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REVIEW PAPER

The role of membrane technology in acid mine water treatment: a review

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Abstract−The activities of mining industries are attracting more scrutiny as the concern of limitations of conventional technology for wastewater treatment and the potential use of wastewater have resulted in accelerated attention in membrane technologies. The paucity of water and industrial environmental guidelines has resulted in the application of membrane technologies in wastewater treatment, especially in the mining industry. Although many conventional physical and chemical processes have been employed to treat acid mine drainage (AMD), they have, however, demonstrated low efficiency and high cost. Membrane technologies have proven to be an important part in the treatment of AMD in order to reduce water paucity. Apart from addressing water paucity, membrane technologies meet high-level application with respect to ease of use, adaptability and environmental impacts. This paper reviews the use of membrane in the published literature for the treatment of acid mine waters and, for the recovery of valuable metals from acid mine drainage effluents. The role of membrane technology in acid mine water treatment is discussed together with the factors that determine membrane performance for AMD treatment. The challenges of membrane technology in acid mine water treatment were reviewed and some solutions to the challenges are presented.

Keywords: Acid Mine Drainage, Active Treatment, Passive Treatment, Membrane Technologies, Fouling, Pre-treatment, Nanofillers, Brine Management

INTRODUCTION

Mitigation and treatment of mine water in mining operations are the most vital difficulties confronting the mine water industry. A typical by-product of the mining industry is acid mine drainage (AMD); this is a type of mine impacted water (MIW). MIW is detrimental to the surroundings and causes paucity of water [1]. AMD is considered a paramount problem to the surroundings and it takes place in running and deserted mines [2,3]. During the operational stage, AMD forms from the processes of enhancement, dewatering water, and drainage from the depository of the material that overlies an ore [4]. AMD can continue for years if the oxidation of sulfides in the mine is not controlled. Hence, the level of acidity, the composition, and the percentage amount of metal in a specified outflow of acid water is governed by the quantity, kind of sulfides and related neutralizing chemical compounds, like calcite and dolomite. Furthermore, the access options of air and oxygen present in the ore before the discharged into the public stream also influence the level of acidity, the composition, and the percentage amount of metal in a specified outflow of acid water [5]. Acidic water containing metals from the waste storage areas can leak under a dam or through the base of the area into surrounding groundwater or perhaps, straight through the dam into the perimeter drainage ditch and the surface waters. The oxidation products of sulfides are formed on the walls of the mine as the mine fills up with water. The mines are washed out in the backfill which results in the spread of metal-bearing water along fractures in the bedrock into the groundwater and possibly through overflow into surface waters [4].

The characteristics of AMD are high acidity of pH between 2-4, high sulfate concentration between 0.001-0.02 kg/L, an excessive amount of metal ions and some undesirable chemical substances like aluminum, calcium, cobalt, iron, magnesium, manganese, and nickel [6,7]. These characteristics result in a severe contamination of soil, surface, and groundwater [3]. The stages of chemical equations that occur during AMD production from pyrite are given in the literature [3,8]. The accumulation of $Fe(OH)_3$ results in the manifestation of by-product from mining companies, hence, an important contributor to the contamination of water [3]. The advancement of several investigations on the processes of oxidation and the interception of mine water resulted in the need for preventing AMD formation [9].

Numerous wastewater treatment technologies such as adsorption process [10-12], ion exchange [13,14], chemical precipitation process [15,16], process of electrochemical [17,18], phytoremediation process [19] and membrane filtration process [20] have been extensively used to treat AMD. As a result, the benefits of membrane filtration technique, which include operation at room temperature, low energy consumption, simple operation processes, high efficiency, and small investment, has invited much consideration than other listed treatment techniques used for the purifying AMD. Membranes with a pore size of about 0.1-5 μm are microporous and can only be used for treating wastewater with particle size range of 1-10 μm. This restricts their utilization in water treatment [21]. However, nanoporous membranes have high performance capacity of purifying wastewater. Hence, the technology of membranes can be used to treat AMD due to their capability of removing suspended

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solids, dissolved organic compounds, metallic ions, microbes and multivalent ions from wastewater. In this review, two main conventional methods of treating AMD are discussed together with the details of the advantages and disadvantages of the two conventional methods. Attention is drawn to the role of membrane technology in acid mine water treatment. The advancement of membrane technology in acid mine water treatment is discussed. Finally, challenges and solutions to the challenges of membrane technology in acid mine water treatment are highlighted.

ACID MINE DRAINAGE TREATMENT

Surface mine control and metal recovery requires that mine water from all active and inactive mines be treated in order to comply with effluent criteria before discharging to the environment. Before the interest in membrane technology, different kinds of methods of treatment were used for treating mine water. AMD purification can be achieved by either passive or active treatment systems [22].

1. AMD Treatment Using Passive Treatment Set-up

Passive treatment is a technique of removing metals from acid mine drainage. Quite a number of types of passive treatment systems exist. They can be utilized on their own or combined to purify AMD effluents. The choice of passive treatment type is reliant on AMD chemistry and the flow of the discharge. Power is not required for these systems. When compared to active treatment systems, they are less expensive and less maintenance is required with the utilization of passive treatment for AMD [23]. There are numerous types of treatment systems used for eliminating metals from AMD. Some of the more commonly used ones are aerobic wetlands, anaerobic wetlands, successive alkalinity producing systems, anoxic limestone drains, limestone pond, open limestone channel [23]. All these function by making contaminated mine water retentive for a long time for the purpose of reducing contaminant concentrations to acceptable levels [24]. Hence, passive treatment systems for AMD are envisioned to improve the quality of waters that pass through them. These systems are modelled after wetlands and other natural processes, with adjustments focused on meeting precise treatment goals [25]. The characterization the waters to be treated is an important step required in designing passive treatment system. This can be achieved by evaluating the discharge, the flow of those waters, and the water-quality constituent's concentrations of concern, for a long enough period in order to determine how these quantities vary seasonally. This information allows the determination of elemental concentrations and flow volume to be treated. These design conditions, and site characteristics influence passive treatment system selection [25]. Furthermore, the chemical and biological processes that eliminates contaminants differ between metals. These processes are affected by the mine water pH and oxidation reduction potential [24].

