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Improving acid mine drainage treatment by combining treatment technologies A review

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Review

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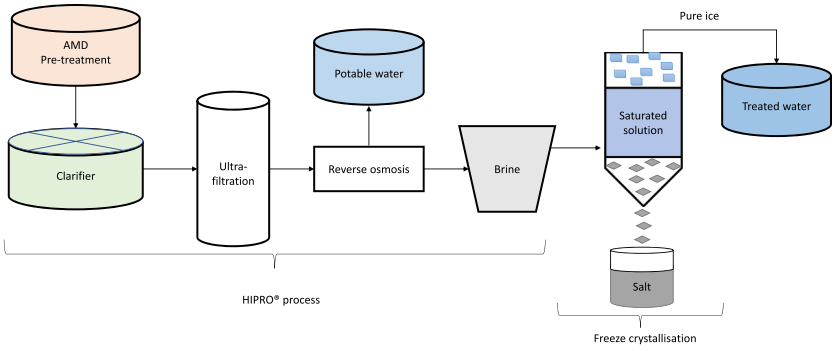
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HIGHLIGHTS

- No "one-size-fits-all" solution to mine waste treatment
- AMD treatment technologies that target different elements in AMD can be combined.
- Effluent of one AMD treatment technology can be the influent of another technology.
- Combinations of AMD treatment technologies can be selected using a designed software.
- The technologies will work efficiently when properly lined-up on application.

GRAPHICAL ABSTRACT



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ABSTRACT

The mining and processing of some minerals and coal result in the production of acid mine drainage (AMD) which contains elevated levels of sulfate and metals, which tend to pose serious environmental issues. There are different technologies that have been developed for the treatment of wastewater or AMD. However, there is no "one-size-fits-all" solution, hence a combination of available technologies should be considered to achieve effective treatment. In this review, AMD treatment technologies and the possible alignment in tandem of the different treatment technologies were discussed. The alignment was based on the target species of each technology and AMD composition. The choice of the technologies to combine depends on the quality of AMD and the desired quality of effluent depending on end use (e.g., drinking, industrial, irrigation or release into the environment). AMD treatment technologies targeting metals can be combined with membrane and/or ettringite precipitation technologies that focus on the removal of sulfates. Other technologies can be added to deal with the secondary waste products (e.g., sludge and brines) from the treatment processes. Moreover, some technologies such as ion exchange and adsorption can be added to target specific valuable elements in AMD. Such combinations have the potential to result in effective AMD treatment and minimum waste production, which are not easily achievable with the individual technologies. Overall, this review presents combinations of AMD treatment

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technologies which can work best together to produce optimal water quality and valuable products in a cost-effective manner.

1. Introduction

Many countries including South Africa, Bulgaria, Canada, Chile, China, Russia and the United States of America have been highly involved in the mining of coal and precious elements (Tiwary, 2001; Castro and Sánchez, 2003; Mudd et al., 2018; Bildirici and Gokmenoglu, 2020; Kjarsgaard et al., 2022). Some of these elements are used in several industries due to their unique physical and chemical properties and their demand has been increasing over the years (Humphries, 2010; Mudd, 2012; Seehra and Bristow, 2018; Acharya and Kharel, 2020; Moschovi et al., 2021; Rahmadianti et al., 2021; Cowley, 2023). Even though mining has a positive impact on the economy of the world, there are serious environmental issues associated with the mining of the elements, including, ecological deterioration and pollution that is challenging and costly to manage (Rybicka, 1996; Dong et al., 2019; Li et al., 2023). The mining and processing of precious elements generates a large amount of wastewater containing different substances including toxic elements (Cr^{6+}, As^{3+}, Cd^{2+} and Hg^{2+}), anions (e.g. sulfates), oil and grease which have a long lasting negative impact on the environment (Artioli, 2023; Fu et al., 2020; Tiwary, 2001). Because of this, several laws have been put in place to ensure that mining companies manage the waste they produce (Thomashausen et al., 2018). However, in the past, these laws were not in place, hence mining wastewater was released into the environment without any treatment or management, and the results are still felt to date (Egiebor and Oni, 2007). Thus, many countries depending on mining activities are faced with an urgent need of treating hazardous mining wastewater which is costly.

Acid mine drainage (AMD) is one of the most prominent mining waste products whose effects can last for decades if not treated (McCarthy, 2011). It is formed when sulfide-rich waste is exposed into the environment and is known for having a very low pH resulting from the formation of sulfuric acid, and a high concentration of elements (e.g., Al, Mn and Fe) (Egiebor and Oni, 2007). Gold and coal mining are the major contributors to the generation of AMD due to high content of sulfur-bearing minerals which are exposed to water, oxygen and bacteria in order to obtain the required mineral resource (Egiebor and Oni, 2007; McCarthy, 2011). Groundwater can infiltrate the underground mines and generate AMD as soon as it gets into contact with the sulfur-bearing minerals (Tiwary, 2001). In the case of opencast mining, water runoff resulting from rainwater and groundwater that infiltrate the mines has been found to be acidic due to the generation of AMD (Mal and Adhikari, 2021). The polluted water often ends up in other waterbodies thus, causing instabilities.

AMD is a serious problem that many countries are faced with, mainly due to the flooding of abandoned, closed and active mines (Park et al., 2019). Closed mines (underground or opencast) which are not properly managed contribute highly to water pollution caused by the mining industry, due to the formation of mining voids which can hold large amount of water (Park et al., 2019). The water in these voids can be discharged into nearby waterbodies including surface water and groundwater (Tiwary, 2001). Also, during mining, pits may intercept groundwater which necessitates pumping, however, when the mines are closed or abandoned, the pits will fill to groundwater level and deteriorate water quality due to ceased dewatering (Cidu et al., 2001). This automatically results in the contamination of nearby waterbodies. Moreover, the vicinity of the closed or abandoned mines continues to undergo environmental changes including the structure of the mine strata and groundwater recharge as well as degradation of the surrounding soils (Liu et al., 2020). There are >20,000 abandoned mines around the world and most of them result in AMD (Mohan and Chander, 2006; Rezaie and Anderson, 2020). There are thousands of abandoned mines in the United Kingdom which discharge mine water to waterbodies, including rivers (Johnston and Rolley, 2008). The discharged mine water contains metals and other pollutants in high concentrations. As a result, waterbodies in various places including England and Wales have failed to meet their water framework directive (WFD). The affected waterbodies have been found to lead to some of the biggest discharges of metals to other waterbodies near Britain (Johnston and Rolley, 2008). Coal and gold fields of abandoned mines in South Africa have also been found to discharge acidic water to nearby rivers where, high concentrations of toxic metals such as uranium and cobalt were detected (Mhlongo and Amponsah-Dacosta, 2016).

