenzi et al., 2018a).

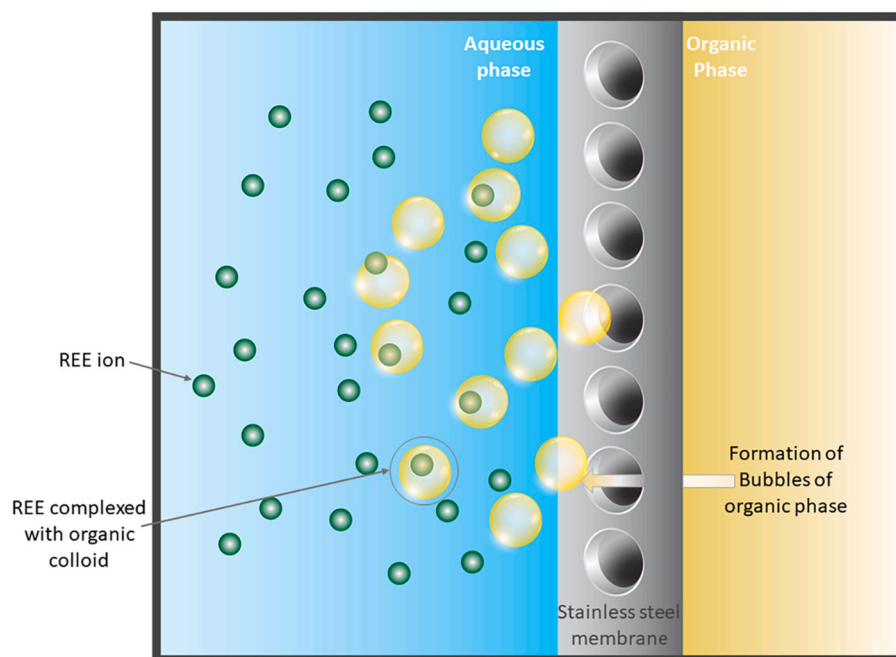


Fig. 5. Removal of REEs via membrane dispersion microextraction using a MF membrane for organic phase dispersion.

Table 4

Extraction of rare earth elements (REEs) by a stainless-steel microfiltration membrane.

Membrane	Extractant	pH	Rejection efficiency %	Rejected REEs	Ref.
Stainless steel with a pore size of 5 $\mu\text{m}$	2-Ethylhexyl phosphoric acid-2-ethylhexyl ester (EHEHPA)	3.33	94	La	(Hou et al., 2013)
		3.14–3.63	99	Ce	(Hou et al., 2015a)
		>3.5	99	Pr	(Hou et al., 2015b)
2-Ethylhexyl phosphoric acid-2-ethylhexyl ester (P507)		4	99	Nd	(Chen et al., 2018a)
			96	Eu	(Chen et al., 2018a)
		–	>90	Nd, Y, Sm, Gd, Dy, and Er	(Chen et al., 2018b)

Although MF membranes have high removal rates for suspended solids and large molecules, they do not have the ability to reject REEs.

#### 2.4. Reverse osmosis and forward osmosis

Reverse osmosis is a hydraulic pressure-driven process in which water molecules diffuse through a semipermeable membrane, whereas most of the dissolved metal ions do not. In RO, the hydraulic pressure of the draw water must exceed the osmotic pressure of the feed water (Hafiz et al., 2020). Although RO is mostly used for desalination, it has been used for the recovery of REEs due to its satisfactory rejection efficiency, high water flux, membrane resistance to biological attack, high chemical stability, and resistance to high temperatures (Pereao et al., 2018). The primary shortcomings of using RO for the recovery of REEs are high energy consumption and membrane fouling, which make RO less feasible for the recovery of REEs (Pereao et al., 2018). The rejection of REEs, specifically Gd, was investigated across different treatment