AMD treatment using passive treatment set-up rely on biological, geo-chemical, physical, and hydro-chemical processes based on matter-of-course; however, if the set-up is not discreetly chosen and developed, the set-up can be deficient [26]. In many passive treatment set-ups, limestone is usually used as a dissolution of a neutralizing material for the neutralization of the acidity in AMD. For the dissolution to happen, adequate residence time is required in the set-up; hence, the residence time is dominant as the limestone dissolution is a function of the residence time in a limestone bed for passive neutralization of AMD. Again, reducing and alkalinity producing systems have an extra deposit of compost or other organic material on the limestone layer. The compost deposit yields anoxic conditions, which reduce Fe(III) to Fe(II) or limit oxidation of Fe(II) and inhibit passivation of the calcite layer. Hence, huge areas of land are usually needed for this kind of treatment; they are appropriate for both running operations and abandoned mine sites and should be considered during the earliest stages of mine development [22]. The reaction that takes place between water and alkalinity-generating materials like limestone and alkaline steel slag is a determinant factor for geo-chemical passive treatment. Skousen et al. [5] described a world-wide utilization of purifying contaminated water from mining operations using a passive set-up. Passive treatment systems are characterized by retaining AMD long enough to reduce contaminant concentrations to tolerable levels. The generation of the factors that promote the methods that rapidly eliminate aimed contaminants is done by an effective passive treatment set-up. Hence, the development of a passive treatment set-up should be based upon the firm putting the chemistry of AMD into consideration and how several passive scientific know-how influences the chemistry [27].

1-1. Cons and the Limitations of Passive Treatment

Passive systems usually contain several treatment units such as organic substrates and limestone. Organic matter employed for passive treatment systems is commonly a locally available biological waste [28]. Different organic substrates have been used for passive treatment systems; however, their effectiveness is site dependent and water quality [29]. The life expectancy of this kind of treatment is reliant on the quantity of organic matter and limestone in the set-up. The availability of the porosity within the organic matter and limestone can influence any anticipated existence of the treatment because the pores govern the magnitude of storing treatment precipitates. Thus, this kind of operational set-up could be unsuccessful because of the organic matter and limestone pores blockage with treatment precipitates in the set-up [30]. Furthermore, should mine drainage surpass precise thresholds, huge quantities of neutralizing material are needed to make sure suitable treatment is achieved. In such a situation, a huge passive system would be needed, which can be susceptible to failure, and active treatment is likely to be a better choice [22]. Again, in order to treat AMD with high flow rates and a low pH and/or high acidity AMD with passive treatment systems, a huge system is required to attain a longer residence time for neutralization and a very huge quantity of neutralizing material is essential to uphold system durability. However, very huge systems could be susceptible to short-circuiting and failure, because the advantageous pathways may be developed over time. Instead of building one large system, splitting the flow among several parallel systems would be preferred [22]. Flow rates must also be taken into account along with the practicalities of treatment. Nonetheless, passive systems are usually more cost-effective when compared to active systems, specifically for abandoned mines [26].

2. AMD Treatment Using Active Treatment Set-up

Active treatment involves a continual cost for continuous dosing with chemicals, mining engagement and sustentation, and the usual delivery of electricity. Furthermore, this treatment involves the hazard of accidental release of stored agents. NH_3 and NaOH are examples of stored agents that can lead to detrimental surroundings or public hazard. The hazard of using NH_3 and NaOH for AMD treatment is the danger in handling the chemicals, the incertitude relating to nitrification, denitrification, and acidification downstream, and the outcome of extreme application rates [31]. However, passive treatment is excluded from these obstacles [5]. Active treatments usually include traditional treatment processes such as neutralization, precipitation, aeration, adsorption, and ion exchange. Generally, active treatment systems need equipment such as tanks, mixers, and pumps; however, they are very much more dependable than a passive set-up [32]. The main advantages of active treatment systems are: (1) their potency in eliminating pollutants such as acidity and metals from acid mine water; (2) Their provision of exact process regulator such as flow rate and acidity load in a way that they can be controlled; (3) Their ability to be manoeuvred to yield an explicit water chemistry, and (4) appropriateness of active treatment in mine sites where a limited land space is obtainable for remediation systems [22]. It is expected that the application of calcium-based chemicals such as limestone for active treatment should continue to be the main option for neutralizing AMD because of the non-retention scenery of the chemicals, their broad accessibility, the simplicity of implementation and profitable factor [30]. Nonetheless, active treatment systems have some disadvantages. The key shortcomings of this treatment method are the continuous running, elevated maintenance costs and elevated capital cost.

THE ROLE OF MEMBRANE TECHNOLOGY IN ACID MINE WATER TREATMENT

The increasing demand for clean and portable water in countries that have mining industries is as a result of the hazard that AMD poses to indigenous natural water resources. Furthermore, AMD affects the wildlife around the affected body of water. Many aquatic animals such as species of fish cannot also survive water affected by AMD [33]. The inflow of AMD into surface waters such as streams, rivers and lakes often leads to the degradation of aquatic ecosystems because of the compounded effects of elevated hydrogen-ion concentration, metal ions, siltation, and the iron precipitate known as yellow-boy [34]. In addition, mine drainage is usually sporadic and diffuses by flowing into affected surface waters at several points through surface runoff and ground-water inflow. Therefore, the site of point sources used in comparing unpolluted and polluted areas within the same is one of the difficulties involved with field investigations concerning the effects of mine drainage on aquatic ecosystems [35]. This has made the strategy of treating water before discharging to the rivers and reusing water a recent topic of great significance. Membrane separation processes utilized for water and wastewater purification afford the use/reuse of water resources [36]. Hence, the successful used of membrane separation processes in many industries is because of their selectivity, ease of application and adaptability characteristics [37].