Residents living near mining activities are prone to the negative side effects resulting from AMD. Prolonged exposure can lead to serious health effects including cancer, diarrhoea, dermatitis, impaired morphology in infants and asthma among others (Yang et al., 2019). Aquatic plants and animals are also threatened by the presence of AMD. Ingestion of vegetation exposed to AMD is another way that humans can attain the side effects of AMD. To further confirm the seriousness of the effects of AMD, the U.S. Environmental Protection Agency (EPA) has categorised its environmental risks as second to global warming and ozone depletion (Acharya and Kharel, 2020).

Different mining sites which are active and inactive have varying AMD qualities and quantities which require different treatment strategies (Brake et al., 2001; Espana et a., 2005; Kefeni et al., 2017). The majority of AMD treatment methods and technologies are still in the development stage and cannot be applied in large scale as they are deemed non-economical due to their efficiency, cost and energy consumption (Simate and Ndlovu, 2014; Madzin et al., 2020). There are few technologies that have been successfully tested in large scale and have been found to be commercially feasible (Neba, 2006; Karakatsanis and Cogho, 2010; Mulopo, 2015; Dlamini et al., 2019). These technologies can adapt to different compositions and qualities of AMD however, they are not widely known in other countries. Thus, instead of trying to come up with new AMD treatment technologies that often take time to be approved or validated, the available technologies from different areas can be used or improved. Some of these technologies can be integrated with others in order to achieve the best treatment solution, but this has not been widely considered (Taylor et al., 2005). The treated water from the combined technologies can be used for irrigation and other applications depending on quality. This is very important especially in arid and semi-arid areas (Al-Hwaiti et al., 2016). Moreover, using mine treated water will reduce the rate of water scarcity due to reduced use of fresh water, thus, protecting water resources (El Ayni et al., 2011; Wang et al., 2022).

This review highlights synergies in technologies which can lead to improvements in acid mine drainage treatment (Masindi et al., 2017; Chatla et al., 2023). The synergistic approach is considered for AMD treatment technologies which are completed and/or tested in large scale. Some studies have indicated that more than one technology could be required to treat wastewater to the required or desired quality (Simate and Ndlovu, 2014). Jaén-Gil et al. (2021) found that combining biological processes with UV/H_2O_2 can significantly improve the removal efficiency of metoprolol and metoprolol acid from hospital wastewater. Vinardell et al. (2020) combined reverse osmosis, forward osmosis and anaerobic membrane bioreactor technologies for the treatment of municipal wastewater and concluded that the combined technologies improved the treatment capacity, and forward osmosis membrane fluxes of 10 LMH were achieved. These studies among others increase the attractiveness of combined technologies for the treatment of wastewater. Moreover, most technologies used to treat AMD deal with specific targets, for example, BioSURE™, a biological sulfate reduction

process, focuses mainly on sulfate removal, but there are other important pollutants that should be considered (Neba, 2006). Thus, combining the technologies can ensure that all the important and toxic pollutants in AMD are removed and that minimal secondary waste is produced.

The hypothesis of this review is that the synergistic approach of AMD treatment technologies can increase the efficiency of treatment and help reduce the adverse effects of AMD on the environment by producing less toxic waste, which is supported by the different research and review manuscripts which are cited herein. Some of the treatment technologies which are discussed have been commercialised due to their high efficiency, but are not known throughout the world. This review will help with increasing the visibility of such technologies. Moreover, it is crucial for mining companies to consider the idea of using a combination of technologies for the treatment of AMD to prevent high output of secondary waste, which is normally released into the environment. Thus, using a combination of technologies will ensure that the treatment of AMD is effective and environmentally friendly, therefore reducing environmental stress.

2. Main components of AMD

The mining of gold, coal, copper, nickel and silver exposes sulfur minerals (pyrite and pyrrhotite) to air, water and microorganisms (e.g., acidophilic bacteria) (Tong et al., 2021). This leads to the oxidation of sulfur, resulting in the formation of sulfate, sulfuric acid and the dissolution of metals (Eq. (1)) (Akcil and Koldas, 2006; Fernando et al., 2018). The resulting high concentration of acid produced, often leads to the weathering of the surrounding rocks which results in the leaching of elements such as zinc, copper etc. into the water (Madzivire et al., 2011). Thus, AMD normally contains high concentration of sulfate and metals at low pH.

