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A comprehensive review of recent advances in membrane innovations for efficient heavy metal removal from mine effluents

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ABSTRACT

The growing global challenge of water scarcity, intensified by industrialization and population growth has heightened the need for effective wastewater management in industries, including the mining sector. Mining operations discharge substantial volumes of wastewater laden with toxic metal such as copper, iron, cobalt, lead and mercury which poses significant environmental as well as human health risk. Efficient wastewater treatment is crucial to mitigate these effects. While technological advancements have improved mine effluents treatment, there remains a need for advanced methods that enable not only removal of the toxic metals but also recovery of resources such as valuable metals and water. Due to its high efficiency, selectivity and low environmental footprint, membrane technology has gained attention especially in the treatment of various mine effluent. Though fouling is a major challenge in its implementation. The review gives an updated overview on the membrane technology in mining effluent treatment, examining the performance of various membranes (pressure driven membrane, thermal and concentration) in removal of metals and recycle of valuable resources from mine effluents such Acid Mine Drainage (AMD) and other mine effluents. It also examines innovative approaches such as pre-treatment processes, hybrid membrane system as well as the use nanocomposites polymeric membrane. Furthermore, the recent advances in membrane modification techniques such as chemical vapour deposition, sol-gel process, lithography, Atomic layer deposition, layer by layer and electrospinning have been discussed. Studies show that >95 % separation efficiency,> 85 % water recovery and >90 % metal recovery for hybrid membrane processes and chemical precipitation. The recovered metals show high purity of >99 %. Studies indicate that standalone membrane system have limitations in recovery of metals but hybrid systems (membrane coupled

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Abbreviations: AMD, Acid Mine Drainage; Al_2O_3, Aluminium oxides; $CoFe_2O_4$/CuO, Cobalt ferrite-copper oxide nanoparticles; CVD, Chemical Vapour Deposition; DCMD, Direct Contact Membrane Distillation; REEs, Rare Earth Elements; GO, Graphene Oxide; FBR, Fluidized Bed Reactor; FTIR, Fourier-transform infrared; NF, nanofiltration; RO, Reverse Osmosis; MF, Microfiltration; UF, ultrafiltration; Zr_2O_3, Zirconium Oxide; MWCNT, Multi-Walled Carbon Nanotube; SEM, Scanning Electron Microscopy.

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with other complementary methods) can achieve better results. This review identifies future direction for advancing membrane technology in sustainable mine wastewater management for improved environmental as well as mine operations.

Introduction

The recent increase in global demand for finished metal products accompanied by increased industrialization and population growth, has led to deterioration and scarcity of freshwater sources [1]. It is estimated that approximately 359 billion cubic metres of industrial wastewater is produced globally per year, with only 52 % of this water treated while the remaining 48 % is discharged untreated into the ambient environment [2]. Anthropogenic activities such as mining play a pivotal role in this process especially in the generation of large volumes of wastewater [3–5].

In mining, water is a primary commodity used for various mining operations. For instance, processing copper sulfide ore through a conventional crushing-grinding-flotation-thickening steps requires 1.5 to 3.5 tons of water per ton of ore [6]. Furthermore, water in mining is used for: 1) dewatering the mines to have access to the ores, 2) dust suppression on hauls and cooling of the drilling bits, 3) separating crushed minerals in chemical processes like during froth flotation and 4) transportation of slurries and suspensions to the tailing storage facility. In general, surface water bodies such as rivers and lakes are preferred sources of such water [7]. AMD, dewatering wastewater, process effluents and tailings are the main wastewater from mining operations. These are often laden with high metal content and coarse material [6].

Heavy metals are the main pollutants in mine wastewater emanating from electroplating, ore processing and extraction industries [8–10]. Even in minute concentrations, these elements pose a significant threat to ecosystems due to their non-biodegradable nature. In human populations, exposure to heavy metal, including, chromium (Cr), cobalt (Co), zinc (Zn), copper (Cu), cadmium (Cd), iron (Fe), lead (Pb) and mercury (Hg) poses major health risks. Their carcinogenic and neurotoxic properties, underscore the urgency of effective wastewater treatment [11–13]. To avert such toxicity and meet the stringent permissible discharge limits, it is imperative to treat mining wastewater effectively.

At present, various methods have been explored in their efficiency to treat mine wastewater and these include; Chemical, physical and biological methods especially for the removal of heavy metals. Chemical treatments include chemical precipitation, neutralization, flocculation, and coagulation while physical methods such as sedimentation, filtration and adsorption [14]. Biological methods such as bioreactors and sulphate reducing bacteria has been used extensively. However, collectively these methods have many challenges; formation of enormous sludge [15], high energy consumption [16] and selectivity limitations [17]. Furthermore, these processes are time consuming and non-selective especially for high metals [18]. These drawbacks contribute to the persistent discharge of mine effluents laden with heavy metal into the ambient environment. Given the inevitability of wastewater generation in mining operation, it is imperative to explore novel treatment methods which are robust in targeting metals even at lower concentration.

Membrane technology has attracted the attention of many researchers especially in the treatment of various mine wastewaters such as acid mine drainage (AMD) [19], acidic mine wastewater [20–22] and synthetic mine wastewater [23]. In their comprehensive review, Panayotova and Panayotov [23] highlighted the extensive application of membrane technology in mining including the treatment of flotation water, pregnant leaching solutions, lithium brines, the extraction of metals from wastewater as well as the remediation of AMD and the recovery of valuable reagents and metals [24]. Owing to their high selectivity, small surface-to-area ratio and low environmental footprint, membranes are the best alternative for treating mine wastewater, especially in removing heavy metals from mine wastewater [4,25,26]. However, one of the major drawbacks of this technology is membrane fouling which reduces the performance of the membrane and increase the operational cost of the membraneas well as reduce the longetivity of the membrane [27–29].

The recent advancement in nanotechnology has provided a way to circumvent these shortcomings by the utilization of nanomaterials which can be incorporated into the membrane matrix. Owing to their small size to volume ratio, physical properties, and easy manipulation, nanoparticles such as metal and/ or metal oxides, organic and nano rods have extensively been used in membrane for the removal of heavy metals from industrial effluents [30,31]. However, in their individual processes membrane system are limited by fouling, which reduces their efficiency and performance. Therefore, to this end, membranes with enhanced selectivity, productivity, resistance to fouling, and stability, all while being more available at lower cost and with fewer manufacturing problems are required [25]. In particular, with the new concept sustainable mining focused on not only removal but also the recovery of valuable resources such as clean water, precious metals and metals to achieve a circular economy has opened opportunities for the hybrid application of membranes to reduce the adverse limitations of single membrane processes [32–34].

The aim of the current review is to provide an up–to–date compendium of latest research on the feasibility of pressure driven membranes in mine wastewater treatment by exploring literature on the strategies have can be used to mitigate membrane fouling with a focus on the nanocomposite membranes, membrane modification techniques and the use of hybrid membranes system for effective removal of heavy metals in mine wastewater. Furthermore, this review gives an overview of membrane technology, the factors influencing membrane performance and classification of membranes.

Fundamentals of membrane technology in wastewater treatment

Membranes are artificial structures that exist independently, typically with pore size dimensions ranging from 1 to 100 nm. They exhibit significant increase in surface area to volume ratio making them selectively permeable barriers. Their pores are sized to allow the passage of materials that are smaller than the pores while retaining a wide range of particulate and dissolved ions, depending on their nature that is, their surface charge, pH, and concentration [26,35]. Characteristically,membranes can be classified according to: i) structure ii) material and iii) driving force [36,37]. Fig. 1 summaizes the classifiation of membranes and highlights examples of each specific type of membrane.

Classification of membranes

Structure

Membranes can be classified as isotropic or anisotropic depending on the structure of the top layer. Isotropic membranes are symmetrical membranes made up of a single material. These are categorized into three as:1) microporous membranes; 2) dense film nonporous and 3) electrically charged. Microporous membranes are also known as sieving membranes and the separation is based on the principle of size exclusion. These membranes have a high flux, for example, ultrafiltration and microfiltration membranes. Nonporous membranes are membranes with no pores. Membranes including nanofiltration and reverse osmosis are some of the examples. Electrically charged membranes represent membranes whose surface have been deposited with either a positive /negative ion. Ion exchange membrane is the principle of exchange in these membranes [38,39].

On the other hand, anisotropic membranes are unsymmetrical membranes made up of different materials. These contain material that vary in composition and, or in structure. They are made up of a highly selective thin top layer that is supported by a dense permeable layer. Example of such membranes are forward osmosis membranes and reverse osmosis membranes [40,41].

Material

Membranes can be categorized based on the material used for synthesised as organic, inorganic and hybrid membranes. Organic membranes also known as polymeric membranes are composed of carbon as the fundamental material. Although carbon serves as the basis for these types of membranes, the most basic compounds such as the carbon oxides, carbides, cyanides, and carbonates are not used in the manufacturing because of their gaseous or liquid state. Polymeric materials such as cellulose acetate, polyether sulfone, polydimethylsiloxane, polyacrylonitrile, polyvinyl chloride, and polytetrafluoroethylene have been extensively used in the fabrication of polymeric membranes. Organic membranes can further be grouped into polymer (single) and co-polymer membrane. Single polymer organic membranes consist of the use of one polymer in the fabrication process while co-polymer membranes involve the use of two-polymers in the synthesis of the membrane to increase the efficiency of the membrane. Owing to their excellent permeability, selectivity and stability, organic materials are good candidates for the synthesis of organic membranes including commercial nanofiltration and reverse osmosis membrane. However, these materials are limited in temperature range of between (0–30 °C).

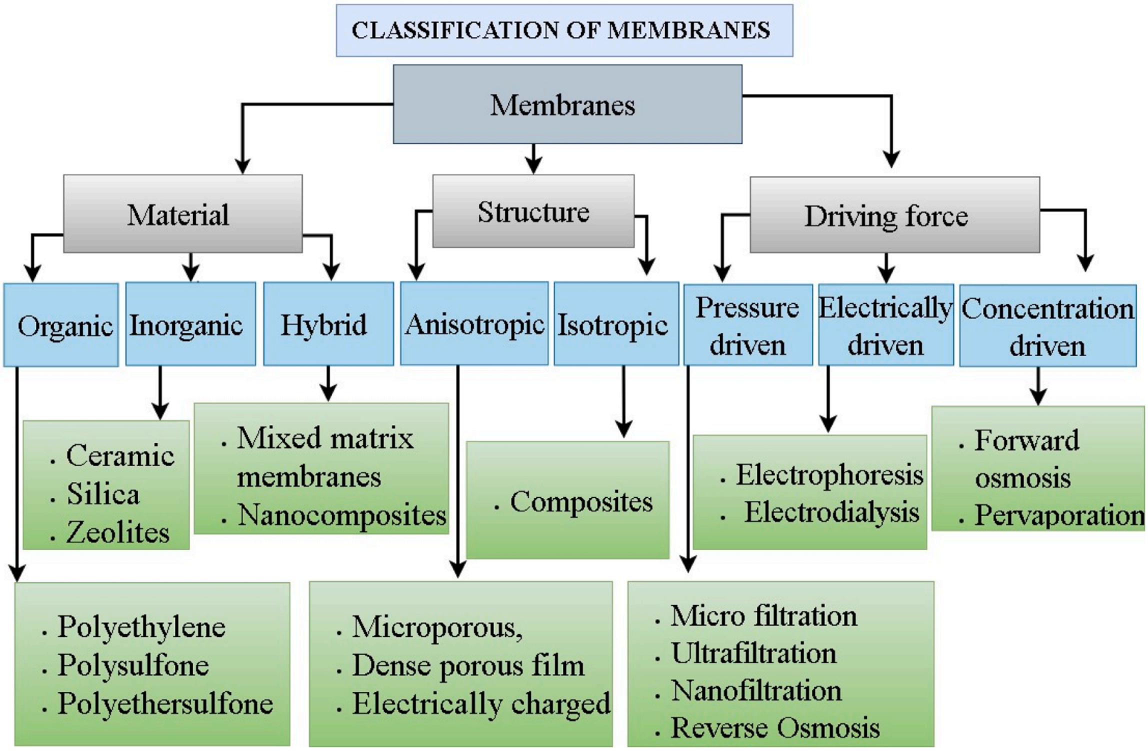

Fig. 1. Classification of membranes.