AMD treatment done by employing membrane technology is very rare owing to the moderately high cost of the membrane and high membrane fouling because of the proneness of membrane systems to low pH and high salt concentration of AMD. Nonetheless, in recent times, application of nanofiltration (NF) and reverse osmosis (RO) processes for AMD treatment has gained interest owing to their high capacity of salt and metal retention [38]. Membrane processes have been introduced in mining industries for the purpose of treating mine water, reusing process water and for the potential reuse and recovery of by-products. However, there is need for the pre-treatment of mine water to remove suspended solids in order to reduce fouling of membranes in membrane systems. During the membrane process, AMD first goes through neutralization in order to adjust the pH. Furthermore, to avoid scaling of the membrane because of the presence of calcium and magnesium in mine waters, softening is added prior to microfiltration and/or ultrafiltration process steps [39]. The pre-treatment choices for nanofiltration and reverse osmosis are microfiltration and/or ultrafiltration processes. Membrane technology processes hence play a crucial role in the sustainability of water and are relatively new state of the art method used in treating AMD. An ideal membrane process would recover all the water, with the renunciation of only the salt. However, membrane processes have different features and specific application [36]. Diverse membrane separation processes are employed in the purification of water, sewer and industrial wastewater. Among the membrane processes, pressure-driven membranes (microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO)) and electrodialysis reversal (EDR) are the ones used for the treatment of wastewater [36]. The membrane process is effectively a size exclusion process where pressure is exerted on one side of a semi-permeable membrane, except for dense membranes such as the RO, or gas separation membranes. Hence, theses membrane act as a semi-permeable barrier and they could have different selectivities for different ions or compounds. From these membrane processes, EDR, NF and RO are the commonly used membrane separation processes due to their unique characteristics.

In the past 20 decades, electrodialysis reversal (EDR) has been used in desalinating non-scaling mine water due to their technical viability [40]. EDR is an advanced membrane separation process that uses the movement of ions to desalinate water with the capability of achieving high water recovery. An EDR was used to obtained high-recovery inland brackish water, which was employed to find the maximum permissible $CaSO_4$ saturation index during a continuous long-term operating system [41]. EDR is the desalination membrane process used for separating dissolved ions across ion permeable membranes based on the influence of an electric made of alternating films of cationic and anionic ion exchange membranes [42]. Fig. 1 shows an electrodialysis reversal cell. The cell is made of AMD feed section and a concentrate section created by an anion exchange membrane and a cation exchange membrane place in the middle of two electrodes. The membranes permit the permeation of oppositely signed ions and separate the ionic species from an aqueous solution and from other uncharged species with the aid of an electrical force [43].

The salts and small organic molecules are separated from liquid streams through reverse osmosis. Reverse osmosis process is the application of membranes with active dense films, in which the preferable mechanisms of transport are usually ascribed to solu-

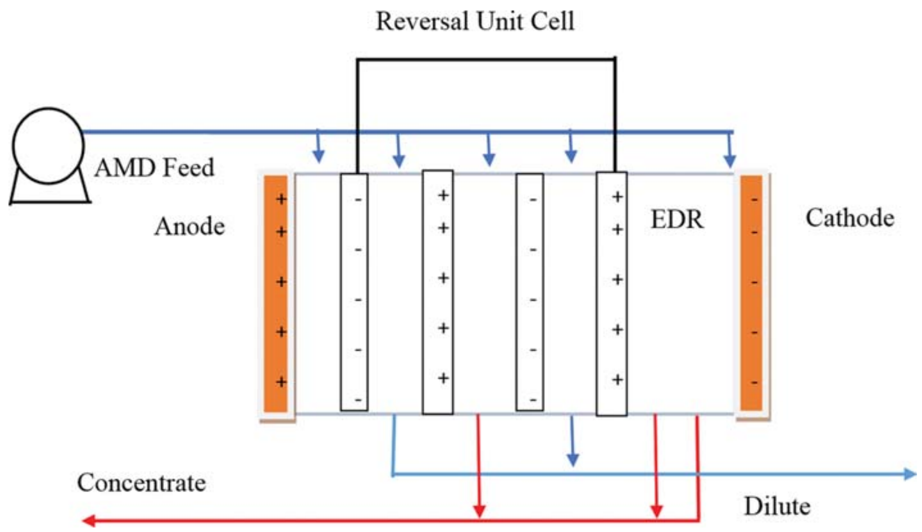

Fig. 1. Schematic illustration of electrodialysis reversal cell.

tion/diffusion. Because of the high density of the active film, operating pressures are required to be more than the operating pressures employed in microfiltration and ultrafiltration [36]. Nanofiltration process shows performance characteristics, which fall in the middle of ultrafiltration and reverse osmosis membranes. NF is generally utilized for the separation of organic solutes with low molecular weight (200-1,000 Da) and in the partial demineralization (principally polyvalent salts) of liquid streams [36]. Transport of species in the course of nanofiltration process is subject to three different mechanisms shown in Fig. 2. The mechanisms are: (1) convection, which takes place because of the exerted pressure difference across the membrane. (2) diffusion, which takes place as a result of the concentration gradient across the membrane, and (3) charge effects, which are due to potential gradient or which take place as a result of electrostatic repulsion amid the charged membrane and a charged organic compound [44,45]. Hence, the rejection of organic compounds occurs as a result of diverse membrane features such as molecular weight cut-off, membrane charge and compound properties such as molecular weight, hydrophobicity and ionization constant [46].