Mining operations in South Africa, Mexico, China, Ireland, Canada, Germany, Spain and France have reported high sulfate concentrations (>2000 mg L^{-1}) which have also been detected in natural water resources (Fernando et al., 2018). The World Health Organisation (WHO) has established that consumable water should not have >250 mg L^{-1} sulfate. It is therefore crucial for mining companies to treat wastewater before discharge into the environment (Fernando et al., 2018). Due to this, many researchers have developed methods and technologies that can remove sulfate from AMD (Costa et al., 2008; Zheng et al., 2020). AMD treatment techniques focusing only on the removal of sulfate lead to the generation of secondary waste which often contains very high concentrations of other elements some of which are toxic even in very low concentrations (e.g., Cd^{2+}, Pb^{2+} and As^{3+}) (Rodríguez-Galán et al., 2019). Thus, technologies which can target more pollutants are of high importance and crucial in addressing the global availability of clean water which has become a scarce commodity.

$$2FeS_2\ (\text{pyrite}) + 7O_2 + 2H_2O \longrightarrow 2Fe^{2+} + 4H^+ + 4SO_4^{2-} \quad (1)$$

3. Effect of AMD on humans

Soils, sediments, food crops and waterbodies have been found to be heavily affected by toxic and non-toxic metals from AMD (Liao et al., 2016). Humans can ingest metals through the food chain, since some farmers use untreated groundwater or surface water for irrigation purposes (Liao et al., 2016). These metals can accumulate in the human body and may cause serious health issues over time. Non-essential and toxic metals such as cadmium (Cd^{2+}), lead (Pd^{2+}), chromium (Cr^{6+}), arsenic (As^{3+}) and mercury (Hg^{2+}) have often been found in high concentrations where AMD is concerned. Cadmium is carcinogenic and can also inhibit transport pathways, cause cell death, lead to organ dysfunction and interfere with the physiologic action of Zn or Mg in the body (Satoh et al., 2002; Bernhoft, 2013). Lead has been found to cause damage to the brain and kidneys. It can also lead to high blood pressure, anaemia and miscarriage (Ab Latif Wani and Usmani, 2015). Liao et al. (2016) studied the effect of metals (cadmium, copper, zinc, arsenic and lead) emanating from the Dabaoshan mine in Guangdong Province, China. The metals were high due to AMD, and highly affected the soils and crops (sugarcane, paddy rice and vegetables) in the vicinity. The local inhabitants have been faced with high cancer mortality for years due to ingesting affected crops with high concentrations of toxic metals. Despite these results, the inhabitants were still forced to plant crops so that they have the daily provision for food. Kagambega et al. (2014) assessed the effect of stockpiles of waste from a gold mine in Poura, Southwest Burkina Faso which has been operating for a decade. The results of the study indicated that the exposure of the waste to water and the atmosphere results in AMD with a series of dissolved toxic metals. The metals (arsenic, cobalt, chromium, copper, lead and nickel) from AMD were detected in the surrounding waterbodies and soils. Abandoned mines in the United States are known to produce AMD which contains a wide range of metals which affects >23,000 km of streams (Kim, 1982; Skousen, 2014). Thus, nearby populations are negatively affected. Some metals such as copper, zinc and iron are essential to the human body, but they can also be toxic once they exceed the maximum allowed limit (Rai et al., 2019). For example, high concentrations of copper have been found to affect renal and metabolic functions in humans (Hough et al., 2004). Zinc (Zn) is considered a multipurpose element due to its ability to bind >300 enzymes (Chasapis et al., 2020). It is also involved in deoxyribonucleic acid (DNA) replication and DNA damage repair however, it can lead to respiratory problems when the allowed limit in the body is exceeded (Hough et al., 2004). Thus, the removal of toxic elements in AMD is of high importance.

4. Deriving value from AMD

It has been estimated that about 50–100 % of resources which are lost during mining, processing and manufacturing can be found in wastewater, hence valuable elements have been detected in AMD (Puyol et al., 2017). Thus, AMD can be considered as a potential source of valuable elements which can be recovered and re-used to prevent high natural resource depletion. Currently, there are many resource limits due to increasing population and urbanisation, therefore, the recovery of valuable elements from wastewater is a great alternative. Rare earth elements (REEs) which occur in large deposits in few countries, have been found in some AMD waters. Thus, AMD has been proposed as the secondary source of REEs. As an example, AMD resulting from uranium mine in Minas Gerais State, Brazil, has been found to contain >100 mg L^{-1} of REEs (Felipe et al., 2021). The price of REEs has increased sharply over the last decade and their supply has also been restricted by China who is the main supplier. Because of this, many studies have been conducted in order to come up with cheap and efficient ways to recover REEs so that demand can be met (Felipe et al., 2021). Copper, cobalt, nickel, manganese and zinc are some of the elements that can be recovered from AMD. Moreover, the production of these elements has been increasing annually, hence demand might exceed supply in the future. Copper is used for the transmission and distribution of electricity in many parts of the world (Henckens and Worrell, 2020). Nickel is used for coating stainless steel and metal alloys as it can protect them from corrosion (Costa et al., 2022). Nickel is also an important element in military, transport, architecture, aerospace and marine technological advances (Kurniawan et al., 2021). Cobalt is another element that is very fundamental in today's technology. It is one of the most important components in the production of electric cars since these cars require lithium-ion batteries which demands cobalt to function (Campbell, 2020). Cobalt is also used in catalysts, magnets, superalloys and tool materials. However, >40 % is used in rechargeable batteries (Alves Dias et al., 2018). Zinc is also an important element which is used to prevent corrosion of steel and iron. Moreover, it is also used in other applications including alloys and batteries (Van Niekerk and Begley, 1991).

Due to the sulfide-bearing minerals, AMD contains large amount of sulfuric acid which can be recovered and re-used in other processes such

as leaching elements such as copper, cobalt, nickel etc. from their respective industries (Nleya et al., 2016). Sulfuric acid is also widely used in electroplating, steal making, paper bleaching, gasoline, sugar bleaching and many other industries. Recovering sulfuric acid from AMD will also be good for the environment since it is very corrosive. Removing acid from AMD will also be important since neutralising agents such as lime and limestone that are normally added result in huge amount of sludge that should be disposed of (which is often challenging).