steps in two advanced water treatment plants (Lawrence et al., 2010). The major treatment steps in the treatment plants were MF, RO, and advanced oxidation using UV irradiation followed by peroxide. The concentrations of REEs in samples obtained during different steps of treatment were analyzed, and the concentrations of REEs in the MF feed sample and MF permeate were found to be identical, which suggested a lack of removal of REEs during MF. Moreover, very low concentrations of REEs were detected in the RO permeate, implying that highly efficient removal occurred during RO. The average concentration of Gd reduced from 0.39 nmol/kg to approximately 0.59–0.7 pmol/kg in the RO FS, and a significant rejection efficiency of 99.8% was achieved. Reverse osmosis was used to treat wastewater released from the REE smelting industry (Gui et al., 2020). The applicability of commercial RO membranes for the treatment of wastewater produced by the REE smelting industry was examined. Reverse osmosis was employed to treat wastewater generated during extraction, washing, and precipitation stages in a rare earth separation plant in Nanchang, China (Gui et al., 2020). The wastewater contained abundant ammonium nitrogen ( $\text{NH}_4^+\text{-N}$ ), very low concentrations of REEs, chemical oxygen demand (COD), heavy metals, and total P (TP).

Forward osmosis (FO) is a relatively new membrane technique and an osmotic pressure-driven membrane process where water moves across a selectively permeable membrane from the FS with low solute concentration to the draw solution (DS) with higher solute concentration (Alfahel et al., 2020). It results in the concentration of the feed stream and dilution of the highly concentrated DS (Thabit et al., 2019). The advantages of FO over RO are lower rate of membrane fouling, the possibility of reverse fouling as a cleaning method, and lower energy consumption (Hafiz et al., 2019). The primary limitations of FO are the scarcity of suitable membranes for the rejection of REEs and difficulty in the regeneration of DS (Fu and Wang, 2011). Recovery of REEs from acid mine drainage using a FO membrane was analyzed (Pramanik et al., 2019). Results demonstrated that when the active layer faces the DS (AL-DS), higher water flux and higher fouling rate were obtained as compared to those in the case when the active layer was facing the FS (AL-FS). Nevertheless, higher REE rejection efficiency was acquired using the AL-FS orientation owing to the lower impact of the level of internal concentration polarization on osmotic pressure, which in turn influences the inside of the membrane support layer. The observed

variation in the REE rejection efficiency was caused by the differences between the molecular weights, shapes, and crystal structures of REEs. At high temperatures, the water flux increased for both membrane orientations. However, the REE rejection efficiency decreased because of the enlargement of the membrane pore size. The highest REE rejection efficiency was achieved at a FS and DS temperature of 20 °C due to the increase in reverse salt flux and at high pH as at basic pH, electrostatic repulsion between the REE ions and membrane molecules increased. Lanthanum(III) and cerium(III) ions were steadily rejected in the cases of both membrane orientations and at different pH values, whereas the rejection of Dy(III) exhibited a different trend owing to alterations in membrane surface charges; the highest and lowest rejection efficiencies were obtained at a pH of 6–7 and 3, respectively (Pramanik et al., 2019). Table 5 presents the performances of RO and FO for the rejection of different REEs.

## 2.5. Dialysis processes

Diffusion dialysis (DD) and electrodialysis (ED) are two ion-exchange membrane (IEM) processes in which different types of membranes, such as UF membrane, are utilized for the removal of metals including REEs (Lin et al., 2015). Dialysis membranes are semipermeable IEMs where ionic functional groups are attached to the backbone of the membrane, which can be dissociated or ionized. These functional groups facilitate the concentration, separation, and recovery of metal ions from aqueous media. The IEMs are of two types: cation exchange membranes (CEMs) and anion exchange membranes (AEMs). Separation of metal ions by an AEM or a CEM is primarily achieved by the exclusion of ions carrying the same charge as the fixed ions of the membrane structure and partially realized by the pores of the membrane (Besha et al., 2020). In ED, a direct electric current is passed through an external circuit, causing cations and anions to move toward the cathode and the anode, respectively. This technique selectively separates ions through the IEMs, resulting in two streams: dilute and concentrate streams (Gurreri et al., 2020). IEMs and ED membranes are synthesized with different functional groups and materials. Fig. 6 shows the configurations of DD and ED cells.