Reverse osmosis (RO) and nanofiltration (NF) are acceptable techniques for metal ion retention from the aqueous medium, offering great capability in treating mine wastewater for water reclamation [48]. Some researchers have successfully used NF and RO membrane technologies to eliminate contaminants in mine water [49-52]. The difference between RO and NF is based on the size of particulate that each is able to remove. Hence, the main difference lies in the construction of the membrane material itself, where

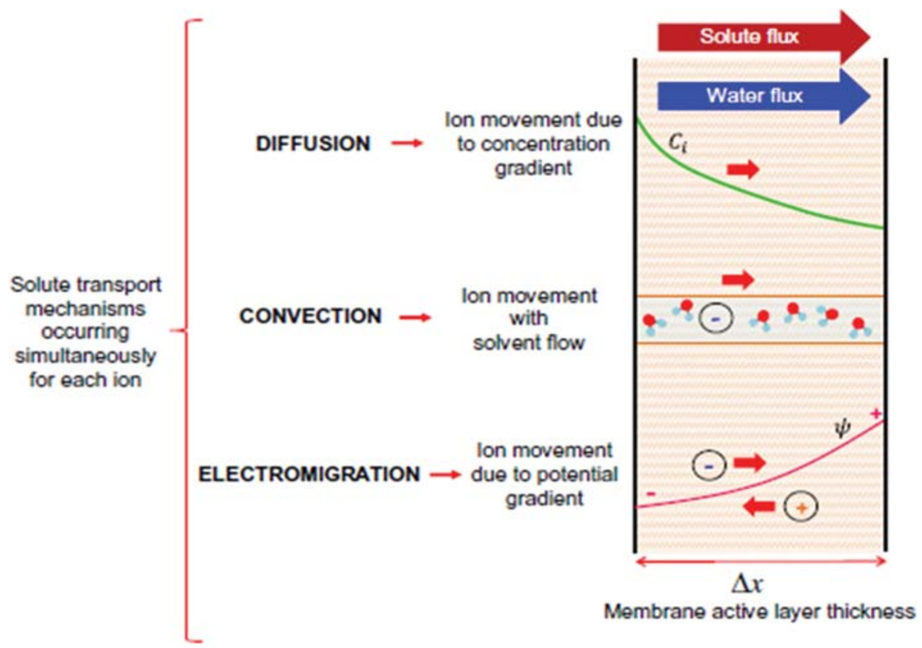

Fig. 2. Nanofiltration membrane transport mechanisms of solutes across the [Adapted from Ref. 47].

the pore structure of a NF membrane is "looser" than that of an RO membrane owing to less cross-linking of the membrane. Besides, NF is recommended to be the superior membrane used in eliminating of metal ions in effluent because of its unique properties: lower operating pressure, substantial permeate flux, lower utilization of electricity, lower principal investment and lower cost of running [53-58]. Solute separation by NF membranes occurs via several mechanisms such as diffusion, convection, steric hindrance, Donnan and dielectric effects [59]. Hence, NF membrane surface charge and pore size have an impact on the retention of ions and molecules. Because obtainable NF membranes in the market are typically hydrophilic and susceptible to be hydrated and ionized in the effluent, the configuration and ionization of polymer chains in the membrane will be altered with the influence of different surrounding conditions, particularly with distinct pH and ionic strength. As a result of the pore at nanoscale sizes which is approximately 1 nm and NF membrane materials that are electrically charged, a slight alteration in pore size or charge pattern would even have a vibrant effect on the penetrability of membrane and molecules [60].

Aguiar et al. [60] examined the impact of effluent pH in the nanofiltration of gold acid mine water. The membranes investigated were commercially available, NF270, NF90, and MPS-34, and they were selected for their supposed elevated capability for the purification of acid mine water based on high rejection of metals. The outcome of their investigation revealed that the MPS-34 and NF90 membranes operating at the normal acid mine water pH of 3.2 attained higher permeate fluxes than in other experimental settings. However, when looking into conductivity retention, the performance of MPS-34 membrane based on the rejection of salts was as low as about 44% for all experimental settings. The NF270 membrane offered elevated permeate fluxes and elevated rejection of sulfate than NF90 membrane. For NF270 at pH 5.5, the fouling resistance was marginally lesser than NF270 at pH 4.2, demonstrating that these conditions could be the most suitable experimental settings for the gold AMD purification [60]. Variations in NF membrane properties like the effective intra-pore solute diffusivity, effective thickness, and the effective membrane pore radius could probably be the reason for lower fouling observed at higher pH. Another reason could be due to the alteration of the NF membranes active layer charge with the operating temperature. Mullett et al. [51] investigated the performance of DOW NF 270 polyamide thin film and TriSep TS 80 polyamide thin film nanofiltration membranes for their rejections of ionic species when treating mine affected water streams at the different variety of acidic pH values. The efficiency of the iso-electric point of the membranes was tested by the alterations in rejection over a small range of pH by treating solutions of sodium sulphate. The two membranes presented alterations in rejections at pH 3, signifying a zero-net charge at pH 3. After the retrievals of different variety of cations, the appropriateness of nanofiltration was relatively based on the pliability with discharge standards of mine water and the retrieval of treasured metal products. The confirmation of NF technology has benefits in retrieving metals from mine water, concurrently allowing discharge standards for the separation disposal to be fulfilled.

AMD, especially abandoned mines are mainly acidic with elevated concentration of metals and sulfates [61]. NF membranes have been evaluated for the purification of abandoned mine drainage. The purification of abandoned mine drainage with NF was investigated on a laboratory and pilot-scale for the retrieval of valuable products appropriate for industrial reuse [62]. Eight NF membranes obtainable in the market were evaluated using synthetic and real AMD on a laboratory scale with the aid of dead-end and cross-flow cells. The systems were operated for 208 h with the real AMD at 10 bar, and more than 99% of total dissolved solids (TDS) was rejected and 57% water recovery was achieved [62]. Sierra et al. [63] examined the efficiency of NF technology for the treatment of AMD from an abandoned mercury mine. The sediment geochemistry and the origin of acid waters were analyzed in order to understand the geochemical factors involved in nanofiltration. The researchers found that NF has the capability of rejecting up to 99% of aluminum, arsenic and iron content, and 97% of sulfate content [63]. Hence, having a good comprehension of the retention behavior of a specific membrane for AMD challenge is important in evaluating the strategy of an NF design [55].