Table 1 shows the concentrations of some valuable elements in AMD from different locations. These concentrations are significant and will add value when recovered. Thus, suitable and efficient recovery techniques are required. In order to select a suitable recovery technique, the quality of AMD must be known since a combination of technologies may be a necessity in some places. For example, AMDs in South Africa contain high concentrations of sulfate ions or sulfuric acid, therefore metal recovery technologies should factor in the influence of sufates (Madzivire et al., 2011). Moreover, some of the targeted elements may be in lower concentrations than competing elements, hence selective techniques will be highly crucial. The selected techniques should be able to adapt to changing conditions such as pH, acidity, temperature and composition. Many studies have focused on the recovery of these elements but very few methods have been commercialised due to feasibility (Hassas et al., 2020; Sephton et al., 2019; Mokgehle and Tavengwa, 2021). Though most studies have indicated success on the recovery of the elements, they are not applicable in large scale due to high costs resulting from cementation, high energy consumption, high cost of chemicals and producing parts of the techniques such as adsorbents and solvents (Gaikwad et al., 2010). Hence, very few technologies for the recovery of elements and sulfates from wastewater have been approved and commercialised. Also, in many developing countries, there are no water treatment plants that can efficiently remove trace elements from water.

In some cases, treated wastewaters containing particular concentrations of metals and sulfates can be used in some processes thus, avoiding the use of potable water where lower quality water can be used (Van Zyl et al., 2001). This means that some technologies leading to a certain quality of water for a particular process can be used to produce the suitable water so that natural groundwater and surface water can be avoided. Water quality guidelines can be used to determine the suitable quality of water for an application and this may be particular to a specific country or district (Grewar, 2017). Fig. 1 shows the concentrations of sulfate in water allowed for some potential applications (Van Zyl et al., 2001). It should also be noted that as the purity of the treated water increases, the cost of treatment also increases.

5. AMD treatment methods and technologies

Various AMD treatment methods and techniques can produce different qualities of treated water (Brake et al., 2001; Espana et al., 2005). The treated water can be used for different purposes including irrigation, drinking (potable), mining processes or released into the environment, where it can end up in waterbodies (Kuyucak et al., 2003; du Preez, 2021). The uses are often determined by the treatment methods used and the quality of water produced. Many countries especially those in arid and semi-arid regions are faced with water scarcity problems, hence re-using water is of high importance. Water scarcity in such regions is also escalated by high population growth, urbanisation and high living standards which drastically increase consumption and threatens supply (El Ayni et al., 2011). This section focuses on AMD treatment methods and technologies which are widely used and have been found to be successful and efficient. Moreover, different commercialised and pilot scale tested AMD or mine water treatment technologies are provided. Some of the technologies have been tested in large scale and have a technology readiness level (TRL) >7, meaning that they have been completed and tested in small and large scale to confirm their eligibility (Meystel et al., 2003).

5.1. Adsorption

The metals in AMD are usually targeted separately from other constituents such as sulfate ions since they behave differently. The adsorption technique which is basically the transfer of solids from the aqueous phase onto the solid phase has been widely studied and found to be very efficient in removing metals from AMD (Ighalo et al., 2022). It is also preferred because there is minimal waste produced and most adsorbents can be re-used. Different natural and modified adsorbents including zeolite, saw dust, manure compost, eggshell, fly ash and nano materials have been used (Table 2) (e.g., magnetic graphene oxide) (Motsi et al., 2009; Mokgehle and Tavengwa, 2021; Ighalo et al., 2022). Though cutting-edge research has been done using different adsorbents, very few adsorption techniques have been tested or used in large scale. This may be due to the impracticality of producing large amounts of adsorbents that should treat the amount of AMD available. Also, most studies are in batch mode or column mode in small scale thus, the possibility and feasibility of large scale is not clear.

5.2. Sulfate reducing bacteria

Bioremediation, a process that requires algae strains or microbes such as bacteria to remove elements from wastewaters has been used for

Table 1
Approximate chemical composition of AMD from selected locations around the world.

Place and location	SO_4^{2-} (mg L^{-1})	Zn (mg L^{-1})	Ca (mg L^{-1})	Co (mg L^{-1})	Ni (mg L^{-1})	Fe (mg L^{-1})	Mg (mg L^{-1})	Mn (mg L^{-1})	Al (mg L^{-1})	REEs (mg L^{-1})	pH	References
Gauteng western basin, South Africa	3500	–	800	2	4	750	120	100	50	–	–	Agboola et al., 2017
Mpumalang, South Africa	42,862	8.4	599	189	3	8158	399	88	474	–	2.5	Madzivire et al., 2011
Poderosa Mine, Brazil	–	101	161	1.4	0.3	1535	1826	–	375	0.01–3.3	3.3	Hermassi et al., 2021
Southeast of Brazil (Caldas Municipality, Minas Gerais State)	999	13	182	–	–	5	10.4	87	147	0.03–49.9	3.4	Felipe et al., 2021
Chessy-les-Mines mine-site, France	5800	320	–	0.06	0.4	70	–	5.5	210	–	2.6	Foucher et al., 2001
Copper mine, China	2570	22.5	189.7	–	–	292.6	225.4	–	112.5	–	2.5	Fu et al., 2020
Sweden	1278	0.068	397	20	69	6.3	57.3	5.2	1.09	–	3.2	Grawunder et al., 2014
Brukunga mine site, South Australia	5465	24	–	–	–	479	–	35	526	–	2.8	Sephton et al., 2019
Pennsylvani, U.S.	2.03	–	178	0.72	1.21	0.17	324	33.4	33.7	0.012–0.16	3.7	Hassas et al., 2020