Furthermore, increased humidity and organic solvents such as alcohols and benzene impair the structure of these membranes [36,42,43].

The second category of membranes are inorganic membranes. These are usually made ceramic, zeolites, silica, carbon nanotubes and metals such as oxides of aluminium, titanium, selenium and zirconium [26,38]. In harsh environmental conditions such as acidic or alkaline wastewater, inorganic membranes have good mechanical strength, better resistance to chemical attack and highly tolerant of pH and oxidation [44]. Furthermore, they are thermally and chemically stable [45]. inorganic membranes are potent materials for improved wastewater treatment [42]. The introduction of these materials in the polymer matrix is a potent way of treating wastewater as they improve the performance of the membrane. However, expense and low selectivity are the major shortcoming with these types of membrane [39,46].

Hybrid membrane are membrane that exist in combination of organic and inorganic or and both inorganic or organic to compensate the shortcoming of either the organic or inorganic membrane [45]. Hybrid membranes represent the most recent classification of

Table 1
Technical and scientific comparison of membrane and hybrid filtration systems: types, applications, and performance analysis.

System Type	Description	Applications	Advantage	Disadvantages	References
MF	Membrane filtration with pore size of 0.1–10 µm	Used as pre-treatment for higher pressure driven such as RO, NF for the removal of suspended solids, bacteria and other large particles	low energy consumption, effective for large particle separation	Less selectivity for viruses, dissolved substances such as dissolved metal ions and anions	[26]
UF	Filtration membrane with pore size of 0.001–0.1 µm with pressure between 1 and 2 bar	Wastewater treatment especially drinking water removal of suspended particles	Low capital operational and operational cost Low pressure	Separation is dependent upon size exclusion. Unable to remove dissolved ions and small organic matter	[49]
NF	Filtration membrane with allowing the removal of divalent ions with pore size between 1 and 5 nm and operating pressure of 10–20 bar	Treatment of industrial effluents for heavy metal removal, dye removal	Strong rejection of divalent ions Reduced energy consumption Moderate pressure	Membrane scaling and fouling Difficulty in removing monovalent ions such as Na^+,	[19]
RO	Pressure driven membrane filtration with pore size 1–0.5. pressure between 20 and 100 bar	Mine wastewater treatment, tannery wastewater, drinking water treatment	Highly selective even for monovalent ions, effective in industrial wastewater treatment ad concentrating metal ions for recovery	High energy consumption due to high operating pressure, generation of brine which needs further reprocessing	[50]
MD	Thermal difference filtration membrane dependent upon the change from liquid to vapour phase	Desalination of brine, textile wastewater, radioactive wastewater	Alternative heat sources such as solar and geothermal energy as such can be used in renewable energy, separate highly	High cost of operation Membrane fouling and wetting.	[51]
ED	Uses electric potential difference to separate ions through ion exchange membrane (cation –exchange & anion exchange	Removal of dissolved ions from wastewater electroplating industry, industrial wastewater	High water flux, high recovery, no chemicals pressure needed, high metal recovery	Separation is limited to ionized molecules, unsuitable for high saline wastewater Cannot remove viruses	[52]
MBR	Leverages the benefits of biological treatment methods such as SRB with membrane filtration to remove solids and biological contaminants	Wastewater treatment to remove bacteria, viruses, suspended solids	High quality water effluent, smaller space requirement, shorter hydraulic retention time	Membrane fouling, higher operational cost, sludge generation, aeration needed	[53]
FO	Osmotic driven membrane. Separation is dependent upon the difference in concentration between the DS and the FS	Wastewater treatment for the removal of heavy metals	No external hydraulic pressure needed, easy in cleaning, regeneration of the DS	Selection of the suitable DS For efficiency it needs to be coupled with other membrane systems	[54]
NF-RO	Filtration membranes combining NF with RO in which NF is a pre-treatment.	Wastewater treatment to get high quality water and concentration heavy metals for recovery	High flux, lower operating cost reduced energy consumption, high removal percentages for heavy metals	Scaling on the surface of the RO membrane and fouling	[50] [22]
FO-RO	Membrane filtration system in which FO and RO are combined based on both pressure difference and concentration gradient.	Advanced water reuse, desalination of challenging feed water.	Better performance, high water recovery, high selectivity, reduced membrane filtration	Complex designing and maintence, high capital and operational cost	[55]
ED-RO	Hybrid membrane system that combines the advantages of ED with RO. In which RO removes the dissolved salts while ED concentrate the brines to remove more and recover more water.	Concentrating wastewater and recovery of water and other valuable resources from brine	High concentration brine, anti-scaling, water recovery, reduced TDS, possibility of zero liquid discharge	High energy consumption, complexity in operation, limited efficiency at high salinity	[56]

membranes-based material. Metal organic framework are hybrid inorganic-organic material with unique structures and fascinating properties with great potential in wastewater treatment [3]. Hybrid membranes such as nanocomposites membranes have been used extensively in wastewater treatment [47,48].

Driving force

Generally, membrane can be grouped into three categories based on the driving force assisting the feed through the membrane: Concentration driven membranes (Forward Osmosis, liquid membrane), electrically driven membranes (membrane dialysis, electrophoresis) and pressure driven membranes (UF, MF, NF and RO). Table 1 shows the advantages of membrane filtration and the disadvantages.

Herein the next section discusses in details, pressure driven membranes.

Membranes propelled by pressure represent the most frequently utilized category of membranes, which can be classified into four types:microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) [11]. They leaverage the pressure difference between the feed and permeate streams driving mechanism to facilitate solvent transport through the membranes [57]. The difference between these four types of membranes is on their pore size and the particles rejected [58]. Rejection, a critical parameter, is influenced by membrane charge exculsion, size exculsion, and the chemical/physical interactions among the solvent, solute and the membranes [59]. Microfiltation uses low static low pressure as driving force for separation. However, one of the limitation of microfiltration in its application for treating mine impacted water is the size of the pores 0.1–1 μm which can not incept heavy metals and other dissolved solid particles [60]. Table 2 shows the difference among the four types of pressure driven membranes.

Ultrafitration on the other hand, has pores range from 2–100 nm. Microorganism, macromoecules and colloids are retained using UF which operates in the pressure range of 1 to 10 bar [61]. In the context of wastewater treatment, NF and RO membranes have gained a solid recognition for their effectiveness in removing metal ions [25]. NF membranes are considered tight membranes because of their pore size of 1 nm and molecular weight out of 300–500 Da Owing to this, these membranes can retain metal ions, microorganism and pathogens [35]. RO is known to be the most efficient of the four pressure-driven membranes because of its small pore size of 0.1–1 nm allowing for the retaination of all polltants including monovalet ions such as Na^+ [62]. Commercially, both RO and NF membranes are made from thin film composite. Numerous studies have shown that NF membranes are preferable than RO removing of pollutants from wastewater due to their lower operating costs, low energy consumption, sustained flux, and low pressure [21,22,63]. The separation in these membranes is dependent upon the size exclusion and the charge exclusion discussed below.

The mechanism and principle of separation in membrane

Separation in membrane is achieved through several mechanisms such as diffusion, convention, electrostatic, steric hindrance and Donnan effect. By acting as a barrier, the membrane limits the passage of metal ions and other solutes from passing through. Additionally, the membrane's morphological features such as pore size, pore distribution, degree of hydrophilicity and the existence of functional groups play a cardinal role in the separation process [65]. The mechanism of separation is dependent upon size exclusion and charge exclusion. These principles govern the separation of heavy metals from mine wastewater in NF membranes. Other mechanism include dielectric [66,67] and hydration mechanism [35].

The retention of metal ions on the membrane's surface is contingent on both the size of the pollutant and the pore size of the membrane. This process, known as the sieving mechanism regulates the separation of uncharged molecules [35,64]. The rejection rate for metal ions in wastewater increases as membranes with smaller pores such as NF and RO are utilized [68].

Donnan mechanism also known as charge –charge exclusion used for the separation of charged solutes. This exclusion arises from the repulsion between the charged membrane surface and the metal ions. The membrane's surface may contain functional groups including carboxyl, amine and sulfonic acid which dissociate when exposed to wastewater stream forming a charged surface. The pH of

Table 2
Diffference among the four pressure driven. Adopted and modified from [9,37,64].

Membrane processes	MF	UF	NF	RO
Pore size	0.1–1 μm	100–200 nm	< 5 nm	<1 nm
Examples of pollutants rejeted	Suspended particles (plastics	Macro and micromolecules, solution with colloids (bacteria, virus, microplastics, oragnic, sugar)	Divalent cations and ions, lactose, sucrose	Monovalent ions and all contaminants
Molecular weight cut-off	100–500	20–150	2–20	0.2–2
Membrane characteristics	Porous isotropous	Porous asymmetric	Finely porous asymmetric/ thin –film nanocomposite	Non-porous asymmetric /composite
Operating pressure	1–3	2–5	5–15	15–75
Advantages	Less energy requirement, Low capital and operatationalcost.	Less energy consumption and low pressure needed	Greater removal of heavy metals and a more enhanced separation compared to UF	Remarkable heavy metal removal efficiency
Disadvantages	Requirement for pretreament	Secondary pollutants and requirement for pretreatment	Lower water permeability	More energy requirement

the feed water is a critical factor in separation mechanism because at the pH of isoelectric point (the pH at which no electrostatic interactions occur) there is charge on the surface of the membrane [35]. Mullett et al. [65] investigated this effect by employing a NF270 to remove metal ions from mine wastewater. The authors observed that at the pH greater than IEP, the membrane rejected more cations because it was positively charge while at the pH below the IEP the membrane became more negatively charged hence the flux reduced because there was a concentration polarization on the surface of the membrane.

In membrane separation, the separation of divalent ions is much higher than monovalent ions. The dielectric exclusion arise when an ion interacts with the bound electrical charges, induced at the solvent-membrane interface because of the interaction between ions and membrane materials with different dielectric constants [70].

Factors affecting membrane performance

The role of membrane technology in sustainable mining is anchored on understanding the operating conditions for obtaining optimum results. Agboola [29] identified three main factors that affect membrane performance: 1) membrane characteristics; 2) feed characteristics and 3) operating parameters. This section discusses in depth the operating conditions like pH, temperature, feed concentration and the pressure, and their impact on membrane performance (Fig. 2).