RO systems, on the other hand, utilize a membrane that is significantly less permeable to dissolved salts but permeable to water, hence, suitable for the removal of dissolved metals, ions, and organic molecules of lesser molar mass [48]. Zhong et al. [64] used commercially available polyamide ultra-low-pressure RO and NF membranes to reclaim acid mine drainage and separate metal ions. The performance evaluation of the membranes was based on the retention of ions and total conductivity, which were respectively more than 97 and 96% for the ultra-low-pressure RO tested. This suggested that the membranes are suitable for AMD treatment and wastewater reclamation. Nonetheless, it was observed that NF process had the capability of removing close to 90% of the ions in the AMD with 48% reduction of the total conductivity. Andrade et al. [48] used RO and NF90 membranes to assess the best conditions for gold mining effluent treatment based on feed pH and permeate recovery rate with the aid of crossflow cell. In comparison, the treatment of mine effluent with NF90 membranes was much more effective than RO membranes. The reason is that NF90 membranes enable permeate fluxes of 7 to 12 times more than the RO membranes and exhibit a good compatible rejection effectiveness. Hence, NF90 performed much better than RO. An incorporation of oxygen furnace slag, lime, soda ash and RO was employed to purify drinking water and recover treasured minerals from AMD [64]. One pass of RO system was replicated in RO system analysis. The water obtained from this system met the standards of the South African National Standard 241 Drinking Water Specifications. The authors thus deduced that the new scientific know-how has proven that drinking water and treasured minerals can be retrieved from AMD using RO membranes [65].

It is obvious that the characteristics of AMD feed and the type of membranes are responsible for the performance of NF and RO. The review shows that RO performed better than NF in a particular mine [64,65], while NF performed better than RO in another type of mine [48]. Hence, a fundamental knowledge of the components and the functionality of materials used for synthesizing membranes are required for the design and implementation of RO and NF membranes. Furthermore, the knowledge regarding the characteristics of a type of mine is very vital as different mines have different metal ions and different concentration of ions, which will, of course, have an impact on the performance of the chosen membrane (RO or NF). For example, Nordstrom et al. [66] presented a report of AMD from an iron mine in California called the Richmond Mine at Iron Mountain. The AMD that comes from the iron mine was represented by a very high sulfate content of 760 g/L, 200 g/L of total dissolved metals and a negative pH value of −3.6. The characteristics of this mine water could have a severe impact on any chosen membrane; thus, a knowledge of the membrane and AMD characteristics is important. In addition, the specific requirements of the rejection and permeate will also govern the appropriate membrane selection between RO and NF. For instance, if the permeate produced is required for discharge and there exist very tight limits on the permeate quality, then an RO would be a better fit. If the product water will be re-used as process water within the mine, the requirements may not be as strict and an NF would be more appropriate. Similarly, if the acid is to be recovered, then an NF would be more appropriate than an RO. A few of the significant characteristics to be considered when selecting a suitable membrane for the treatment of AMD are AMD feed analysis, the polymer composition of membrane, pore size, operating conditions (pressure, flow rate of feed), hydrophilicity and hydrophobicity. The following sub-section discusses some of the factors that determine the performance of membranes for the treatment of AMD.

1. Factors that Determine Membrane Performance for AMD Treatment

For the treatment of AMD, some significant factors such as membrane characteristics, feed characteristics and operating condition determine the membrane performance.

1-1. Membrane Characteristics

The structure and the separation principles of membranes have the capacity to determine their performance. The membrane characteristics, especially the pore size, charge and hydrophilicity are strictly integrated to the performance of the membrane [67]. Membrane charge and pores are the main transport parameters of membranes. Membranes contain pores in a different range of sizes, depending on the principle of separation used; hence, filtration is based on particle size. In the interest of attaining high selectivity, membrane pores must be relatively smaller than the particles in the AMD feed. Thus, membrane pore size is a crucial factor influencing the level of membrane rejection on uncharged contaminants found in AMD. Membrane separation process must also put into consideration the charges of the membrane and solute. When an aqueous solution moves onto or close to the membranes, an electric charge is obtained by some mechanisms like adsorption of ions from the solutions and adsorption of polyelectrolytes, dissociation of surface functional groups, ionic surfactants and macromolecules [68]. To conserve the electro-neutrality of the system, the mechanism of the charge could occur on the exterior surface of the membrane and in the membrane pore interior because of the ion distribution in solution [69]. If there is a difference between the membrane charge and the solute charge, the process could result in a gravitational force, hence, the chance of fouling [67]. Furthermore, hydrophilicity is a primary membrane property connected to a weak solute interaction and connected to a high water interaction. Generally, it is known that hydrophilic membrane is accorded to a membrane with lower fouling ability than hydrophobic membrane [70,71]. Hence, enhancing hydrophilicity during the design of membrane should be considered a significant strategy in alleviating fouling in a membrane during AMD treatment. In addition, the optimization of the synthesis procedure for the production of membranes and process conditions could be needed in order to improve the performance of the membranes for AMD purification [72].

1-2. Feed Characteristics

Feed characteristics such as pH, salt concentration, the hydrophilicity of solute, the structure of chemical and fouling potential are also significant factors that affect membrane performance. AMD pH has the capacity to influence membrane charge, even alter it [67]. Hence, the AMD solution pH has a very strong influence on the effectiveness of membranes and the separation of cations and anions. The pH of the feed governs the charge density and charge polarity of the membrane by instituting the zeta-potential at the surface of the membrane [55]. The membrane iso-electric point (IEP) is an important parameter that occurs when the pH value of molecules/colloids carries no electrical charge; hence, at pH where there is no charge, the net charge of the membrane is zero. When the pH values are below the IEP, the membrane is positively charged, and negatively charged when the pH values are above the IEP; hence, zeta potential turn out to be more negatively charge as pH upsurges and more positively charge as pH decreases. When working with pH value below the IEP, the very high rejection of metal is established; thus, the membrane is positively charged. The pattern is reversed for anions, such as sulfur; minimum rejection is experienced when feed pH is low, signifying that the IEP is in the area of low pH values [51,55]. Furthermore, the pH and the hardness of cations have a very strong impact on the charge of the membrane and on the properties of the molecules in the solution. Thus, the pH has the capacity to protonate and deprotonate membrane functional groups and the molecules in solution over its range [73]. The potential of a pH will alter the charge and pore sizes of the membrane, thus reacting to the membrane performance [69, 74]. Hence, having a good knowledge of the connection between AMD pH and membrane charge is essential in order to ascertain the compliance with strict discharge criteria, and to take full advantage of the metal recovery for proceeds [55]. Furthermore, the diffusion gradient is responsible for the solubility limit of the solute within the polymer matrix of membranes. The concentration of the individual feed components at the surface of the membrane is in solubility equilibrium with the feed composition. Hence, sustaining the solubility gradient across the membrane for the components of the feed prompts the flow of the fluids across the membrane [75].