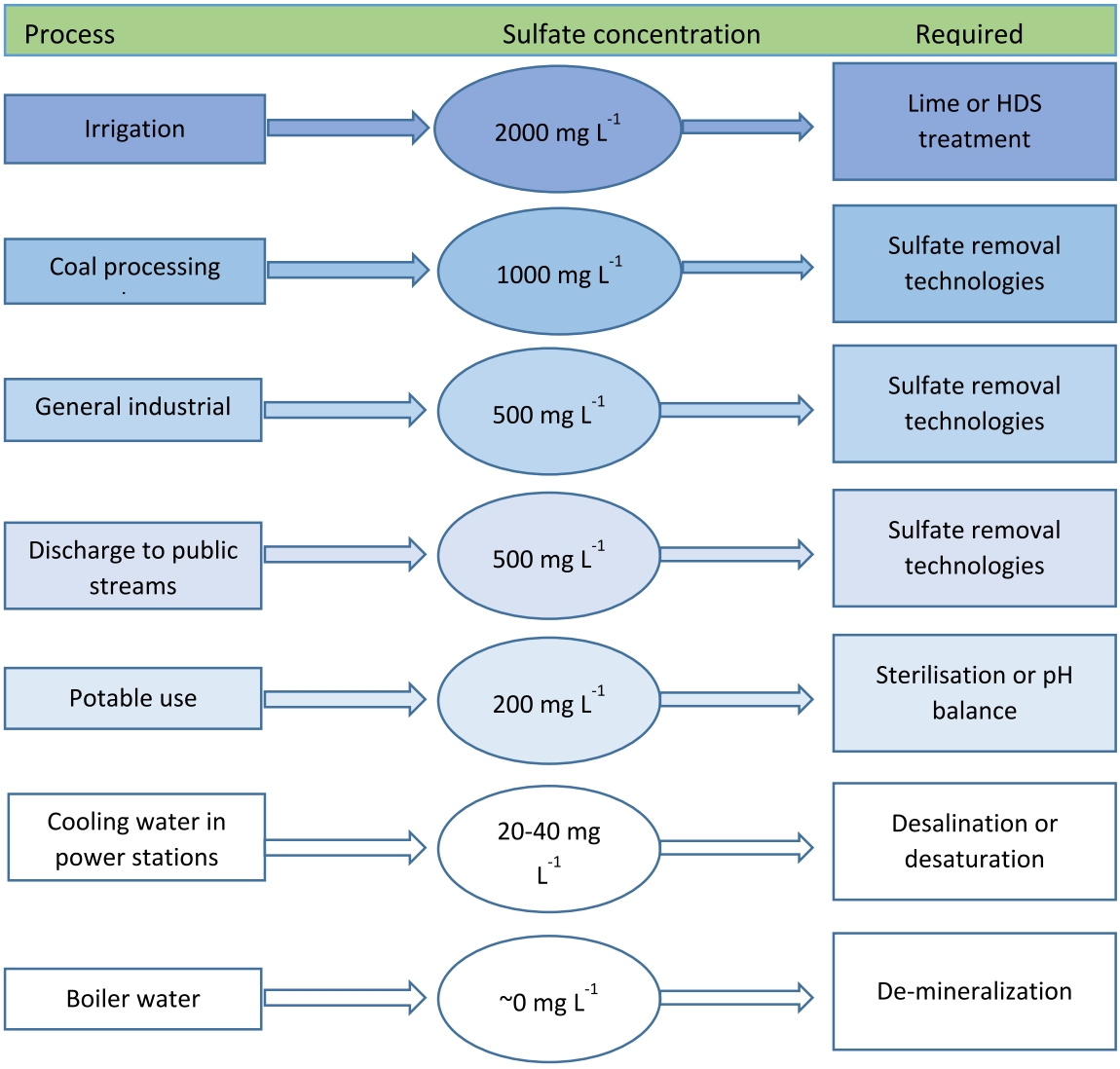


Fig. 1. The application of treated water with different sulfate concentrations (Van Zyl et al., 2001).

the treatment of AMD. Sulfate reducing bacteria (SRB) are widely used microbes in AMD treatment as they can reduce sulfate to sulfide when supplied with a suitable organic carbon source such as lactate, glucose, sucrose and methanol (Eq. (2)) (Postgate, 1979). The carbon source is used as an electron donor and sulfate acts as a terminal electron acceptor (Gibert et al., 2002). The energy resulting from this reaction is used by the bacteria for growth and development (Postgate, 1979). SRB uses the AMD excessive sulfate ions as terminal electron acceptors during the metabolism of the organic matter (Eq. (2)) (Costa et al., 2008). The utilised sulfate is then converted to hydrogen sulfide (H_2S) which interacts with metals, leading to highly stable metal sulfide precipitates (Foucher et al., 2001). This allows for the ease of removal of metals from AMD using filtration.

This process has been found to be cheap, efficient and easy to operate (Rambabu et al., 2020). Moreover, the metabolism of SRB leads to the neutralisation of AMD since it generates alkalinity.

$$Carbon\ source\ (e.g., 2CH_2O) + SO_4^{2-} \rightarrow 2HCO_3^- + H_2S \quad (2)$$

Bai et al. (2013) used SRB for the treatment of AMD utilising bench scale runs. In this study, iron (Fe^0) was added to enhance the activity of SRB. The results indicated that 61 % of the sulfate in AMD was removed as well as some elements (Fe^{2+}, Cu^{2+} and Mn^{2+}). The treatment lasted for 70 days without the replenishment of biomass. An article by Gibert et al. (2002) indicated that SRB can also be applied in permeable reactive barriers (PRBs) for the treatment of AMD impacted groundwater. The use of SRB in PRBs in large scale was first proposed in 1995 where, a full-scale reactive barrier was installed at Nickel Rim mine site, Canada, for the treatment of AMD. The reactive barrier was installed into an aquifer with suitable reactive material to allow for the physico-chemical and biological processes that can remediate groundwater. The reactive material used was made up of pea gravel and a mixture of municipal compost, leaf compost, wood chips and small amounts of limestone. Extensive monitoring system was also installed to monitor important the physical and chemical (pH, concentration of metals, concentration of sulfate, etc.) parameters. The article indicated that the removal of sulfate and metals from groundwater continued for years. After three years of installation, 30 % of the initial sulfate concentration was removed. The majority of AMD treatment technologies using SRB have been applied in small scale, therefore their large-scale capabilities are blurry. Some of the few efficient AMD treatment technologies utilising SRB are presented below. These technologies have been applied in large scale and are commercialised.

5.2.1. CloSURE™

CloSURE™, developed by Mintek, South Africa, is a SRB technology for the treatment of AMD with sulfate content (du Preez, 2021). It

Table 2
Adsorbents used for the removal of metals from AMD.