Operating conditions

Temperature is important in membrane performance. An increment in the temperature reduces the viscosity of the feed solution causing an increase in permeate flux and consequently an increased permeation rate [71–73]. In a study conducted by Dévora-Isiordia et al. [74] the effect of feed water temperature on RO performance was examined, revealing that an increment in temperature increases the diffusivity and the permeate concentration. Subsequently, increasing the mass transfer coefficient. Recently, Harharah et al., (2022) found similar results when the temperature was elevated from 25 °C to 45 °C for the removal of Cu(II) from synthetic wastewater, observing an increase in Cu(II) ions removal from 89.98 to 91.05 %. As the temperature of the feed solution increases, there is an imbalance in the membrane structure, causing the pores to expand and allowing the passage of large amounts of solutes (ions) through the membrane [76]. It is without a doubt that care must be taken in fully understanding the operating temperature least the integrity of the membrane is comprised.

The rate at which water passes through the membrane in a given time called the flow rate is a critical parameter in the performance of the membrane. The flow rate is directly proportional to the permeate flux and inversely to the solute rejection. [75] reported a 2.5 % increment in permeate flux, from 82.2 (kg/m^2.h) to 84.3 (kg/m^2.h) as the feed flow rate was increased from 2(L/min) to 4 (L/min) while maintaining constant pressure (20 bar), temperature (T = 25 °C) and concentration (100 ppm) when treating wastewater in a crossflow system operating RO membranes. Rejection increased as the flow rate increased due to the reduction in concentration polarization especially in nanofiltration membranes [77].

The transmembrane pressure is a crucial factor, particularly for pressure driven like RO and NF. An increase in pressure increases the permeation rate of the water while reducing the transport of solutes across the membrane [76]. Pressure propels the movement of water through the membrane while restricting the movement of ions, hence increasing both the rejection and flux of the membranes especially in small pored membranes [78]. Fig. 3 illustrates the effet of pressure on rejection as well as flux. However, the major drawback for these membranes is high energy consumption and high pressure needed for the separation to occur. Studies have shown

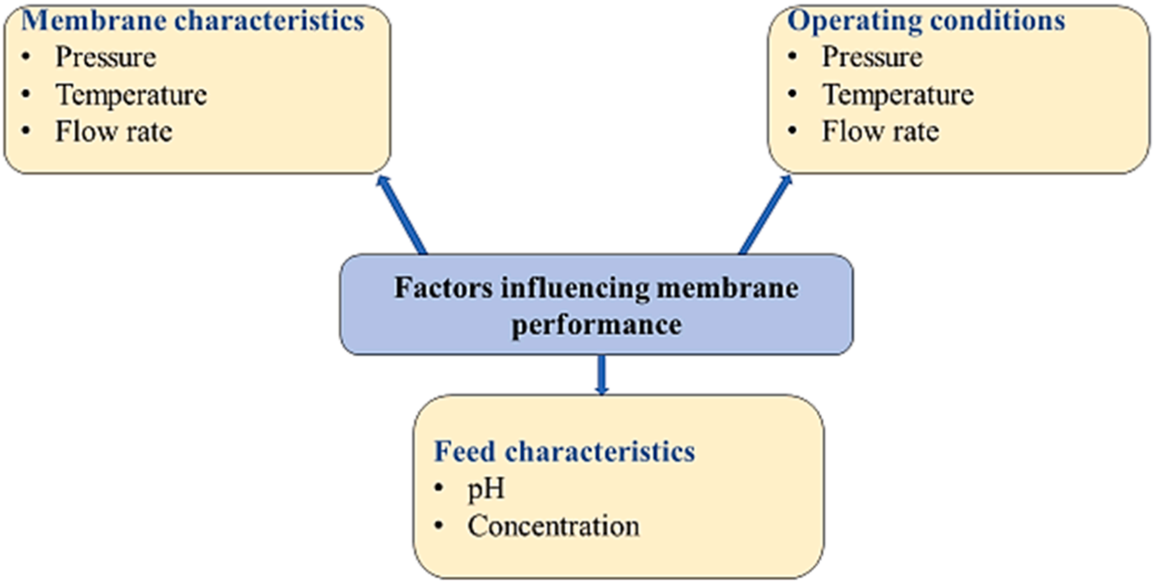

Fig. 2. Factors influencing the performance of the membrane.

excellent performance of these membranes in mine wastewater treatment [79,80].

Membrane characteristics

The membrane characteristics play a vital role in the retention of heavy metals and overall performance of the of the membrane. Pore size, surface charge, roughness and the hydrophilicity are among the most important membrane characteristics [25,29]. The pore size distribution typically must be smaller than the size of the particles in the mine wastewater to facilitate retention through size exclusion and achieve high selectivity. The surface chemistry including membrane charge determines the ionisable functional groups present in the structure of the membrane. The interaction between the feed solution and the membrane surface is vital for the separation of ions especially through the Donnan exclusion. The charge of the membrane is a function of the zeta potential which is the function of the pH. Positively charged membranes have higher retention for metals ions [81]. Hydrophilicity plays a critical role in enhancing the permeability of water as well as the antifouling properties of a membrane system. In membrane systems, the hydrophilicity is associated with the water contact angles and the smaller the water contact angles, the more hydrophilic the membrane [25,82]. Addition of hydrophilic metal oxide nanoparticles to membranes increases the hydrophilicity of the membrane [83,84]. Care must be taken in the manufacture of membrane to ensure improvement membrane characteristics such as surface chemistry, morphological characteristics such as the roughness, the charge, the hydrophilicity. For this purpose, various methods of membrane modification such IP, microwave, lithography and sol-gel have been explored in this venture [25]. Membrane surface roughness also influence the performance of pressure driven membranes. Membranes with smooth or homogeneous surface tend to be less susceptible to fouling. Fouling on heterogeneous surfaces is caused by the continuous trapping of solutes or colloidal organic particles in the valley [85].

Feed characteristics

Mine effluents is usually laden with higher concentration of salts, metals and organic matter. High salt concentration such as sulphate, carbonates, nitrates which are part of the ore processing, these causes scaling and fouling on the membrane surface. In AMD treatment, sulphates cause irreversible fouling which reduces the water flux as well as reduce the rejection of the metal ions [21]. In this case, to preserve the integrity of the membrane it is imperative that a pre-treatment system such as coagulation, sedimentation is done before membrane filtration.

The feed concentration is directly linked to the decrease in rejection rate [76,86]. An increase in feed concentration increases the concentration polarization on the surface of the membrane of negatively charged ions on the surface of the membrane, resulting in a shield negatively charged membrane. Consequently, leading to low permeability of the solute [87]. For example, Harharah et al. [71] reported a decrease in the permeate flux by 4.2 % from 87.41 to 83.86 (kg/m^2·h) when the feed concentration was increased from 25 to 150 (ppm) while the other parameters remained constant ($T = 25$ °C, $P = 20$ bar, QF = 3.2 L/min). [88] found similar results when the Zn feed concentration was increased. The decrease was attributed to the increase in the osmotic pressure which decreases the effective pore size of the membrane increasing the concentration polarization near the membrane surface, leading to membrane fouling.

The feed pH plays a cardinal role in separation of solutes from the wastewater. It influences the zeta potential by governing the charge of the functional groups within the membrane material which is relative to the membrane wettability [30,89]. Mullett et al. [69] investigated the effect of pH on the rejection of metal ions using two nanofiltration membranes (NF 270 and TS80). They noted that at pH less than 3, the cationic rejection of Mg and Ca from the wastewater was higher at 97 % while at pH greater than 3, the rejection rate of the ions decreased. A negatively charge was noted at this pH, attracting the cations in the wastewater to the membrane surface. Consequently, the membrane with low pH than the IEP are usually positive charged while those with a higher pH become more negatively charged. Most NF membranes are made of polymers such as polysulfone, polyvinyl chloride whose functional groups may

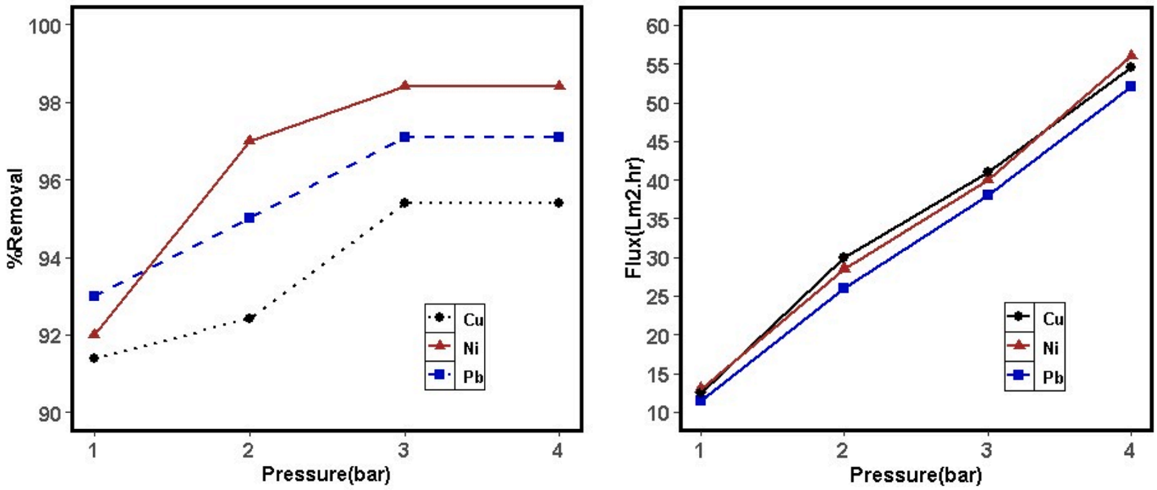

Fig. 3. Effect of pressure on (a)heavy metal removal% (b)permeate flux for RO (Concentration = 100 ppm, Temperature = 30 °C, pH = 5.5, and Feed Flow rate = 30 L/hr) [76].

be amino acids, esters and carboxylic acids. When feed solution passes through the membrane, there is a disassociation of these groups causing a charge to the surface of the membrane. For optimum results especially with wastewater which has a low pH, the rejection of divalent cations such as copper, cobalt, zinc increases because of the repulsion force between the cations and the positively charged membrane surfaces [29].

Utilising membrane technology for mining wastewater management for the removal of heavy metals

The mining sector contributes significantly to the ongoing pollution of freshwater resources by releasing untreated or partially treated effluents particularly mine tails laden with heavy metals and other pollutants [90]. Heavy metals are elements with an atomic weight between 63.5 and 200.6 g with a specific gravity higher than 5 [91,92]. These elements can be categorised into three groups: toxic metals (including Pb, Co, Hg, Cr, Cu, Cd, As, Sn, Ni), radioactive metals (Th, Am, Ra, U) and precious metals (Ru, Pd, Ag, Au, Pt). Membrane technology has emerged as one of the most efficient methods of treating mine effluents to remove heavy metals. Studies have reported the excellency in mine wastewater treatment through the usage of various types of NF and RO.

Al-zoubi, [93] used three commercial NF membranes (NF99, DK and GE) to treat to AMD from a copper mine in Chile. The membrane's performance was assessed for the rejection Cu, Fe, Mn, Ca, Mg, Al, and SO_4, as well as the monovalent cation of sodium (Na). Remarkably, the NF99 membrane achieved a high rejection rate exceeding 99 % for all metal ions, and permeate concentration of these species met World Health Organisation (WHO) standards for potable water. Algureiri and Abdulmajeed [76] conducted an evaluation of operating conditions such as temperature, pressure, pH, concentration and flow rates on the removal of three metal ions (Cu^{2+}, Ni^{2+}, Pb^{2+}) from synthetic wastewater using RO membrane. Rejection rates of 96 %, 98 % and 97.5 % were reported for the respective metal ions. The authors further observed that higher concentration led to a reduction permeate flux due to an increase in the concentration of negatively charged shielding, resulting in heightened attraction forces between the positively charged ions and the membrane. Hence, causing an increase in the concentration polarization. Similar results were also observed by Cséfalvay et al. [94], the rejection rates of Ni (II), Pb (II), and Cu (II) at various concentrations and observed a 97 % average rejection rate for all three heavy metals.