Separation by DD is driven by the spontaneous diffusion and the concentration gradient of ions (Luo et al., 2011). The DD membranes are usually composed of cellulose triacetate (CTA) and PEI due to the high removal abilities of these materials induced by the formation of their complexes with metals and introduction of a charged layer on the membrane surface. The performance of DD for the recovery of REEs from electronic waste was examined (Hammache et al., 2021). The polymeric membranes used herein were CTA, PEI incorporated with one of the following carriers: di-(2-ethylhexyl) phosphoric acid (D2EHPA) (M1), tridodecylamine (TDDA) (M2), trioctylamine (TOA) (M3), and trioctylphosphine oxide (TOPO) (M4). The FS used was diluted leachate and deionized water. To retain the positive charge on the membrane surface, pH was maintained at 4 for all membranes to reduce the strong

**Table 5**  
Performances of reverse osmosis (RO) and forward osmosis (FO) for the rejection of different rare earth elements (REEs).

Process	Membrane	Operating conditions	Rejection efficiency %	Rejected REEs	Ref.
RO	–	pH: ~7	99.9	Gd	(Lawrence et al., 2010)
FO	TFC polyamide	pH: 3–5, T: 20 °C	91	La	(Pramanik et al., 2019)
	Support layer: polysulfone	pH: 6–7, T: 20 °C	93	Ce	
	Orientation: AL-FS	pH: 6–7, T: 20 °C	96	Dy	

electrostatic repulsion between the membrane surface and the positive REE ions. Lower water uptakes were obtained using membranes with P-based carriers (M1 and M4), whereas higher water uptakes were acquired employing membranes with amino-based additives (M2 and M3) because of the presence of more hydrophilic groups in these additives, thus leading to smaller contact angles of the membranes with the water surface. At pH 4, M1 had the highest positive charge, implying its higher stability and therefore high REE rejection efficiency. The low water uptakes in the cases of M1 and M4 are attributed to the difficulty in formation of coordination complexes between the carriers and REEs owing to favorable interactions between PEI and the phosphoryl groups of TOPO and D2EHP; this makes the addition of these carriers to CTA/PEI polymeric membranes unfavorable and thereby reduces the REE removal efficiencies. The positive charge of M2 was less than that of M3 and M4, which indicated its less stability; however, M2 exhibited higher REE rejection efficiency due to its denser structure (Hammache et al., 2021).

An AEM with ammonium functional groups and a CEM with sulfonic acid functional groups were utilized to separate and concentrate Sc<sup>3+</sup> (Li et al., 2021). Sulfuric acid was used as an anolyte and a catholyte to rinse the electrodes. A CEM comprising sulfonic acid functional groups was applied to recover REEs from coal ash (Couto et al., 2020). Sodium nitrate and coal ash suspended in deionized water were used as a catholyte and an anolyte, respectively. Results obtained from these studies demonstrated that sufficient removal efficiencies were achieved using higher applied current, higher applied voltage (Mosadeghsedghi et al., 2018), lower feed concentration of REEs, and uniform acidic pH of the FS to enhance the solubility of Sc<sup>3+</sup> (Couto et al., 2020; Li et al., 2020). The maximum Sc<sup>3+</sup> removal efficiency from synthetic FSs was obtained to be 99.5% using the constant current mode with a low energy consumption of 0.26 kWh/m<sup>3</sup> (Li et al., 2020). The applicability of ED for the separation of REEs from real industrial wastewater was also investigated by designing a similar system that contained Sc<sup>3+</sup> coexisting with Al<sup>3+</sup> and Fe<sup>3+</sup> at higher concentrations as impurities. The total ion removal efficiencies increased with an increase in the feed pH, and the highest Sc<sup>3+</sup> removal efficiency of 65.4% was acquired at pH = 4 (Li et al., 2020). Effects of the addition of EDTA as a complexing agent to enhance the selective separation of REEs on the removal rates of REEs were analyzed. Lower removal rates of Ce and Yb ions were obtained in the presence of EDTA (Mosadeghsedghi et al., 2018). Table 6 presents the removal percentages of the different REEs achieved using DD and ED.