Adsorbent	Metals targeted	Adsorption capacity (mg g-1)	Adsorption mode	Reference
Chicken eggshell	Cd^{2+}	1.57	Column	Zhang et al., 2017
	Cu^{2+}	387.51		
	Pd^{2+}	146.44		
Dairy manure compost	Cu^{2+}	0.460 mmol g-1	Batch	Zhang, 2011
	Pb^{2+}			
	Zn^{2+}	0.428		
		0.237		
Poly (hydroxamic acid) ligand	Cu^{2+}	14	Batch	Mzinyane, 2022
	Fe^{3+}	13		
	Mn^{2+}	11		
	Pb^{2+}	12		
	Zn^{2+}	11		
Un-activated attapulgite	Co^{2+}	0.0044	Batch	
	Cu^{2+}	0.0053		
	Fe^{2+}	0.01		
	Mn^{2+}	0.0019		
	Ni^{2+}	0.0053		
Double-oxidised multiwalled carbon nanotubes	Cu^{2+}	14	Batch	Rodríguez and Leiva, 2019
	Mn^{2+}	6.6		
	Zn^{2+}	4.0		
Zero-valent iron/ phosphoric titanium dioxide	Cd^{2+}	308	Batch	Ren et al., 2022
Graphene oxide–ZnO nanocomposites	Al^{3+}	29.1	Batch	Rodríguez et al., 2020
	Cu^{2+}	45.5		
Molybdenum sulfide modified chelating resin	Cd^{2+}	429	Batch	Fu et al., 2020
	Cu^{2+}	369		
	Hg^{2+}	1315		
Lucerne biochar	Cd^{2+}	6.28	Batch	Bandara et al., 2020
	Cu^{2+}	18.0		
Grafted cross-linked chitosan beads	Cd^{2+}	540	Column	Igberase et al., 2018
	Cr^{2+}	360		
	Cu^{2+}	810		
	Ni^{2+}	630		
	Pb^{2+}	630		
	Zn^{2+}	270		

consists of two processes viz biological sulfate reduction step and oxidation step for sulfide removal and biosulfur production (Fig. 2). SRB can reduce sulfate by disassimilation, since they use sulfate as their electron acceptor for the degradation of organic matter (Niu et al., 2018). During this process, sulfate is reduced to sulfide which can interact with free metals to form metal sulfides which can be easily removed from solutions due to their stability and low solubility (Utgikar et al., 2002). High concentration of sulfide can lead to the corrosion of equipment which can pose safety risk to operators hence, CloSURE™ has a second step which focuses on the biological removal of sulfide from the process (du Preez, 2021). Toxic metals such as cadmium, uranium and mercury can be bio-transformed by SRB. Thus, toxic metals can be removed from AMD through the CloSURE™ technology. CloSURE™ technology is highly suitable for remote locations with no proper services and infrastructure e.g., closed mines and abandoned mines since, it mainly uses anaerobic prokaryotic microorganisms that can be accessed in natural environments (rivers, lakes, sediments etc.). As a result, the technology is very cheap (i.e., low capital and operational costs) and produces significantly less solid waste (du Preez, 2021). Moreover, the resulting treated solution contains very low toxicity and increased stability as compared to conventional chemical precipitation methods (Grewar, 2017). This technology has been applied in laboratory and pilot scales (i.e., in mine sites) (Van Rooyen et al., 2021). It has been found to remove >85 % of sulfate and sulfide with average sulfate reduction rate of 196 g m^{-3} d^{-1}. Moreover, other elements including aluminium, zinc and nickel can also be removed in high quantities (du Preez, 2021).

5.2.2. BioSURE

BioSURE is a biological sulfate reduction process developed by Rhodes University, South Africa, for the treatment of AMD (Neba, 2006). During treatment, sulfate-rich AMD is mixed with a primary sewage sludge which acts as a source of the most freely available carbon source and electron donor (Rose, 2013). There are two stages in the BioSURE process and each is associated with a reactor (Fig. 3). In the first stage, a multi stage reactor which acts as a dual stage sulfate reduction operation is used (Neba, 2006). The primary sewage sludge (PSS) is solubilised and small organic molecules are generated in the first stage. Thus, the purpose of the first stage is mainly to enhance the hydrolysis of PSS as electron donor and carbon source (Rose, 2013). The solubilised PSS is transferred onto the second stage, where the sulfate reduction reaction is optimised using a baffle reactor. Some of the effluent from the anaerobic baffle reactor (reactor 2) which is alkalised and rich in sulfide would pass on to the AMD treatment operation where neutralisation and precipitation of elements occurs. There is also a high rate of algal pond where polishing and disinfection of the final treated water occurs (Rose, 2013). More than 90 % of sulfate removal can be achieved with this process and >10 ML of wastewater can be treated per day. BioSURE is designed to remove sulfate to levels below 250 mg L^{-1}, with the removal of >12 tons per day of sulfate. The residual organic material and nitrogen present in the treated effluent are removed before discharged. To reduce the odour emissions from this process, lime is added to the produced biological sludge (Rose, 2013).

5.2.3. Integrated managed passive treatment process technology

The integrated managed passive treatment process technology (IMPI) is a patented technology which has been widely used because it is self-sustaining, requires less maintenance and it is cheap since it uses available energy sources (e.g., topographical gradient, microbial metabolic and chemical energy as well as photosynthesis) (Pulles et al., 2004; Molwantwa et al., 2010). There are four stages involved in the treatment process (Coetser et al., 2004). The first stage involves the degrading packed bed reactor which is packed with several layers of carbon sources (electron donors) and receives readily available carbon which conditions the received AMD by removing dissolved oxygen in the first portion of the reactor (Pulles et al., 2004). Moreover, the desired redox conditions are established whilst high levels of sulfides and alkalinity