Mine water contains sulphate, acids and different metal ions which are detrimental to the environment if not sufficiently treated. At laboratory scale Rieger et al. [60] investigated the treatment of very concentrated AMD at low pH using nanofiltration (NF99) and reverse osmosis (RO 98pHt). The results showed an overall rejection of about 80.3 and 91 % for NF 99 while a 91 and 95 % rejection for RO98pHt with an applied transmembrane pressure of between 10 and 30 bar Lower scaling (accumulation of inorganic salts such as $Mg^{2+}$and Ca^{2+}) and less energy consumption makes NF more feasible than RO especially in treating highly concentrated AMD mine wastewater. Recently, Ambiado et al. [21] in a pilot study evaluated the treatment of AMD from gold mining with a focus of sulphate removal using the same membranes. Transmembrane pressure of between 4 –50 bar for RO98pHt and 4 - 40 bar NF 99 at temperature $25 \pm 1\ ^\circ C$ was used. At optimal pressure of 15 bar, the total ion rejection was 92 % for NF and 98 % for RO while sulphate rejection was 97 % for NF and 99 % for RO. These results indicate an increase in the rejection of the metal ions from the previous study by [63] at optimal pressure of 20 bar The configuration of the membrane in a spiral wound geometric configuration proved relevant to the better performance of this system. The membrane configuration plays a vital role in the performance of the membrane especially in removal of metal ions from contaminated mine wastewater.

Wadekar and Vidic [95] evaluated the performance of a polymeric nanofiltration membrane in comparison with a ceramic membrane for the treatment of AMD from an abandoned coal mine in Pennsylvania. The polymeric membrane exhibited higher rejection for metal ions than the ceramic membrane. Multivalent ion species rejection was greater than 96 % for the polymeric membrane whereas the ceramic membrane achieved rejections between 55 and 67 %. However, these membranes did not effectively remove arsenic from the AMD until an antiscalant was added to the feed water causing an increase in the removal from 114 to 200 %. The study showed the possibility of treating AMD to standards of drinking water especially when an antiscalant is added to the feed water which reduces scaling and prolongs the life span of the membrane. However, this can increase the cost of operation especially for the cost of the antiscalant.

Pino et al. [74] studied the performance of spiral-wound membranes (NF270 and NF90) in treating acid mine drainage from an active copper mine and three metal ions were studied (Cu, Al, Zn). NF90 showed high rejection than the NF270 prone to fouling. The results showed high rejection for Al and the lowest rejection was observed for Cu. The authors concluded that the results observed were a result of the hydration mechanism and steric hindrance and electrostatic forces which were similar to the finding of [96,97]. These two types of separation mechanism are the most well established separation technique which the metal ions are separated from the wastewater.

Lopez et al. [97] examined the performance of the semi-aromatic polyamide NF270 and a polyethersulfoned (HydraCoRe 70pHT) nanofiltration membranes for treating acid mine drainage (AMD) for metal removal and recovery of sulphuric acid. The NF270 membrane showed a 90 % rejection rate for sulphate and for metals ions like Zn^{2+}, Cu^{2+} and Fe^{2+}, while HydraCoRe 70pHT achieved a 75 % rejection rate. This difference was attributed to the membrane charge with NF270 exhibiting positive charge, which improved metal ion rejection compared to the negatively charged HydraCoRe membrane, which suffered from higher concentration polarization. Both membranes effectively collected dilute hydrochloric acid, though with some iron impurities. This study highlights the economic potential of using membrane filtration in mining, especially within a circular economy era, in which AMD represents a sources for resources products such as water, metals and reagents such as sulphuric acids which can be reclaimed and recycled.

To better understand the long term performance of membranes in acidic conditions [98] exposed six commercial nanofiltration membranes to acidic wastewater from the copper refining plant. After 30 days the permeation and rejection completely stopped. This is because overexposure to acidic wastewater without cleaning caused clogging of the pores of the membranes and eventually resulted into blockage of the pores with the solutes. It is therefore imperative that membranes are cleaned to avoid such problems and another way is the introduction of nanomaterials. Table 3 below shows summarises the various types of membranes and their efficiency in removing heavy metals.

The recent advancement in nanotechnology has opened opportunities for a way to reduce or curb membrane fouling through different strategies. Furthermore, there is need to utilize available nanomaterials to produce highly selective, fouling resistant and higher water permeable membrane. The following section discusses the recent advancements made towards the improvement of pressure driven membranes for improved waste water treatment.

Recovery of heavy metals and rare earth metals from AMD

Mining processes results in the production of large volumes of mine wastewater such as tailings, process water, leachate and AMD etc. AMD is one of the significant challenges facing the mining sector around the global intensified by the presence of waste rock, tailings and open pit. Exposure of sulphide minerals such as pyrite and chalcopyrite to water,air and microbial activities causes AMD, which is a severe environmental challenge [21]. AMD is dark brown reddish appearance is characterized by low pH between 2 and 4, high conductivity, elevated concentrations of toxic metals such as Cu, Cu, Zn and Al, with predominately high sulphate and iron ions which degrade the quality of water as well as sediments near the mining site. The severity in environmental damage of AMD is the

Table 3
NF and RO for eliminating metal ions from various mine effluents NF and RO for eliminating metal ions from various mine effluents.

Membrane process	Membrane type	Type of mine wastewater treated	Removal efficiency (%)	Parameters	Reference
Nanofiltration	Semi-aromatic polyamide(NF270)	Synthetic mine water	Cu^{2+}:100 Pb^{2+}:74 Cd^{2+}:99 Mn^{2+}:89	pH = 1.5–5 Pressure = 4 bar Concentration =1000 mg/L	[99]
Nanofiltration	Semi-aromatic polyamide(NF 270) Sulfonated polyether sulfone active layer(HydraCoRe 70pHT)	Synthetic acidic mine wastewater	Zn^{2+},Cu^{2+},Fe^{2+}rejection >90 for NF 270 Zn^{2+}, Cu^{2+}, Fe^{2+}rejection around 75 for (HydraCoRe 70pHT)	pH = 2.0–2.8 Pressure=8.35 −19.7 bar	[97]
Nanofiltration	(Semi-aromatic polyamide) NF 270	Acid mine drainage	Metal rejection (Cu^{2+}, Zn^{2+}>98 %	Pressure = 3 bar pH = 1	[100]
Nanofiltration	NF90 (thin film aromatic polyamide) NF 270 (Thin film Semi-aromatic polyamide)	Metal plating	Ni^{2+}:99.2 Cr^{6+}:96.5 Ni^{2+}:98.7 Cr^{6+}:95.7	pH = 10 TMP = 30 bar	[101]
NF 90 Nanofiltration	Polymeric membranes	Synthetic mining water	Pd:>99	Concentration = 20–100 ppm TMP = 5,10,15,20 Pressure = 25 bar Model =dead-end module	[102]
Nanofiltration	NF 99(Thin film composite polyester)	Acid Mine Drainage	Cu^{2+}: 99.9 Fe^{2+}:99.9 Mn^{2+}:99.9	Model =cross-flow system Pressure = 20 and 30 bar Concentration = 1x and 2x*	[93]
Nanofiltration and Reverse Osmosis	Polymeric membranes	AMD from copper mining	Zn, Cu, Al, As, Mn, Fe rejection was 92 % for NF,98 % for RO	For RO (Temp = 25 ± 1 °C Pressure =−50 bar Velocity flow=700–1150 l/h) For NF Temp = 25 ± 1 °C Pressure =4–40 bar	[22]
NF (membrane A)	Thin film composite PAN (polyacrylonitrile)	Synthetic AMD	Cu^{2+}, Mn^{2+}, Mg $^{2+}$: rejection >96	Temp = 25 °C Pressure = 10 bar Time = 1 hr pH = 2–7	[103]
Reserve Osmosis	Polymeric membrane	Industrial wastewater	Cu^{2+}:96 Ni^{2+}:98.5 Pb^{2+}:97.5	Concentration =50–200 ppm, Pressure = 1–4 bar Temperature = 10–40 °C pH = 2–5.5 Flowrate =10 to 40 L/hr	[76]

reason some researchers have described it as toxic [104,105]. Fig. 4 below shows the role of membrane hybrid system in sustainable mine wastewater management.

In recent years with the coming of circular economy concept and a shift in paradigm, waste streams have been considered as potential sources of metals and other by-products such as water due to the high cost of mineral exploration and reduction in ore grade [106]. AMD has been explored as a secondary sources for the recovery of valuable resources such as water, sulphuric acid and heavy metals using various methods [107].

Membrane processes have been studied for the purpose of recovery of these valuable resources from mine waste streams such as AMD [108,109]. However, these individual membrane processes have major drawbacks such as high operational costs and the production of a concentrate laden with potential resources such as sulphates, metal ions, REEs and precious Earth metals which may cause potential source of pollution if not managed properly. Therefore, an advanced hybrid system integrating membrane processes with other treatment processes enhances the recovery of more water and metal ions which can be reused for industrial purposes such as washing of the ores, irrigation and as portable water [106,110].

Recovery of water and valuable metal from AMD

Nanofiltration membranes have reported excellent results in water recovery from AMD. Fonseka et al. [106] employed a low pressure nanofiltration membrane with chromium modified metal organic framework(Cr-MIL-PMIDA) at steady permeate of 15.5 LMH and pressure of 3 bar to achieve 80 % water recovery while achieving greater than 98 % solute rejection [19,111]. Furthermore, about 90 % of the copper from the stimulated AMD was recovered.

Metal recovery by pressure driven membrane in combination with other treatment processes has shown excellent results. Recently, Pino et al. [27] employed nanofiltration with NF 270 membrane coupled with solvent extraction for the selective recovery of water and copper from AMD collected from Chile. The system achieved a 97 % recovery of copper from 1.1 g/L copper retentiate and 80 % water recovery combined nanofiltration with solvent extraction to recover copper from acid mine drainage. However, the gypsum (Ca_2SO_4) scaling reduced the efficiency of the membrane to recover more water and concentrate more copper for recovery but it not affects the overall extraction process. Metal sulphide precipitation and microfiltration membrane process demonstrated higher recovery of copper close to 100 % and turbidity was lower than 2 NTU for the sulphide stoichiometric dosage of 120 % synthetic AMD treated using this system [23]. The flux was greater than 0.1 L/m^{2}s, with the microfiltration membrane acting as a clarifying component of the system.

Other membrane system such as the thermally driven membrane distillation (MD) have also been explored for their ability to recover valuable resources from AMD. Ryu et al., (2019) demonstrated the feasibility of a submerged MD integrated with Zeolite as a way of preventing scaling and membrane fouling, 50 % high quality fresh water recovery from AMD was recorded in 30 h of operation. Furthermore, almost 100 % removal of Fe and Al was achieved with this system when pH was adjusted from 2 to 4 [105]. To this end, MD demonstrated its ability to recover up to 90 % of water due to its low sensitivity to solute concentration and composition [112, 113].