## 3. Economic feasibility of using membranes for the recovery of REEs

The costs of REEs have been varying since the last decade due to the changing mining restrictions. The global demand for REEs has been increasing. Charalampides et al. (2015) have summarized the market prices of REEs during the period 2009–2012 (Charalampides et al., 2015). The prices of REEs have continuously increased, except for a sudden fall in 2012. The total demand for REEs is expected to reach 200,000 tons/year in the next few years (Haque et al., 2014). The variation in the prices of rare earth metal oxides over the past four years is presented in Table 7. A decline in the market prices of some elements was noticed after 2019 owing to the development of techniques for the recovery and extraction of REEs.

Economic feasibility of using membrane processes for the recovery of REEs from coal ash leachate was evaluated, and the acquired REE recovery efficiencies were compared with those obtained via other conventional extraction techniques. Solvent extraction and ion exchange are the most commonly used methods for the recovery of REEs at a commercial stage. In SE, the choice of extractants depends on the processed solution and the available budget. Commercially, D2EHPA, 2-ethylhexyl phosphonic acid mono-2-ethylhexyl ester, and Aliquat 336 have been extensively used in the SE of REEs (Xie et al., 2014). The primary



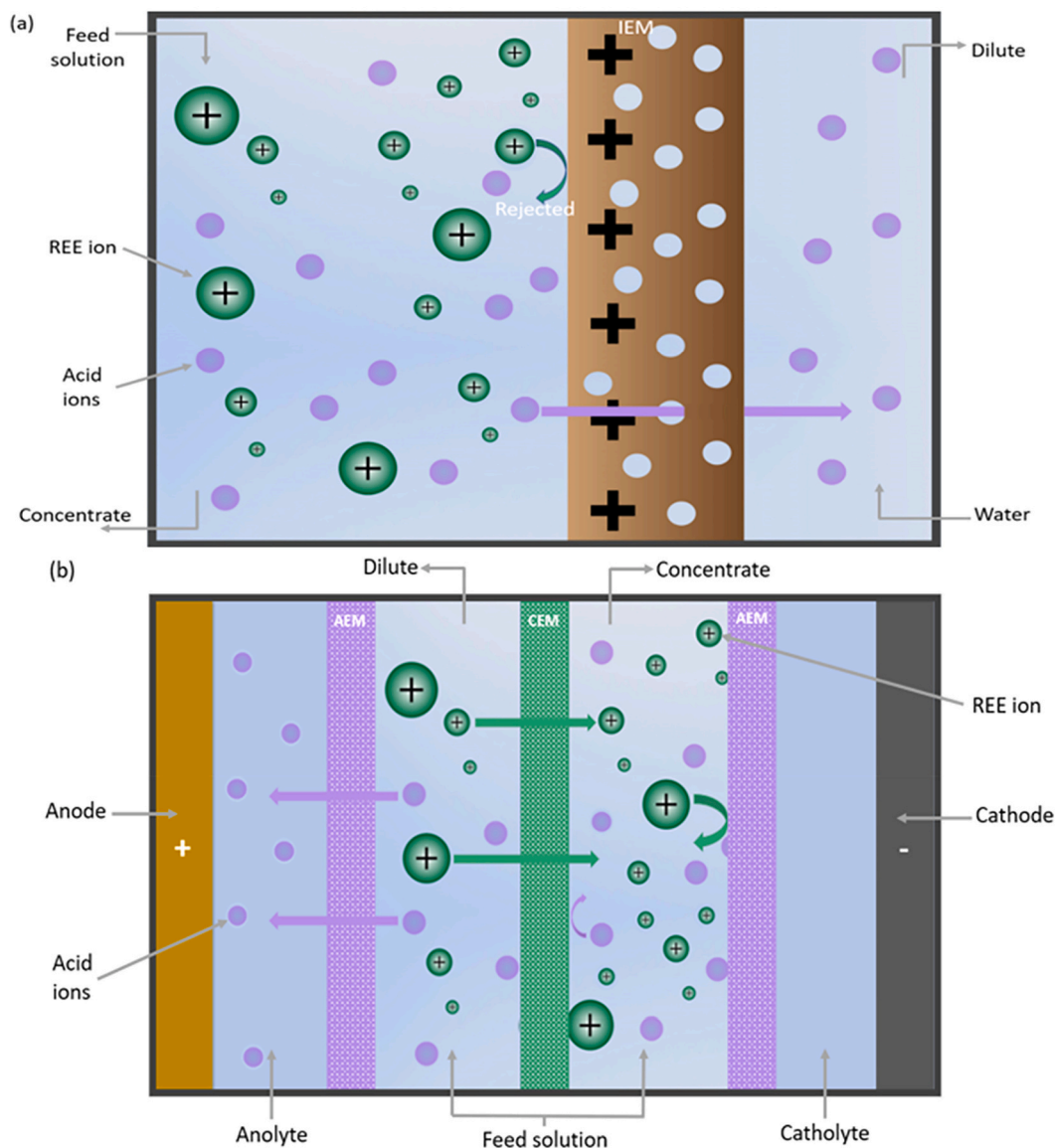


Fig. 6. Schematic of a) diffusion dialysis and b) electrodialysis cell configurations.

**Table 6**  
Rare earth element (REE) rejection by diffusion dialysis (DD) and electrodialysis (ED) membranes.

Process	Membrane	Operating Conditions	Removal %	Rejected REEs	Ref.
ED	AEM: PC-SA CEM: PC-SK polyester reinforcement Electrodes: anode: Ti coated with Pt/Ir and cathode: stainless steel	Voltage (U): 1–3 V Current (I): 50–100 mA pH: 2–4 [Sc <sup>3+</sup> ]: 20–160 mg/L	99.5	Sc <sup>3+</sup>	(Li et al., 2020)
ED	CEM Mixed metal oxide electrodes	Current (I): 50 mA Time (t): 3 days pH: 2–3 at the cathode compartment [REE] <sub>total</sub> : 447 mg/L	55–62 74	Tb and Dy Nd	(Couto et al., 2020)
ED	CEM: CMX AEM: AMX	Voltage (U): >13 V Current (I): 22–31 mA Time (t): 60–180 min [Yb]: 49–90 mg/L [Ce]: 1186–1690 mg/L	–	Yb Ce	(Mosadeghsedghi et al., 2018)
DD	CTA and PEI Carriers: D2EHPA, TDDA, TOA, TOPO	pH: 4 Time (t): 6 h	–	Nd, Pr, La, Ce, Sm, Eu	(Hammache et al., 2021)

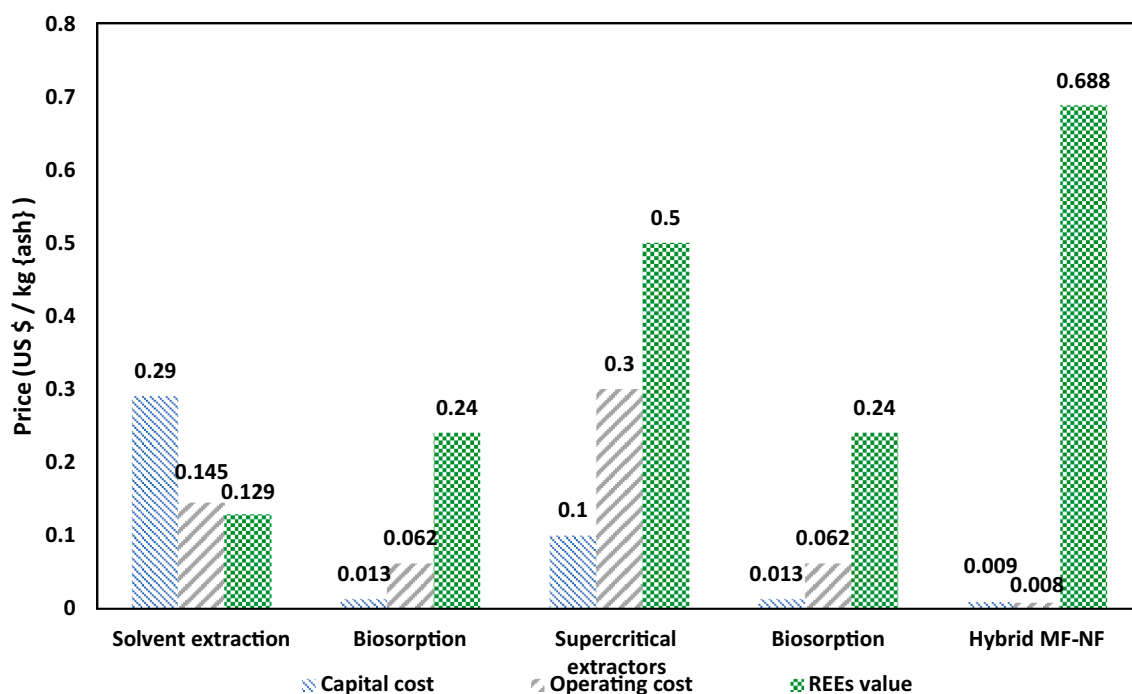
**Table 7**  
Prices of rare earth metal oxides in US \$/ton during the past four years.