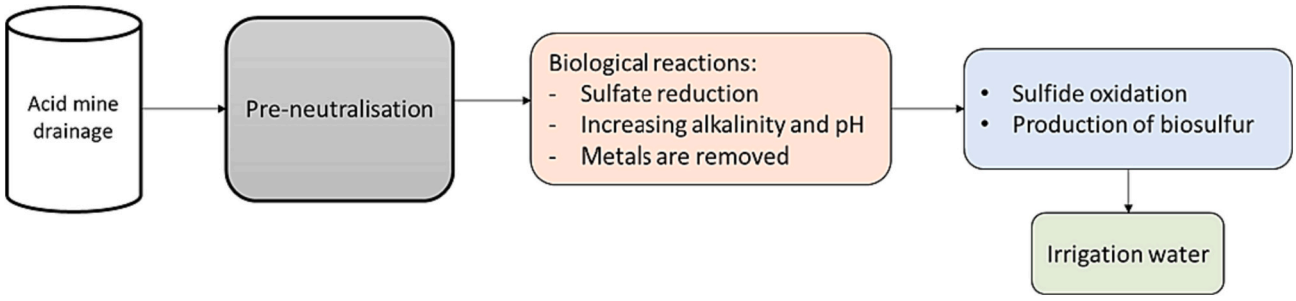


Fig. 2. Processes involved in the treatment of AMD using the CloSURE technology (Utgikar et al., 2002; du Preez, 2021).

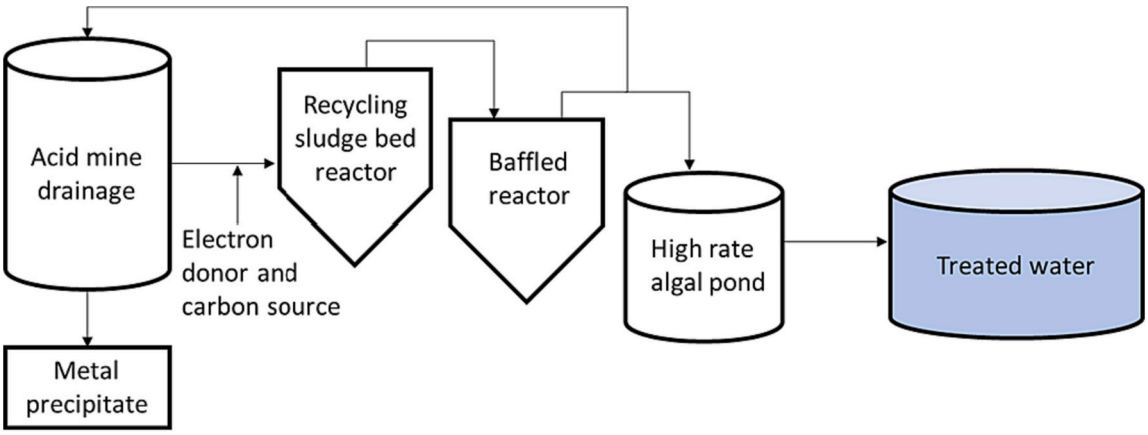


Fig. 3. BioSURE process flow for the treatment of AMD (Neba, 2006; Rose, 2013)

are produced. In the last portion of the reactor, volatile fatty acids are produced and the hydrolysis of lignocellulose is optimised. The effluent from the reactor will contain less metal and sulfate content but, high levels of sulfides, alkalinity, volatile fatty acids and nutrients (Pulles and Heath, 2009). The second stage uses the primary sulfide oxidising bioreactor where sulfide from the previous reactor is oxidised to elemental sulfur which is removed from the reactor. The third stage involves the secondary sulfate reducing reactor where, the remaining sulfate is reduced to the desired concentration using specially selected single carbon source (Pulles and Heath, 2009). The effluent from this reactor is transferred into the secondary sulfide oxidising bioreactor (stage four) where sulfide is oxidised to elemental sulfur and removed from the reactor. The technology can treat 1 ML of AMD to remove 1 ton of sulfate per day. During the process, between 87 and 97 % of metals can be removed. The technology can last for a minimum of 15 years (Pulles et al., 2004).

5.2.4. Fixed-bed column bioreactor

Touze et al. (2008) designed a 200 L pilot plant to treat AMD from an abandoned Chessy-les-Mines (CLM) mine site in the Rhône department, France, by reducing sulfate to sulfide over 36 days. The sulfate concentration on the site was between 1500 and 6000 mg L^{-1}. Metals such as Fe, Al, Cu, Mn and Zn were also in high concentrations. A fixed-bed column was used and filled with pozzolana which was inoculated with *Desulfomicrobium norvegicum*, a sulfate reducing bacteria. Hydrogen gas was used as the electron donor in order to establish a low-cost process. The pilot plant was designed to treat 5000 mg L^{-1} of sulfate at a rate of 20 L h^{-1}, using ambient conditions. In the first phase of the process, AMD effluent is transferred into a mechanically agitated tank where, NaOH is added to precipitate the metals. The resulting supernatant is fed into another tank and mixed with the outflow liquid of the bioreactor. During this process, hydrogen sulfides produced by the bioreactor are used to precipitate the metals which remained after the first precipitation process. In the second phase, the supernatant from the second precipitation is transferred into a down-flow fed bioreactor containing nutritive elements which are 50 times more than the supernatant. The final results indicated that >80 % of metals were removed and the sulfate content was reduced by 75 %. The economical evaluation showed that 0.88 USD would be required to treat 1 m^3 of AMD.

5.2.5. Anaerobic stirred batch reactor

Castro Neto et al. (2018) used anaerobic stirred batch reactor (ASBR) to treat AMD based on the activity of sulfate reducing bacteria. A 7.0 L ASBR reactor with an operational volume of 5.5 L was used. The reactor had a Fiberglass jacket and a water circulation system in order to control the reaction temperature. 1.0 L of granular sludge biomass was added to the batch reactor at the beginning of the process and no further addition was required. Ethanol was used as the source of carbon and electron donor. The results indicated that after 218 days, the highest removal efficiency of sulfate from AMD was 65 % from the initial concentration of 1500 mg L^{-1}, and the removal efficiency of metals such as Fe, Zn and Cu was >99 %.