A two steps forward osmosis(FO) with sodium chloride (NaCl) as the draw solution achieved 80 % water recovery from AMD. As the concentration of the direct solution(DS) increased from 0.5 M to 2.5 M the water recovery increased from 50 % to 80 % respectively

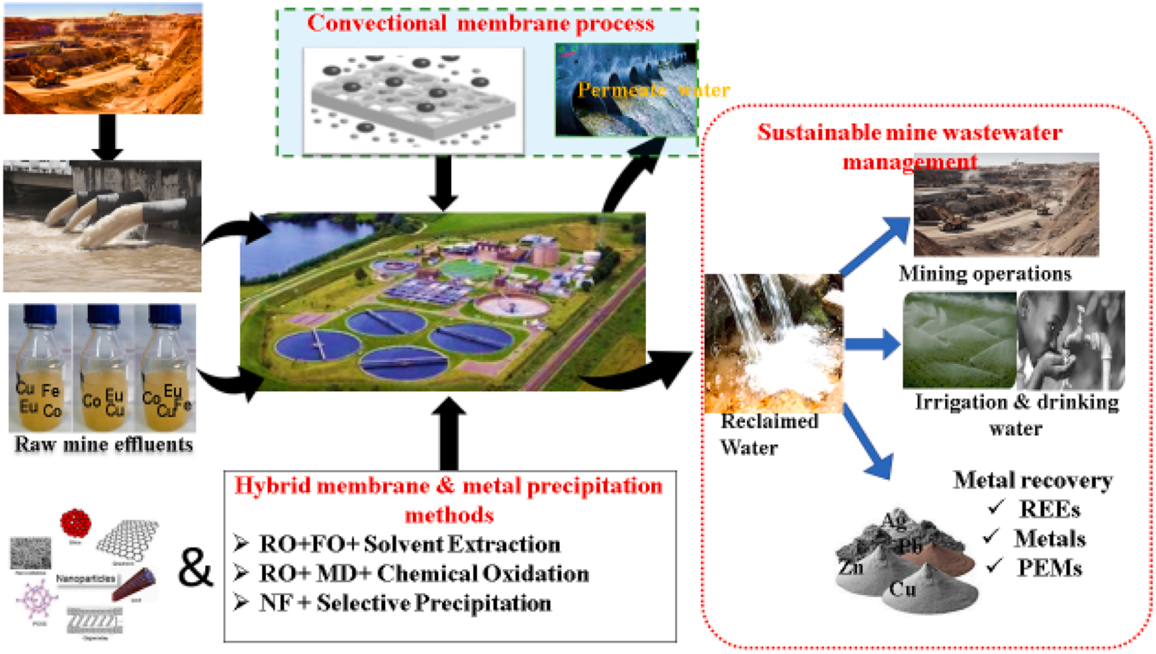

Fig. 4. Sustainable mine wastewater treatment towards a circular economy.

[114]. In this system, the concentration of the draw solution plays a critical role in the increment of the flux [54].

Despite these successful metal recovery studies from AMD, the metals recovered are not sufficient to add more value to the mining process. This is because these metals exist concentration that are not sufficient to recover in large quantities. Against this background, researchers have explored other alternative to recover metals. Lithium –ion spent batteries have emerged as another source for metals such as Li,Ni and Cu [34]. Li, Co, Mn and Ni were recovered from used lithium ion batteries by employing nanofiltration membrane in a crossflow mode. Metal rejection greater than 92 % was achieved for the metal ions with a permeate flux of 7.5 L/m^2·h. the system was. Further coupled with crystallization to recover the metal ions, over 90 % recovery was achieved for Mn,Co and Ni while 89 % of Li was recovered with 99 %purity when 4 M K_2CO_3. This study underscored the ultization of other resources seen as waste for recovery of metal ions [115].

Removal and recovery of rare earth elements (REE)from AMD

The demand for REEs has increased over the years due to their potential application in the modern fourth industrial revolution characterized by digital high technology. REE are the 17 elements on the periodic table with similar physical and chemical properties like malleable melting point and boiling including 15 lanthanides, yttrium and scandium [17]. REEs can generally be classified into two groups based on their electron configuration; the light rare earth elements (La, Ce, Pr, Nd, Pm, Sm, Eu), and the heavy rare earth elements (Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Y). Globally, china is the world's highest exporter of REEs. REEs are abundant in the earth's crust. However, their rare character is due to their inability to exist in large quantities independently. Therefore, the recovery of such elements from AMD can significantly help in meeting the increased demand of REEs especially in high technology industrials. Studies have indicated higher concentration of REEs in AMD [113,116].

Despite the various techniques employed for the recovery of REEs such as chemical precipitation, ion-exchange, adsorption, membrane filtration and ion flotation, individually these methods lack selectivity because of the characteristics of AMD. Though adsorption and ion-exchange are the most preferred methods of recovery of REEs from AMD, these have their own limitations. Integrated system of recovery were such methods are paired with membrane filtration are efficient as they not only recovery REEs but also recover other useful resources such as water [117].

The role of membrane technology in recovery of REEs is to concentrate the REEs in aqueous solution for selective extraction using selective adsorption. For this purpose membrane processes such as MD, FO and RO have been extensively employed (R. Kumar et al., 2023).For example, a FO membrane was used to recover REEs (lanthanum, Dysprosium and cerium) from stimulated AMD. The pH, temperature and membrane orientation played a critical role in the role in the recovery process. Higher water flux was reported when the active layer(AL) faces the DS (AL-DS) in comparison to when the active layer was facing the feed solution(Al-FS). REEs rejection using the Al-DS was 85 % while for Al-FS rejection was 91 %. Due to the internal concentration polarization which had a lower impact on the osmatic pressure gradient, low REEs rejections were observed in Al-DS orientation. The molecular weight, shapes and crystal structure of the REEs influenced their difference in rejection by the FO membrane. Higher rejections of REEs was noted when the FS and DS temperature was 20 °C as a result of increased electrostatic repulsion between the REEs and the membrane molecules. Additionally, the rejection of dysprosium was pH dependent. At varying pH, the rejection was different which would be attributed to membrane surface charge and different forms of dysprosium. Nevertheless, the rejection efficiency of Lanthanum and cerium remained steady [118].

An ion imprinted membrane have gained attention in recent research as they are able to selectively separate metals of interest from their concentrate. A study by [119] used an ion-imprinted membrane modified with chitosan to selectively recover Neodymium(Nd) and Yttrium(Y) from artificial coal fly ash extracts. The synthesized membrane showed higher retention of the two metals of 90.11 % and 80.95 % respectively.

A mixed membrane modified with highly europium selective metal organic framework, Cr-MIL-PMIDA embedded in the sulfonated poly (ether ketone) (SPEK) polymer membrane matrix to preferentially concentrate europium ($Eu^{3+)}$ ions in the presence of other competing cations. The activated membrane notably reduced ionic conductivity for Eu^{3+} compared to other multivalent ions. Membrane extraction experiments further confirmed the selective behaviour, demonstrating slower diffusion for Eu^{3+} compared to Mg^{2+} and Zn^{2+} cations. Especially, at pH 5, Mg^{2+} and Zn^{2+} recovery was greater than 30 %, whereas Eu^{3+} recovery remained lower than 4 % [120]. In a study by Kumar et al. [104] precious metals can be recovered through membrane system. Molybdenum, a precious element used in photocatalytics, lithium batteries and electronics was recovered from industrial wastewater using a hybrid membrane system consisting of UF and NF operated in recirculation. NF coupled with chemical precipitation when operated for 15.7 h at pH (1.7) and temperature (62 °C), 98.7 % of the molybdenum was recovered as ammonium molybdate (NH_4^+/Mo).(Kumar et al., 2023).

MD is effective in concentrating metal ions for recovery of REEs from AMD. In a study by Fonseka et al. [121], a hybrid system of a highly efficient adsorbent SBA15-NH-PMIDA adsorbent and direct contact membrane distillation (DCMD) was used to selectively recover Europium(Eu) from real AMD. The authors successfully demonstrated a two-step process combining adsorption and MD for Eu recovery. The adsorbent achieved an 80 % adsorption rate for the Eu when the pH was adjusted. Eu recovery exceeded 90 % for the DCMD system. Operation of the DCMD for 11.8 h yielded water recovery of 80 %. Such system offers greater benefits especially in addressing the adverse environmental impact of AMD.

Recent advancement made towards improvement of membranes

Recent advancement in membrane modification of membrane are centred on the philosophy of improving selectivity, and mitigating membrane fouling, the major drawback the utilization of membranes for mine wastewater treatment. Membrane fouling reduces the efficiency of the membrane and increase the operational costs, arises from the accumulation of solutes on the membrane surface causing the pores to constrict, and subsequently, reduction in water flux as well as the quality of the permeate [59]. To address this challenge, various strategies have been employed including pre-treatment methods, optimization of operating conditions and membrane modification approach. Herein, this section discusses in depth these some of these strategies

Fabrication techniques for modified membranes

The synthesis methods play a critical role in determining the physical properties of the membrane particularly the morphology, which significantly impact the performance of the membrane especially in wastewater treatment. In comparison to other strategies, membrane modification approaches are preferred as only small changes can be made to the membrane structure [59]. This sub-section explores in detail different fabrication methods that have been used to produce modified membranes. As shown in Fig. 5, there are different techniques that can be used to synthesize and modify membranes.

Lithography

Lithography is one of the methods used to synthesize membranes. In this process, the driving force for the use of this method is the desire to produce membranes with small features but excellent resolution [122]. Lithography is grouped into two types: masked and unmasked. In unmask lithography, patterns are inscribed directly onto the foundational material, whereas in masked lithography, the patterns are transferred onto the material [123]. Photolithography and nanoimprint are examples of mask lithography. Photo lithography can further be divided into Contact printing, proximity printing and projection printing, with the ability to produce features in the range of 2;3 nm [124,125]. Unmasked lithography include the electron lithography which involves the use a hard mold to imprint into a polymer film for nanoscale printing. [126] used lithography to produce isoporous polyvinylidene fluoride (PVDF) membranes for industrial separation and purifications processes. The membrane exhibited high water flux and the pore size were 100 nm and 20 nm.