Metal oxide	2016	2017	2018	2019	2020
Lanthanum oxide	1856	2021	2185	1877	1810
Cerium oxide	1565	1856	2146	1899	1815
Terbium oxide	405	430	454	504	510
Dysprosium oxide	192	185	177	235	240
Europium oxide	68	60	51	35	30
Neodymium oxide	39,327	44,566	49,804	44,578	41,700
Praseodymium oxide	47,988	55,737	63,486	54,017	52,200

Statista, 2020.

drawback of SE is the use of organic solvents, which increase environmental pollution, are hazardous to the health of the operator, and expensive (Tao et al., 2021). A techno-economic assessment report on the recovery of REEs from coal ash using SE was published by Argumedo et al. (2020). The estimated project lifetime was 20 years, and the daily processed amount of ash was 30 tons/h. They speculated a capital cost of \$76.4 million and an operational and maintenance cost of \$38 million/year. The calculated capital cost and operational cost per kg of ash were estimated to be \$0.29 and \$0.145, respectively. They reported revenues of approximately \$34 million in a year from the sale of rare earth oxide at a cost of \$0.129 per kg. The revenues from the sales of REEs alone were not sufficient to cover the costs of the process; therefore, profits were increased by selling the byproducts, primarily ferric chloride. Ion exchange is the reversible exchange of ions between a liquid phase and a solid phase without significant changes in the structures of the solids (Mohebbi et al., 2019). The main drawbacks of ion exchange are the high usage of hazardous chemicals, the inability to remove large amounts of non-ionized hazardous sludge, and high cost (Sole et al., 2017). Recently, improved methods, such as supercritical extraction, biosorption, and membrane processes, have been established to overcome the limitations of the conventional processes. Supercritical fluid extraction is a leaching process where a supercritical fluid is employed instead of large quantities of acids. Recently, a cost analysis on the recovery of REEs from coal ash using a supercritical extractor was conducted (Das et al., 2018). Carbon dioxide (CO<sub>2</sub>) was used as a