The fouling potential of the feed is an incredibly imperative characteristic of the membrane. Membrane fouling can be defined as undesirable formation and the deposition of unwanted impurities such as colloidal matters, organic molecules, sparingly soluble inorganic salts and microorganisms, found in the feed on the membrane surface or into the membrane pores, which eventually block the membrane pores [76,77]. Hence, fouling results in rapid decline of the permeation flux with time and causes poor membrane performance [78]. Colloidal fouling, organic fouling, inorganic fouling and biological fouling are the common types of membrane fouling. Colloidal fouling arises owing to the deposition of colloids and particulates on membrane surface or pores. The main sources of biological fouling are microorganisms and bacteria in the water. The adsorption of organic species instigates the organic fouling of membrane. While, inorganic fouling arises because of scale formation of sparingly soluble inorganic salts [77]. Apart from the types of fouling mentioned in this sub-section, fouling can be classified into reversible and irreversible. This classification depends on the intensity of the foulants' attachment to the membrane surface [79]. Reversible fouling can be removed easily with certain cleaning methods, such as water flushing or backwash because it is a temporary membrane fouling [80]. Irreversible fouling is a permanent fouling. The foulants of an irreversible fouling remain on the membrane after cleaning of the membrane, and replacing the membrane is the only solution [81]. The degree of fouling usually depends on the following factors: operating parameters, such as pressure and stirring speed, feed characteristics, such as solute size and charge, salt concentration, and membrane characteristics, such as porosity, pore size and surface roughness [79].

During processing/mining of ores, the effluents produced usually have high salt concentrations because the salts are typically connected with valuable components in ores and concentrates [82]. Membrane fouling is a multifaceted phenomenon in higher salt concentration environment, instigated by inorganic fouling (in which the key phenomenon is salt scaling), organic fouling (in which the key phenomenon is colloidal fouling), biofouling, and their activities [83]. The higher salt environment has the capacity of causing scaling on membrane [84]; furthermore, it has the capacity to aggravate colloidal fouling [85]. In addition, metal ions/salts built-up on the surface of the membrane as a result filtration give rise to concentration polarization. In the course of time, the membrane active pores get blocked and thus decrease the membrane flux [86].

The detrimental effects of membrane fouling are an increase in transmembrane pressure and the reduction in the effectiveness of separation performance, which are low permeate flux, low water recovery and low rejection rate. These detrimental effects could ultimately cripple the economic feasibility of membranes in separation processes [87]. Several measures have been taken to lessen membrane fouling for the purpose of increasing membrane flux, water recovery, rejection rate, and extend membrane life span. Membrane modification [88], chemical cleaning [89,90], and hydrodynamic adjustments [91] have been employed to alleviate membrane fouling. Hence, the transformation of membrane material or membrane surface is the usually employed to enhance membrane performance and to meet diverse requirements [92].

1-3. Operating Conditions

Operating conditions like pressure, flow rate, and temperature intensely affect the performance of the membrane; hence, membrane performance for AMD treatment has been evaluated under different operating conditions [93].

1-3-1. Operating Pressure

The operating pressure of membrane separation processes has an influence on the metal separation efficiency [63]. Increased pressure is directly proportional to flux increase: the higher the pressure applied, the higher the permeation fluxes for a fixed feed salinity, linearly. This is due to the consequence of the impact of driving force taking place during the membrane operation. The elevated pressure makes the thrust that transport of the feed solution through the membrane gets higher in order for the chemical species embodied in the feed solution to rapidly move into the pores of the membrane and out as infiltrates [94]. However, at a certain point, the linear relationship between operating pressure and fluxes is not applicable owing to fouling and concentration polarization [95]. Furthermore, the operating pressure does not only affect the flux, but also on the salt rejection.

1-3-2. Flow Rate

Increased feed flow rate influences the performance of the membrane by increasing the permeate flux and mass transport [96]; even at constant pressure, the recovery is not fixed in this context. This could be linked to the possible decrease in the effect of the concentration polarization. The concentration polarization is directly connected to the thickness of the boundary layer, which is of high significance for effective separation and could lead to a decrease in the process performance of membrane during AMD treatment. Increase in flow rates remarkably decreases the boundary layer, hence resulting in the decrease of mass transfer resistance of the boundary layer on the upstream of the membrane, which in turn leads to increased permeation flux [97].

1-3-3. Temperature

The productivity of NF and RO is extremely susceptible to the alterations in AMD feedwater temperature. Increase in water temperature will result in a linear increase in water flux, primarily owing to elevated diffusion rate of water across the membrane. This relationship is also due to viscosity and reduction of the level of concentration polarization at the membrane surface [98]. Nonetheless, the occurrence of fouling results in flux decline despite temperature increase [99]. Furthermore, increased feedwater temperature leads to lower salt rejection. The effect of feedwater temperature on flux and salt rejection is because of higher diffusion rate for salt across the membrane and the variation of the diffusion coefficient and component absorption for the flux coming from the membrane [67]. Hence, the capability of a membrane to endure high temperatures upsurges operating latitude, and is imperative through cleaning operations for the reason that it allows the use of stronger, faster cleaning processes [100].