Sol-gel process

The sol-gel process is wet method utilized in the production of inorganic/ceramic materials like metal oxides/ polymers and

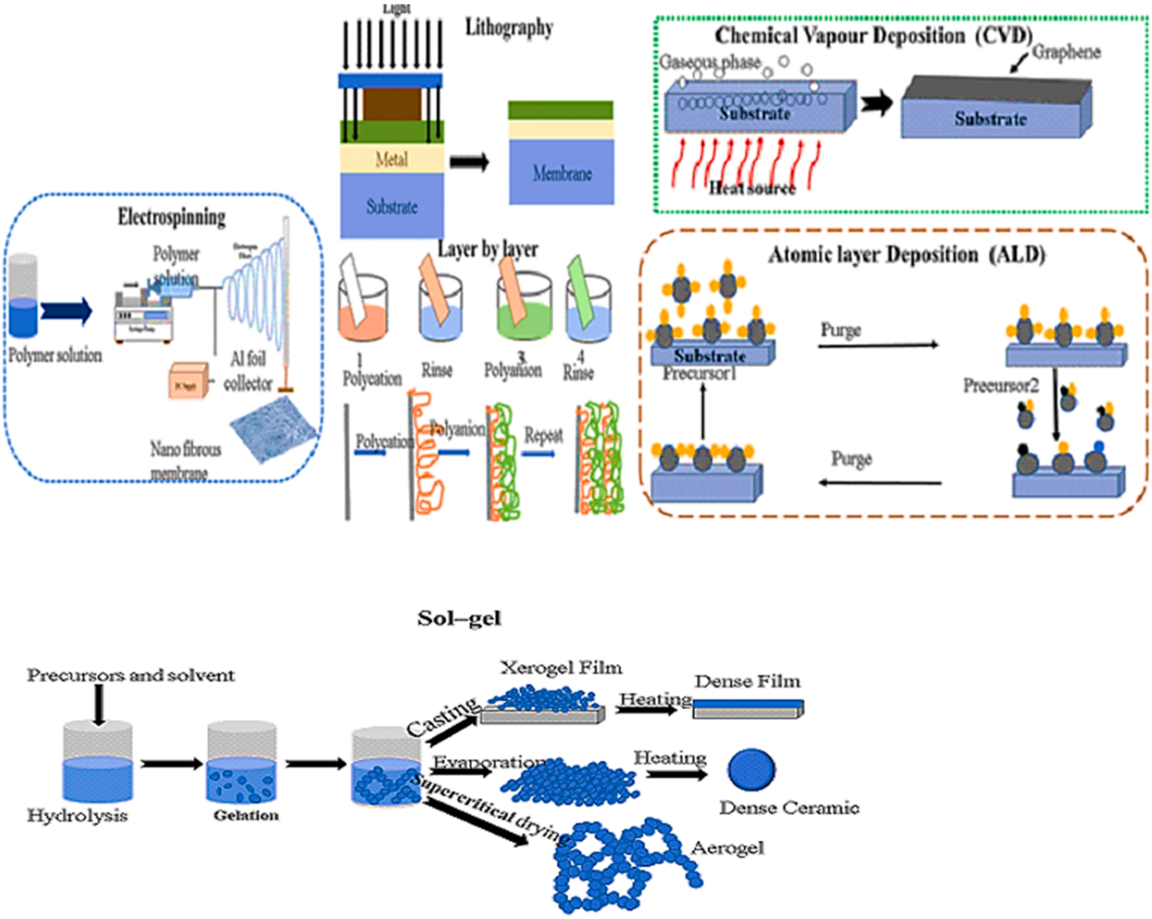

Fig. 5. Synthesis and fabrication techniques for modified membranes.

membranes through the transformation of liquid precursors to a sol-colloidal suspension [127]. There are four stages involved in the process of sol-gel; first the homogeneous solution with salt or a metal must be prepared which is also the precursor (metal alkoxide). During the second stage, the concentration of the homogeneous solution is increased. The third stage involves the formation of a gel by condensation. Finally, depending on the application of the material the gel is dried. Structure with excellent optical, mechanical, electrical and thermal properties can be obtained by using this method. The advantages of this methods that its cost-effective, uses simple equipment, highly controllable synthesis and low temperature chemistry. In addition, modified sol-gel methods such as ultra-sonication, precipitation and aerogel are equally used. These modified methods can improve the mechanical and porosity properties of the membranes [122,128]. Cu(II) ions was successfully selectively separated from an aqueous solution by using an ion-imprinted silica nanotube membrane synthesized using the sol-gel method.

Chemical vapour deposition (CVD)

Chemical vapour deposition is a fabrication method used to produce thin film structures. It is used to synthesise carbon nanomaterials and is considered as a cost effective method because the process can be conducted at low temperatures. This technique ensures control of both the morphology and the structure of the nanomaterial to be produced. The process involves the formation of a solid heater resulting from a chemical reaction in the vapour phase. At higher temperature, the precursor decomposes and the resultant gaseous atoms are absorbed and deposited on the substrate during the chemical vapour deposition process. Catalytic CVD was employed in the production of a vertically aligned CNT composite, with spaces between them filled by fluorocarbon polymer films. The produced membrane exhibited uniform distribution. Furthermore, the deposition of the fluorocarbon film polymer on the surface of the walls of the MWCNTs did not affect the alignment of the membranes. Defect–free deposition of the polymer was achieved without distracting the carbon nanotube alignment [129]. Therefore, CVD can be used to fabricate nanomembranes that can be used for the treatment of mine effluents.

Layer-by-layer

In this method, membranes are produced by the adsorption of molecules with different charges. Initially, the substrate, for instance, is submerged in a cationic polyelectrolyte solution. The polyelectrolyte is adsorbed in a single monomolecular sheet with a thickness of approximately 1 nm [123]. New ion exchange membranes have been synthetized using a layer-by-layer coating method through chemical crosslinking. The interactions between the layers involve various types of bonds, such as hydrogen, ionic, and covalent bonds [130]. By utilizing glutaraldehyde as a crosslinking reagent, a cationic membrane was assembled layer by layer. This approach improved the adhesion between the layers. In comparison to the pristine PVC membrane, the prepared membrane showed a smoother and more hydrophilic surface, which helped the fouled membrane regenerate effectively with ultrasonic waves. The electrodialysis tests also showed that the prepared membrane was capable of effectively removing heavy metal ions, such as copper, nickel, and lead. On the surface of ion exchange membranes, hydrogels can be used as modifying agents to enhance ionic interactions and regulate ion transport [131].

Electrospinning

Electrospinning is an electro hydrodynamic fabrication method used to manufacture ultrathin nanofibers membranes from the spinning of the polymeric dope using electric field and is considered to be one of the best methods for creating continuous nanomaterials with diverse biological, chemical and physical properties. With this method, it is possible to synthesize membrane with diameter from 2 nm to several nanometres. This method depends solely on the viscoelastic qualities of a high voltage solution to generate and grow a single jet on a collector [132,133]. Different types of polymers are used in the electrospinning process such as natural and synthetic polymers as well as co-polymers. Natural polymers such as chitosan while synthetic polymers include PVA, polyamide, polyoctide, polyglycolide, polystyrene, polyethylene have been explored in the synthesis. Furthermore, co-polymers such as a combination of the natural and synthetic polymers such as gelatine/poluethyleneoxide blend, polycaprolactone–poly-l-lactic acid blend and chitosan-polyethylene oxide [134]. One of the major advantages of this fabrication method is the formation of membranes that can operate at low pressure or gravitational force, hereby, use less energy. Furthermore, electrospun membranes are characterised by the formation of nanofiber membranes that are uniformed and have controllable morphologies. Different parameters that affect the electrospinning process such as the voltage and the flow rate. The effects of parameters such as voltage and flow rate on electrospinning of nanofibers are well investigated by [135]. The authors noted that with an increase in the voltage applied, the diameter of the of the electrospun nanofibers increased while increasing the flow rate decreased the diameter of the fibre electrospun. In a review by [134] parameters such as polymer concentration, molecular weight of the polymer, feed rate and the temperature were identified as important for the morphology of the electrospun nanofiber.

Atomic layer deposition

ALD is a fabrication method used to synthesize highly precise uniform thin film membranes or coating on the substrate layer with atomic level control. This technique is compactable with a broad range of substrates and produces a homogeneous coating over the uneven support that not only keeps the original pore structure but also reaches the complete coverage. The process of ALD is a self-limiting reaction happens when the precursor (polymer) reacts with a substrate to form a monolayer and the reaction stops once the substrate has been used up [136,137].

Membrane technology based on polymer nanocomposites for treating mine effluent

In comparison to their traditional counterparts, polymeric nanocomposite membranes have improved mechanical, thermal, chemical properties, antifouling properties and improved surface properties. Subsequently, leading to improved rejection of metal ions sand water flux [138].Polymer nanocomposite membranes are formed when nanomaterials such as nanoparticles, nanotubes, and nanosheets are incorporated into the porous polymer intermediate/substrate or support of the membrane. Polymeric nanocomposite membranes can be classified as thin film nanocomposite and blended or mix matrix membrane [139]. Polymeric nanocomposite membranes utilizes the benefits of nanomaterials and polymers to enhance the functions of membranes [140]. Hence, can be used in several environmental applications such as the treatment of mine effluents to remove metal ions [141].

Ahmed et al. [142] observed an increased adsorption of heavy metals to the chitosan when CS/PVA blended graphene oxide with gum resin and silver nanoparticles for the removal of heavy metals. Metal rejection above 95 % for all investigated heavy metals was reported. Ghaemi, (2015) examined the removal of copper ions using a polymeric PES embedded with γ-alumina nanoparticles at varying concentrations of 0.1,0.01 and 1 wt.%. Increasing the γ-alumina nanoparticles concentration from 0.01to 0.1 wt%, increased the Cu^{2+}rejection. This enhancement was attributed to the smaller size nanoparticle which facilitated a larger surface area for adsorption of Cu^{2+}.Furthermore, incorporation of these nanoparticles increased the hydrophilicity of the membrane leading to a higher water flux. This study underscored the play that surface area as well as the size of the nanoparticles incorporated play in the effective removal of metal ions from wastewater even in nanocomposite membranes.

Polymeric membranes have been used in the treatment of wastewater from various industries. However, one of the drawbacks has been the accumulation of the bacteria on the surface of the membrane resulting into a biofilm which reduces the selectivity and permeability of the membrane. In reducing such inefficiency, nanomaterials such as silver, titanium dioxide, zinc oxide and carbon nanotubes have been explored as nanofillers in this area [143]. A silver nanocomposite polymer inclusion membrane was used in removing cadmium, cobalt, nickel and copper from both synthetic and real wastewaters near a mine tailings dam by varying the concentrations of the Ag nanoparticles. The authors observed that the increase in silver concentration improved the hydrophilicity of the membrane and an increase in the anti-fouling properties of the membrane. Furthermore, the rejection of the metal ions was in the order $Cd^{2+}>Cu^{2+}>Ni^{2+}>Co^{2+}$ despite the ions ionic radii been nearly the same. This trend is attributed to the membrane affinity towards Cd^{2+}because this ion can easily shed off the hydration shell to remain with a hydrophobic shell. Owing to its lower hydration energy than the other ions [144]. In another study, silver nanocomposite membranes were synthesized by modification of PVDF and PES with silver nanoparticles by physical vapour deposition. After 1200 min filtration time, the PVDF showed a smooth flux reduction slope and better bio-fouling slope than the unmodified membrane. Silver nanoparticles proved their ability to increase the anti-bacterial activities in wastewater [145].

The ZnO nanoparticles have been reported in literature to have enhancing hydrophilicity properties, porosity, and high permeability of the membranes modified with ZnO due to their anti-bacterial, anti-fungal and anti-corrosive properties [146]. The anti-fouling ability of the PVF ultrafiltration membrane enhanced when Fan and the group modified it with Poly(vinyl) Chloride. The hydrophilicity of the membrane was increased resulting into an increase in the anti-fouling abilities [83].

Among the available nanoparticles, Titanium dioxide (TiO_2) has attracted the attention due to its chemical stability, low cost, availability and non-toxicity especially in water [84]. Furthermore, under UV radiation TiO_2 becomes photocatalytic producing free radicals that exhibit anti-bacterial and anti-viral properties against many microorganisms such as Gram negative and positive bacteria [147]. This is because of the large band gap, making it able to absorb UV radiation [148]. Because of these properties, the introduction of TiO_2 NPs in the membrane enhances the anti-biofouling the membrane leading to an increased water flux of the membrane especially in wastewater treatment [149].