supercritical fluid because of its low viscosity and high diffusivity, which facilitated its application in SE. The processing cost for extracting REEs from one ton of coal ash varied between \$380 and \$1200 for 550 g of REEs. The value of REE oxides that might be obtained per ton of ash was estimated to be between \$6 and \$557, with a median of \$250. The lowest total cost reported for the extraction of REEs from coal ash using supercritical extractors was \$0.4 per kg of ash. However, the value of the extracted REEs was approximated to be \$0.5 per kg of ash. Economic feasibility of using biosorption for the recovery of REEs from several feedstocks, including coal ash, was evaluated (Jin et al., 2017). Biosorption was found to be suitable for the extraction of REEs due to the high capacity and affinity of cell surface for metal binding. The value of REEs ranged from \$437 to \$526 per kg of total REE oxides, whereas the capital cost was between M\$ 5.7 to M\$ 6.4. An industrial-scale plant was planned to process 500,000 tons of coal ash/year, supposing a plant life of 20 years. Based on this, the total cost of recovery was estimated to be \$0.075 per kg of ash, whereas the value of the extracted REEs was predicted to be approximately \$0.24 per kg of ash. Techno-economic feasibility of using a hybrid MF–NF membrane process for the recovery of REEs from coal ash was investigated (Kose Mutlu et al., 2018). Separation by NF was performed under a pressure of 12–24 bar at a pH of 1.5–3.5 on a lab-scale. The most optimized total cost was less than \$0.02 per kg of ash. The value of the extracted REEs was approximated to be \$0.735 per kg of ash, which was higher than the total cost of recovery. Fig. 7 shows the cost analysis and comparison between the capital and operational expenditures (CAPEX and OPEX, respectively) of MF–NF and conventional recovery processes. A general trend noticed herein was that the value of the recovered REEs was higher than the total cost of recovery mainly because of the increasing market prices of REEs. It is to be noted that the value of the recovered REEs depends on the type and source of the coal ash used and the efficiency of recovery. Moreover, the profitability of REE recovery substantially depends on the recovered amount of expensive REEs such as Sc, Nd, and Pr. The primary finding of this analysis is that the total costs of using membrane processes for the recovery of REEs are considerably lower than those of the conventional processes due to the minimal use of chemicals required for operation and the use of low pressures (approximately 12 bar) (depending on the



**Fig. 7.** Comparison between the capital and operational expenditures of solvent extraction (Argumedo et al., 2020), supercritical extraction (Das et al., 2018), biosorption (Jin et al., 2017), and hybrid MF–NF (Kose Mutlu et al., 2018) for the recovery of REEs from coal ash.

FS). Capital expenditure (Capex) for the membrane for the membrane process was almost the same as that for biosorption and 11 times lower than that for supercritical extraction. Operational expenditure (Opex) of the membrane process was eight times lower than that of biosorption and 37 times lower than that of supercritical extraction. The total cost of the membrane process was four times less than that of biosorption and 23 times less than that of supercritical extraction. This indicates that membrane processes can be promising methods for the recovery of REEs.

#### 4. Summary & future prospect

This review evaluates the techno-economic feasibility of using various membrane techniques for the recovery of REEs. Discovering an efficient process for the recovery of REEs from waste streams has become urgent because of the increasing worldwide demand for REEs and their limited supply due to mining and export restrictions imposed by governments. Moreover, REEs must be completely removed from wastewater owing to their high toxicities. Therefore, using membrane techniques for the recovery of REEs from waste streams can be an appropriate solution specifically because of their high removal efficiencies and economic feasibility when compared with those of the conventional separation processes. Nanofiltration has been found to be the most suitable membrane process for the recovery of REEs, and UF and MF can be integrated with other conventional techniques to improve their overall REE recovery efficiencies. Reverse osmosis was considered unsuitable for the recovery of REEs because of the high rejection of most solute ions in the waste stream and its high energy consumption. There are still some challenges that need to be overcome before using membrane techniques for the recovery of REEs on a commercial scale, which are as follows:

- Enhancing the ion selectivity of NF membranes is essential for the recovery of REEs using these membranes at a commercial stage. Therefore, more research efforts must be directed toward developing highly selective NF membranes targeting REEs.
- Very few studies have been conducted on the economic feasibility of using NF membranes for the extraction of REEs from waste streams. Thus, more research must be performed to evaluate the economic feasibility of using NF membranes for the extraction of REEs from different waste streams.
- Membranes have low stability in aggressive media such as acids and alkali. Therefore, more studies must be conducted to enhance the chemical stabilities of membranes, particularly when influents produced during the recovery of REEs are used.

#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Acknowledgement

This research is made possible by graduate sponsorship research award (GSRA6-1-0509-19021) from Qatar National Research Fund (QNRF). The statements made herein are solely the responsibility of the authors. Open access funding was provided by Qatar National Library.

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