CHALLENGES AND SOLUTION OF THE MEMBRANE TECHNOLOGY IN ACID MINE WATER TREATMENT

There are certain limitations to using membranes for the treatment of mine wastewater. Abrasion and breakage of the membrane material can occur from solid particles. Solid particles could obstruct flow channels and feed spacers of spiral wound membrane elements [101]. Complex physical and chemical interactions between the several fouling elements in the feed and between these elements and the membrane surface are other limitations that instigate fouling [102]. Usually, membranes encounter fouling after an extended period of filtration time. The quality of the product could be affected and further cause the replacement of the membrane [103]. Membrane fouling takes place in the course of an upsurge in transmembrane pressure to sustain a precise flux or during the reduction in flux when the system is functioning at a constant pressure. Membrane fouling is categorized as reversible fouling and irreversible fouling. The difference between these two types of fouling is completely reliant on the perspective whereby membranes are functioning and cleaned. Reversible fouling, either back-washable or non-back-washable, occurs because of the cake layer or concentration polarization of materials at the membrane rejection surface. The restoration of membrane with back-washable reversible fouling via suitable physical washing procedure, such as backwashing or hydrodynamic scouring, is possible. This process is known as surface washing. The non-back-washable reversible fouling can be only taken away through chemical cleaning. Irreversible fouling takes place through chemisorption and pore plugging mechanisms [102].

Sulfate forms salts with calcium, magnesium, and sodium, and each of these salts can easily foul membranes and equipment, as they could be present in AMD at a concentration ranging from few hundred to several thousand milligrams per litre [104]. This could be due to the low rejection of magnesium and sodium, which is owing to the weakening of the membrane surfaces. This occurs because of high formation of scale as a function of the inorganic fouling, which causes a reverse effect onto the membrane performance. Furthermore, an unforeseen upsurge of magnesium and sodium salts rejection than divalent salts could be because of increasing of the applied pressure, which is equivalent to triple the salt osmotic pressure [105]. In addition, the solubility of magnesium and sodium depends on temperature; their solubility increases with decreasing temperatures [106]. Sulfates are known to be drastic membrane foulants, and the control of these foulants is vital for a successful mine water treatment. Hence, fouling in mine wastewaters is usually because of inorganic foulants such as calcium sulfate [107]. The fouling layer elimination and hence the cleaning of the fouled equipment is very demanding. Thus, pre-treatment is usually required for excessive sulfate concentrations [3]. Furthermore, solid particles can be eliminated by either screening or particulate filtration. Ultrafiltration as a pre-treatment or pre-filtration is very vital in making sure optimum system performance is achieved, particularly relating to the rapid growth of industry adoption of ultrafiltration for RO pretreatment. Arevalo et al. [108] operated a submerged membrane ultrafiltration plant at pilot scale, as pre-filtration of complex brackish surface water to examine the effectiveness of filtration. The outcome of their investigation shows the effectiveness of the membrane, chemical reagent needed, water quality, and the effectiveness of the cleaning protocol of an ultrafiltration pilot plant employed as pre-filtration for a reverse osmosis system. A reasonable substituted chemical cleaning protocol was used, which maximized permeability recovery and permitted stable operation.

Additionally, in scaling, when the solubility of salts is surpassed, slightly dissolved ions deposit and crystallization can occur on the membrane surface and operational equipment. The characteristics of mine waters possessing a low pH are dependent on the caliber of bedrock and its propensity to form AMD. Hence, either regulating the pH or addition of anti-scalants to the feed water can manage scaling [109]. Anti-scalants are pretreatment materials designed to remove scale and lessen fouling during membrane system processes, irrespective of the source of feedwater. Companies such as Suez are developing hyper-sperse anti-scalants to control scale precipitates and lessen particulate fouling within membrane separation systems. Outstanding outcomes are attainable in membrane separation processes like RO, NF, and EDR [109]. Genesys International [110] is another company involved in the development of anti-scalants that will help to ensure the plant runs as efficiently as possible. Despite the successful utilization of NF and RO membranes for the treatment AMD, there is the necessity for investigation at advanced level for particular application in a most appropriate system and the selection of membrane type. This is especially true if commercial spiral wound elements are used. Gu et al. [111] developed a new one-dimensional predictive model for spiral wound modules (SWMs) for reverse osmosis membrane systems by integrating a comprehensive description of the geometric characteristics of SWMs and taking into account the flow in two directions. It was observed that the proposed model was in accordance with existing experiment, with comparable accuracy to the widely used plate model in which the SWM was presumed to be made of multiple thin rectangular channels. Furthermore, operational conditions such as AMD feed pH and the rate of permeate recovery need to be considered. This type of evaluation would be directed to searching for ways of increasing retention efficiencies, minimize the formation of membrane fouling, cut budgets and improve the system as a whole [48].

ADVANCEMENT OF MEMBRANE TECHNOLOGY IN ACID MINE WATER TREATMENT

The technologies of NF and RO are developed, synthesized, and constructed for the elimination of dissolved ions and salts, not for particulate matter [112]. However, organic substances, particulate, and other solids that might not be adaptable with RO and NF membrane processes are frequently embodied in wastewater from mines. Hence, suitable pre-treatment for eliminating organic substance, particulates and any scale-forming pollutants is very vital for membrane performance and the length of span [52]. Roughing filters, primarily known as physical filters such as nanofilters, could be utilized to pre-treat AMD owing to their efficient separation of fine solid particles for extended spans without the inclusion of chemicals [113]. Furthermore, NF and RO technologies are efficacious in the removal of metal and sulfate from mine water; however, scaling can be a critical problem. Although a microfiltration system is used for the removing particulate, it also decreases the amount of scalants in order to delay scaling on the membrane surface and thus increase water recovery for NF and RO membrane processes when employed as pre-treatment [114].