In membrane separation, TiO_2 has been utilized because of its ability to withstand high temperatures as well as work at room temperature. In addition, the physicochemical properties can be altered to a greater extent when the size of the NPs reduces.TiO_2 has been reported as an effective photo catalyst in removing heavy metals from wastewater through the reduction of the heavy metals to lower oxidative states [150]. This degradation lies in the absorption of energies higher than the semi-conductor band gap resulting in the excitation of the electrons from the valence band to the conduction band.

Wanjale et al. [151] fabricated a polyester incorporated TiO_2 composite nanofiber membranes for the removal of Cu^{2+} from water. It was observed that the removal of Cu^{2+} increased and this was attributed to the effective adsorption nature of TiO_2 causing a deep wettability modification of the membrane. Hereby, increasing the rejection of the Cu^{2+} ions from the water. Ultrafiltration membranes synthesised from polysulfone with TiO_2 nanocomposites fibres exhibited higher water flux than the unmodified without TiO_2 [152]. The authors attributed this to the increase in hydrophilicity and wettability of the membrane due to the increase in the concentration of the TiO_2 nanoparticles.

An adsorptive membrane prepared by the modification of Polyether sulfone (PES) with TiO_2 as an adsorbent and Polyaniline (PANi) as a modifier was used in the removal of Cu(II) ions. The authors observed an increase in the pure water flux from 50 kg/m^2.h. bar with the pure PES to 60.5 kg/m^{2}.h. bar for the modified PES/PANi-TiO_2. Furthermore, because of the high adsorption capacity of the PANi-TiO_2 rejection of the Cu(II) increased from 54.5 to 94 % when the adsorbent loading was increased from 3 to 5 wt.%. Nevertheless, at 7 wt.% TiO_2 loading, membrane agglomeration occurred reducing the rejection capacity of the Cu(II) ions to 71 % [153]. Thus, the amount of the adsorbent loaded in the membrane plays a critical role in the efficiency of the membrane especially in removal of heavy metal ions. Table 4 below summarises the use of different polymer nanocomposite membrane in the elimination of metal ions from wastewater. Polymer nanocomposite membrane exhibit stronger rejection capacity of metal ions than pristine membranes.

Table 4
Recent studies of polymer nanocomposite membranes in removal of metal ions.

Nanocomposite / nanoparticles	Polymer	Membrane processes	Target metals ions	Key findings	Fabrication method	Reference
Ethylenediamine eGO	PA	Hollow fibre ceramic Nanofiltration	Zn^{2+},Cu^{2+},Ni^{2+}, Pb^{2+}	-Excellent removal capacity of the target heavy membranes (Zn^{2+}=93.33 %, Cu^{2+}=92.73 %, Ni^{2+}=90.45 %, and Pb^{2+}=88.35 % -High permeate flux of 12. 96 $L.m^{-2}.h^{-1}$ in the 30 h test for the stimulated mining wastewater	Interfacial polymerization	[154]
Carboxylated-GO	PPSU	Nanofiltration	As,Cd, Pb, and Zn	-Enhanced volumetric flux of 27 ± 3 $L.m^{-2}.h^{-1}$ -Increased heavy metal rejection > 98 %, high hydrophility, Negatively charged membrane	Phase inversion	[155]
Alpha-zirconium nanoparticles	PVDF	Nanofiltration	Cu^{2+}, Pb^{2+}, Ni^{2+} Zn^{2+},Cd^{2+}	-Enhanced removal of metal ions, (Cd^{2+}=42 %, Cu^{2+}=93.1 %, Ni^{2+}=44.4 %, Pb^{2+}=91.2 %, and Zn^{2+}=44.2 %) -Mechanical and thermal stability -Improved anti-fouling properties	Phase inversion	[156]
Graphene Oxide (GO) Zinc Oxide Nanoparticles	PSF	Ultrafiltration	Pb^{2+},Cu^{2+},Cd^{2+}, Fe^{2+}	-GO improved the mechanical properties -Permeate flux=1.65 mL min^{-1} -ZnO enhanced hydrophility, pore size and permeate flux -Highest adsorption was recorded for Pb at 279.68 mg g^{-1}.	Phase inversion	[157]
Al_2O_3 & ZrO_2	Polyamide 6(PA6)	-	Al^{3+},Fe^{3+}	-Al_2O_3 and ZrO_2 were charged -Increased the adsorption capacity (Al=98.6 % &99.3 %) -Selective removal of metal ions	Electrospinning	[158]
$CoFe_2O_4$/CuO nanoparticles	PES	NA Nanofiltration	Pb^{2+}, Ni^{2+}, Cu^{2+}	-Reduced surface roughness from 45 nm to 24 nm -Higher rejection that 88 % for all metal ions (Pb^{2+}: 88 %, Ni^{2+}: 92 %, Cu^{2+}=98 %) -Improved anti-fouling and improved water flux capacity	Phase inversion	[159]
PES /Chitosan	PA	-	Mn^{2+}·$Fe^{2+}$$Mg^{2+}$and Ca^{2+}	-Positively charge membrane Cation rejection (90.4, 88.3, 89.3 and 75.7 % for Mn^{2+}, Fe^{2+} Mg^{2+} and Ca^{2+}) −0.75 wt.% CS increased the rejection of cations more than anions -Improved flux from	Phase inversion	[160]

(*continued on next page*)

Table 4 (*continued*)

Nanocomposite / nanoparticles	Polymer	Membrane processes	Target metals ions	Key findings	Fabrication method	Reference
				56 to 93 L/m^2.h for 1 wt.%		
Ethylenediamine (ED)/MWCNTs	PES	nann Nanofiltration	ZZn, Cd, Mg, Pb Ni,Cu and Ca	-Excellent rejection for heavy metal Zn >90 % -Enhanced Water flux of 80.5 L/m^2.h -Positively charged membrane -enhanced surface chemistry (hydrophilicity, small surface roughness)	Self-assembly	[161]
MWCNTs /PEI	PAN	nan Nanofiber membrane Nanofiber membrane	Pb^{2+},Cu^{2+}	-Enhanced adsorption time Improved removal capacity Pb^{2+}, Cd^{2+}, -Adsorption increased with increased dosage of the nanocomposite	Electrospinning	[162]
PANI	polyphenylsulfone (PPSF) Polyphenylsulfone (PPSF)	Nanofiltration	Cd^{2+},Pb^{2+}	-Improved hydrophilic -Enhanced surface properties (CA≤52) -Improved thermal and mechanical properties -Higher water permeability of 1.8 L/h.m^2/bar -Improved high rejections of 99 % and 95 % toward Pb^{2+} and Cd^{2+}	Blending &Phase inversion	[89]
ZnAL-LDHs(ZnAl-layered double hydroxides)	Polyvinyl butyral (PVB) and polyacrylonitrile (PAN)	adadadadadadAdsorptive membranes	Cd^{2+},Pb^{2+}	-Improved metal ion rejection capacity (Pb^{2+}=97.76 %), (Cd^{2+}=87.68 %) -Improved surface properties (hydrophilicity, roughness)	Phase inversion	[163]

Pre-treatment methods

Pre-treatment of the effluents stands as a key strategy in mitigating membrane fouling and energy consumption. According to a review by Ezugbe and Rathilal [37] pre-treating the feed solution before membrane filtration preconditions the wastewater by changing the physical, biological and chemical properties, thereby enhancing its suitability for membrane filtration [37]. Mine effluents are often times laden with metals ions,suspended solids and other reagents which increases the conductivity as well as the fouling in membrane. Because of the high suspended solids and dissolved ions, viscocity tends to be high which results in a high concentration polarization. Consequently, reducing the rejection rate for solute. Furthermore, pretreatments helps in the removal of micrbes like bacteria, algae etc. which would cause organic fouling, limiting the eficiency of membrane systems. Hence, it is imperative that wastewater is pre-treated before membrane filtration with either a low pressure membrane or other conventional methods such as adorption [164].

In the treatment of mine wastewater from an Austrialian mine, Mohsen et al. [165] studied the efficiency of RO as a post treatment method after the pretreatment of the feed water by flocculation, coagulation and sedimentation. The results obtained from the period of 2015 to 2018 showed an improved reduction in the turbidity, Zn, Ni, As and Fe of more than 50 % in the permeate. The efficiencyof the RO system is dependent on the feed quality after pretreatment.

Although these methods prove effective, in contrast, adsorption stands out as the most efficient pretreatment method for membrane filtration due to its removal capacity, cost effectiveness, and environmentally friendliness. Matebese and Moutloali [166] used a system comprised of a flocculation, activated carbon adsorption and finally ultrafiltration. Flocculation played a crucial role in the removal of colour, turbidity while the activated facilitated the adsorption of the metals ions. the overall system showed an increased removal of metals of between 80 and 99.9 %. Many adsorption methods have been used such as batch continuous, mixed tank, packed and fluidized bed absorbers. Among these, the packed and fluidized bed reactors have emerged as potent methods for the removal of metal ions from mine wastewater [167] (Fig. 6).

Hybrid membrane system

Hybrid membrane system(HMS) represents one of the most efficient way to curbing membrane fouling. This system integrates membrane with other existing membranes process or with other bio-physicochemical methods such as coagulation, ion-exchange, adsorption and bioreactors [168]. HMS enhances the removal of organic and inorganic pollutants such as heavy metals, making it potentially suitable for the treatment of mine wastewater. However, the efficiency of this system is dependent on the type of materials used, the dose of the material and the membrane process. As the limitation of one system is offset by the benefits of the alternative method.

Osmotically- driven membrane like forward osmosis is one of the emerging membranes process based on the movement of water molecules from a low feed solution to a more concentrated draw solution. This process uses osmotic pressure without the use of hydraulic pressure for water purification [54]. In mine wastewater treatment, FO has shown potential for the recovery of water and the selective extraction of heavy metals as well as the recovery of heavy metals and REEs due to its low energy consumption, high flux and low fouling [17]. Mine wastewater has been seen as economically valuable resource Despite the many advantages of this system, FO is limited as it does not directly remove heavy metals from AMD; instead just leads to the dilution of the DS and the concentration of the feed solution Carmona and Abejón[125]. Therefore, FO system must be combined with other treatment system to achieve greater metal rejection. The selection and design of the membrane and the draw solution plays a critical role in FO system. Therefore, for efficiency a FO hybrid membrane system have been utilized especially in mine wastewater treatment. The recovery of heavy metals from AMD using a hybrid FO membrane was analysed by [169]. A volume retarded osmosis and Low pressure membrane hybrid process used to recover Mn, Fe, Cu, Zn, As, Cd and Pd from AMD. Poly (sodium 4-styrenesulfonate, PSS-Na) and ethyl- enediamine-tetraacetic acid tetrasodium salt (EDTA-4Na) were used as the DS for the system. Results demonstrated that both PSS-Na and EDTA-4Na were suitable DS in the recovery of metals from AMD because of their negative polarity chelating ligands facilitating an increase in metal adsorption towards them. Complete rejection was observed for Mn, As, Cd and Pd while Zn, Cu and Fe reported 80–85 % rejection. The difference in metal rejection was attributed to the difference in ionization energy and the atomic radius among the metals. The higher the ionization energy of a metal the less likely to form cation or a chelate with the DS. Osmotic pressure of 8.9 showed to be ideal pressure for both volume retarded osmosis –low pressure osmosis (VRL-LPO) and the single FO system for higher water flux and metal rejection [169].