With the advancement in membrane technologies, nanocomposite membranes can be used for mine water treatment because they produce a small amount of concentrated waste and drinkable water [115]. Additionally, there are many developed momentous membrane anti-fouling strategies. These strategies principally focus on membrane surface modification and membrane materials improvement for the purpose of minimizing the interaction between membrane materials and metal ions, which will further improve the anti-fouling property [116]. One of the strategies is the use of direct coating method to modify the membrane surface. This is done by incorporating nanoparticles into a porous polymer membrane [117]. The incorporation of nano fillers to these polymeric materials during the synthesis of membrane could be very effective for mine water treatment. For instance, two-dimensional carbon, graphene, having a one-atom-thick sheet of sp^2-hybridized carbon atoms arranged in a honeycomb configuration, has broad novel innovative ideas that can be used to prepare the next generation of membranes with exceptional separation capacities [118] which can be used to treat AMD. Furthermore, two-dimensional graphene materials have outstanding building block for the synthesis of porous membranes used for wastewater treatment. For example, spacing graphene oxide membranes have the potential to be specifically governed by adding cations, owing to the solid cation-π interactions amid hydrated cations and aromatic rings [21]. The spacing of membrane governed by a kind of cation can competently and carefully preclude other cations with substantial hydrated volume. For example, K^+- governed graphene oxide membranes have the capacity to effectively reject over 99% of Fe^{2+}, Mg^{2+}, Ca^{2+}, Na^+ and Li^+ in AMD and exhibited steady attainment beyond 24 h with a permeate flux of 0.36 L m^{-2} h^{-1} [119].

The joint outstanding performances of conventional membrane materials with those of carbon nanotubes have made carbon nanotubes-based composite membranes established membranes for separation of wastewater treatment. Additionally, carbon nanotubes exhibit promising adsorption, catalytic and electrochemical features. These features are advantageous to combine adsorption, catalytic or electrochemical role with membrane separation process, hence enhancing the performances of carbon nanotubes-based composite membranes for water treatment [120]. Presently, most carbon nanotubes-based composite membranes are useful in wastewater treatment and water desalination, which can now also be used to treat AMD due to their unique properties. They have attracted substantial consideration because of their unique features for improving the performance and capability of available recent membrane processes like NF [121] and RO [122]. Hence, to enhance membrane selectivity to monovalent and divalent ions, the incorporation of nano fillers to polymeric materials during membrane synthesis will enhance the quality of water from acid mine water and eradicate the compulsion of pre-treating AMD and further treatment phases.

MEMBRANES FOR BRINE MANAGEMENT AND THE RECOVERY OF METALS IN ACID MINE DRAINAGE

The management of brine concentrate is very difficult because of the waste product's composition, the necessity of managing the mass of liquids, and the solid generated. Hence, AMD has proven to have a weighty risk of acquiring significant costs, if not identified early and properly managed; however, the cost of treating AMD is location and circumstance specific [123]. For achieving sustainable mining rehabilitation, managing the brine and the environmental aspects all through the duration of the active mining operation is crucial [124]. The options of disposing brine concentrate are limited depending on the origin of the brine, its composition, location, and potentially substantial costs [125]. To completely avoid or lessen the difficulty of disposal, recovery opportunities of valuable products such as valuable metals, road salt of feed for electrolyte production of chlorine and caustic soda are very necessary. Nonetheless, the difficulty associated with discharging concentrated brine to the environment has not been solved. Over two decades, there has been continuous improvement in membrane science and technology, in order to find solutions to this environmental issue. In this regard, other membrane-based processes have appeared to be appropriate solutions. Among them are the thermally driven membrane process and membrane distillation (MD) process, which have been proposed to treat this high concentrated brine for water production and reduction of its volume [126]. Furthermore, RO and improved membrane system brine concentrator and natural treatment systems are among the technologies that can be used for brine concentrate management [125].

Metals in wastewater are one of the utmost vital contaminants in water because metals found in wastewaters are still valuable products that could be recovered from the residual waters and letting the reuse of a clean liquid stream into the production process [127]. Johnson and Hallberg [128] pointed out two crucial facts in selecting appropriate technologies to treat mine waters. The first fact is the basic consideration of mine water remediation as a valuable product. Hence, the encouragement of the recovery and recycling of the valuable products of mine water treatment. The second fact is the legislation that defines the discharge standards that could govern the selection of a system, in order to efficiently take out sulfate, metals, and acidity from mine waters. Membrane processing is a well-recognized technique for recovering and removing toxic metal ions. Reverse osmosis is usually employed for water recycling and recovery of metal [129]; however, RO is substituted by most promising techniques such as nanofiltration owing to the utilization of high pressure, the tendency to foul, and higher energy requirement. Nanofiltration methods are specially applied efficiently for bivalent metal recovery. The practical application of nanofiltration membrane technique for the removal and recovery of metals from wastewater is more effective owing to the lower operating pressure and higher flow rates [130]. Mullett et al. [51] investigated two nanofiltration membranes for their retention of ionic species from filtering contaminated mine water streams at a range of acidic pH values. From the recoveries of a range of cations, two-nanofiltration membranes were found suitable in relation to the compliance with mine water discharge standards and the recovery of valuable metal products. Hence, the NF process was confirmed to provide benefits in the recovery of metal from mine waste streams.

In addition, electrochemical processes are also considered quite appropriate for recovery of metals from diluted solutions because of their high separation efficiency, ability to recover metal and non-generation of sludge [131]. Furthermore, detailed parametric assessment by analyzing the concentration polarization of the separation system, the limiting current, and flux enhancement strategy for the overall recovery of acid constitutes important areas of electrochemical process development [132]. Hence, the development and application of electrochemical separation methods have increased.

CONCLUSION

Acid mine waters contain high concentrations of Fe^{2+}, Fe^{3+}, Ca^{2+}, SO_4; moderate concentrations of Al^{3+}, Ni^{2+}, Na^{2+}, Mg^{2+} and Mn^{2+} ions, which are dangerous to the environment, hence, the need to treat AMD. In the absence of an understanding of the terminologies that describe various aspects of membrane system operation and the relationship between these operating factors, the application of membrane technology for acid mine drainage treatment can be a complex subject. The consideration of factors such as AMD feed pH, concentrations of ions and thermal stability is very important when selecting membranes for the treatment of AMD because feed temperature, feed pH and concentration affect the selectivity of ions and permeate flux. The paucity of water has extended the prospects for advanced membrane technology for the treatment of AMD principally for metal recovery and water reuse. Hence, the need to improve the selectivity of membranes for the treatment of AMD.

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