HMS demonstrates high efficiency in water recovery and effective reagent reclamation. Kumar et.al [109] investigated a hybrid FO—NF membrane system for treating coke oven wastewater. Various draw solutions (DS) including calcium chloride ($CaCl_2$), magnesium sulphate($MgSO_4$) and sodium chloride(NaCl) were examined for their water flux performance at varying concentration. Results indicated that increasing the DS concentration led to an increment in volumetric flux, with NaCl as an ideal DS, achieving flux of 46 Lm^2.h at concentration of 1.5 M Furthermore, coupling the system with NF allowed for 99 % of the DS, making this system highly suitable for the DS regeneration, thereby reducing operational cost. Economically, it was estimated that about $1.5/year sufficient to recover about 1000 L of reusable water from this system underscoring this system's cost effectiveness Kumar et.al [109].

Hybrid membrane–adsorption have been explored for their efficiency in wastewater treatment. Loganathan et al. [170] identified three configuration for the adsorption /membrane hybrid system; adsorption followed by membrane filtration, membrane filtration followed by adsorption and finally, were both membrane and adsorption are integrated together. Furthermore, activated carbon is noted for its efficiency, cost effective and availability. Its negative surface charge and highly hydrophobic nature makes it a potent adsorbent in adsorption-membrane hybrid system [171]. Herein, these hybrid system is discussed with a focus on fluidized bed reactors (Fig. 7).

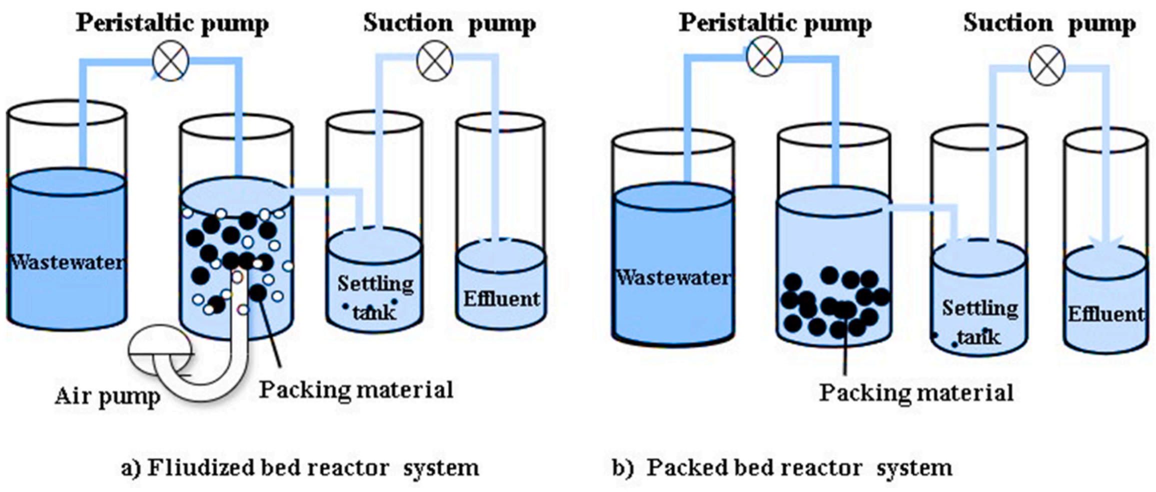

Fig. 6. Fluidized bed reactor and a packed bed reactor. Modified and adopted from [167].

Adsorption-membrane hybrid system- fluidized bed membrane reactor

Many adsorption methods have been used such as batch continuous, mixed tank, packed and fluidized bed absorbers. Among these, the packed and fluidized bed reactors have emerged as potent methods for the removal of metal ions from mine wastewater. FBR offer a superior heat and mass transfer compared to their counterparts packed bed in chemical treatment and mineral separation [172]. FBR system operation are affected by the upward air flow, which suspends the particles forming a fluidized layer with pseudo-fluid characteristics. The performance of such system is dependent upon the pH, the dosage of the adsorbent, initial concentration, temperature and contact time. With such benefits, FBR has been used as together with membrane filtration in a separate system or as a submerged system.

A study by Lv et al. [173] investigated the hydrodynamic and adsorption performance of a fluidized bed containing granular active carbon for removing copper ions from wastewater. The results obtained show that carbon derived from nutshell showed a robust adsorption capacity for copper, facilitated by electrostatic adsorption properties of the oxygen-containing groups within nutshell. The high-frequency contact between the nutshell and the copper ion in the wastewater extended the external diffusion rate, resulting in an enhanced adsorption efficiency of 96 % compared to the 76 % achieved by the packed bed. This investigation highlighted the effectiveness of cost-effective materials such as nutshell in removing metal ions from wastewater and emphasised the potential of such system to be integrated as a pre-treatment method in membrane separation process.

Chang et al. [174] investigated the membrane performance and contaminant removal by a fluidized bed reactor in which a ceramic membrane was submerged and GAC was used as a scouring agent. It was observed that removal of heavy metal ions from real metal plating wastewater is pH dependent. At a pH of 7, 80.7 %, 59.4 %, 48.4 % and 98.3 % were the rejection rates for Ni, Cr, Cu and Zn, respectively. In addition, the fluidized GAC played a crucial role in the reduction of membrane fouling by creating a larger surface area for the adsorption of metal ions prior filtration. 10 % increment of the dosage of GAC resulted in 90 % removal of COD. Furthermore, air bubbles generated by aeration reduce the deposition of solute materials from the wastewater onto the surface of the membrane through air scouring effect of the fluidized GAC.

In another study the effect of adsorbent size was studied, Wu et al. [175] noted that increasing the size of the GAC in the fluidized bed membrane reactor resulted in a reduction membrane fouling. The adsorption capacity of the Granular Activated Carbon (GAC) was saturated within 1hr of the experiment and afterwards the GAC acted as a scouring agent more than an adsorbent. The ratio of hollow fibre spacing to fluidized particle size, dosage of the GAC and the size of the GAC contributed to the reduction of the cake layer. However, it did not reduce the irreversible fouling of the membrane. Johir et al. [176] observed that using GAC size of 300–600 μm increased the membrane resistance to fouling. In their review Devaisy et al. [168] highlighted that hybrid fluidized membrane systems are effective in reducing membrane fouling especially biofouling reduction by usage of minimum energy in comparison to using a single membrane filtration process. However, the limitation of such submerged membrane systems is the formation of a cake layer on the membrane's surface stemming from the dissociation of the adsorbent, which tend to mix with the water. Furthermore, a significant number of membranes employed, particularly in fluidised bed reactor, are pristine, thus not all contaminants are effectively captured within this system. This limitation has been a substantial hindering factor to its widespread application especially in mine wastewater treatment which has elevated concentrations of both organic and inorganic species.

To facilitate a widespread adsorption of the fluidized membrane bed reactor, it is imperative that ideas and concepts from the advancement of nanotechnology are incorporated to ensure effective and high water quality. Modification of the membrane with adsorbents present as a feasible way to not only enhance the hydrophilicity of the membrane but also turn the membrane into an adsorptive membrane. Adsorptive membrane offers more advantages such as increased adsorption.

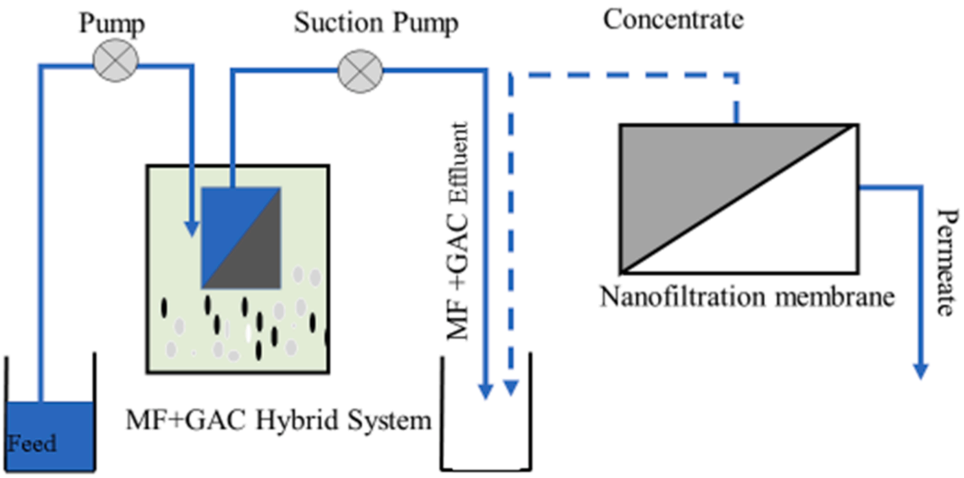

Fig. 7. Hybrid membrane adsorption process with Microfiltration coupled with GAC and NF. Redrawn and adopted from (Loganathan et al., 2023).

Challenges and conclusion

Mine operation produce large amounts of wastewater in form of tailings, AMD, and process water. The adverse negative impacts of such wastewater on the environment can be overemphasised. Propelled by global water scarcity and high demand for finished metal products, it is important to treat this wastewater to recycle and remove toxic metals. Membrane technology plays a pivotal role in this goal. The reviewed literature has highlighted the superior benefits of membranes in mine wastewater treatment including achieving higher removal of metals from the effluents and high water. However, the challenge with these membrane system is extensive energy consumption which increases the operational cost for the membrane which may not be beneficial for the mining industry. Hence, slow adoption in the mining sector.

Studies have shown NF membrane as efficient membranes in mine wastewater treatment because of the efficient energy consumption and high metal removal up to greater than 95 % and water recovery greater than 90 %. However, this technology comes with its own challenges including fouling and energy consumption.

Exhaustive research has been conducted to improve membrane performance and offset the challenges of membrane fouling such as membrane functionalization by using of nanomaterials such as nanoparticles of metal oxides, metal nanoparticles, hybrid membrane systems and pre -treatment methods. All these research has aimed at water recovery and resource recovery.

In a circular economy era, mine wastewater is considered to be a secondary source of resources which can be reprocessed for value addition. The role of membrane processes such as RO, NF, MD and ED is to recover water and to concentrate solute brines for extraction of resources. membrane systems coupled with other chemical methods such as adsorption, ion exchange and solvent extraction can significantly contribute to the sustainable mine wastewater management and improve the quality of the water discharged and recovery of rare earth elements and toxic metals. Literature studies show that high purity of REEs can be recovered from mine effluents such as AMD.

However, further research must focus on studies using real mine effluents to assess the long term effects of mine effluents on functionalized membranes. Literature studies show that the majority of the studies conducted on mine effluents were done with synthetic mine effluents. Furthermore, future research should focus on studies to achieve a zero liquid discharge to avoid the negative impacts of heavy metals on the environment. Membrane technology looks very promising in mine effluents treatment and can have a wide application especially in concentrating brine for recovery. However, a low cost efficient, durable and highly selective membrane should be developed to allow for the longevity and application in the mining industry.

CRediT authorship contribution statement

Eunice Zulu: Writing – original draft, Writing – review & editing, Formal analysis, Software, Data curation. **Subbaiya Ramasamy:** Writing – original draft, Writing – review & editing, Software, Supervision. **Keneiloe Khoabane Sikhwivhilu:** Writing – original draft, Writing – review & editing, Resources, Methodology, Visualization, Supervision. **Stephen Syampungani:** Writing – original draft, Writing – review & editing, Resources, Formal analysis, Supervision, Visualization.

Declaration of interests

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.

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