5.3. Membrane technology

Research has found that the use of membrane technology to treat AMD is a good alternative to conventional methods that have been used. As a result, many studies involving reverse osmosis, ultrafiltration and nanofiltration membrane technologies have been conducted (Sierra et al., 2013).

Nanofiltration (~1–10 nm) which is known as borderline between reverse osmosis (~0.1–1 nm) and ultrafiltration (~10–100 nm); uses different highly water permeable membranes (e.g. NF90, NF99, NF270 and NF2540) to separate metals from AMD. It ensures that metals and salts from the feed solution are retained (Sierra et al., 2013). The performance and lifespan of the membranes are based on the pre-treatment of the feed solution which is often required so that particulate matter or any scale forming contaminants can be removed (Agboola et al., 2017). Using membrane technology also allows for the separation of monovalent ions from multivalent ones since it offers preferential passage of monovalent ions whilst rejecting multivalent ions such as transition metals and rare earth elements which, many conventional methods are incapable of doing (Siew et al., 2020). Thus, nanofiltration can be used to concentrate and recover specific elements through selectivity. Though this is good where selectivity is required, it will be a problem where the overall idea is to treat AMD. The main advantages of using nanofiltration include short operating times, low operational costs, low energy consumption, high flux, low pressure requirement and the ability to adapt to AMD changes such as the concentration of elements and temperature (Aguiar et al., 2018; Sierra et al., 2013). The main issue with using nanofiltration has always been membrane fouling which depends mainly on the properties of the AMD, surface properties of the membrane and the conditions used for operation (Agboola et al., 2017). In most cases, a pre-treated is required to remove suspended solids so as to reduce the possibility of membrane fouling. Nanofiltration is a pressure-driven filtration membrane process that operates by retaining elements and salts from the feed using size exclusion process. The thermal, chemical and mechanical properties of the polymer membranes determine the durability and effectiveness of the nanofiltration membranes (Agboola et al., 2017).

Nanofiltration processes are also good for AMD treatment since they have the ability to concentrate multivalent ions in their retentate side whilst allowing monovalent ions to pass through to the permeate side (López et al., 2020). For example, HSO_4^- and H^+ can be allowed to

penetrate through so that the permeate side contains the acid which can be recycled and used again in leaching processes (Fig. 4). This is a good method to also recover valuable multivalent ions such as REEs, Mn, Zn etc. which are concentrated in the retentate side. Companies such as Dow Chemical and Desal DL have developed commercial nanofiltration membranes for such processes (López et al., 2020).

Unlike nanofiltration membranes, ultrafiltration membranes can separate macromolecules (Galanakis, 2015). Ultrafiltration is a fractionation technique that is driven by pressure difference (Mehta and Zydney, 2005). It can simultaneously purify, concentrate and fractionate macromolecules or colloidal substances in process stream (Hlangwani et al., 2023). Ultrafiltration membranes have a pore size which is between microfiltration and nanofiltration (Zhang et al., 2019). The membranes have been used in water treatment since they can remove suspended particles, colloids and microorganisms with high efficiency, and the treated water is normally of high quality (Ma et al., 2019). Moreover, ultrafiltration techniques have been widely used because they are cost efficient, easy to operate, consume less energy than other filtration techniques and they are environmentally friendly (Wu et al., 2021). When ultrafiltration membrane is applied, the unwanted particles such as metals are separated from the solution through a semi-permeable membrane using low pressure hence minimum energy consumption. Since it is also prone to fouling, which limits its application, many studies have been conducted to focus on organic pollution, inorganic pollution, particulate/colloid pollution and biological contamination which leads to fouling. As such, ultrafiltration membranes have been modified to increase their efficiency, making them to remain one of the preferred methods for wastewater treatment including AMD (Zhang et al., 2019).

Reverse osmosis is a membrane technology that uses osmotic pressure to force a solute concentrated solution through a membrane into a region where no pressure is applied. The membrane used is semi-permeable since it only allows the solution to pass through whilst the solutes are prevented from passing through. The reverse osmosis membranes have a dense barrier in the polymer matrix which allows for the separation of solutes from the solution (Bakalár et al., 2009). There are some issues associated with the reverse osmosis process including fouling, sensitivity to scaling, production of large amount of brine which is expensive to dispose, and highly concentrated solutions require high amount of energy (Masindi et al., 2017). Pre-treatment is often required to avoid all this which, inflates the process costs.

5.3.1. *Application of ultrafiltration and nanofiltration membranes*

The Nanotechnology Innovation Center (NIC) at Mintek, South Africa, designed NICMembrane™, a semi-permiable and low-fouling capillary ultrafiltration membrane that separates solutes from a solution using size-exclusion (Fig. 5) (Petterson, 2018). It is made up of bulk functionalised polyethersulfone (PES) powder, however it has high hydrophobicity than a typical PES membrane (Petterson, 2018; Steenhoff-Snethlage, 2018). It is designed in such a way that it has high surface porosity and narrow pore size which lead to high efficiency and low energy consumption. The membrane also has a lower fouling propensity, meaning that it can remain productive for longer (Steenhoff-Snethlage, 2018). When applied, it can lead to an effluent which used in the mining processing plant. Moreover, NICMembrane ™ can be used for the removal of particulates and pathogens from grey water to produce drinkable water (Parker, 2023). NICMembrane™ is cheap, efficient and the energy consumption is very low (Steenhoff-Snethlage, 2018). The lower fouling ability makes it suitable for use in severe wastewaters and the membrane remains productive for longer periods of time.

Fonseka et al. (2022) used low pressure (3 bar) nanofiltration with NF90 membrane to recover fresh water from AMD. A 2.0 L feed volume was used including a rectangular cross-flow cell with an area of 54 cm^2. The presence of organic matter was found to cause membrane fouling and to prevent this from happening, powdered eggshells were used to adsorb organic matter before introducing the AMD to the membrane. The results of the study showed that 80 % of water was recovered with >95 % solute rejection. Wadekar et al. (2017) also used NF90 membrane

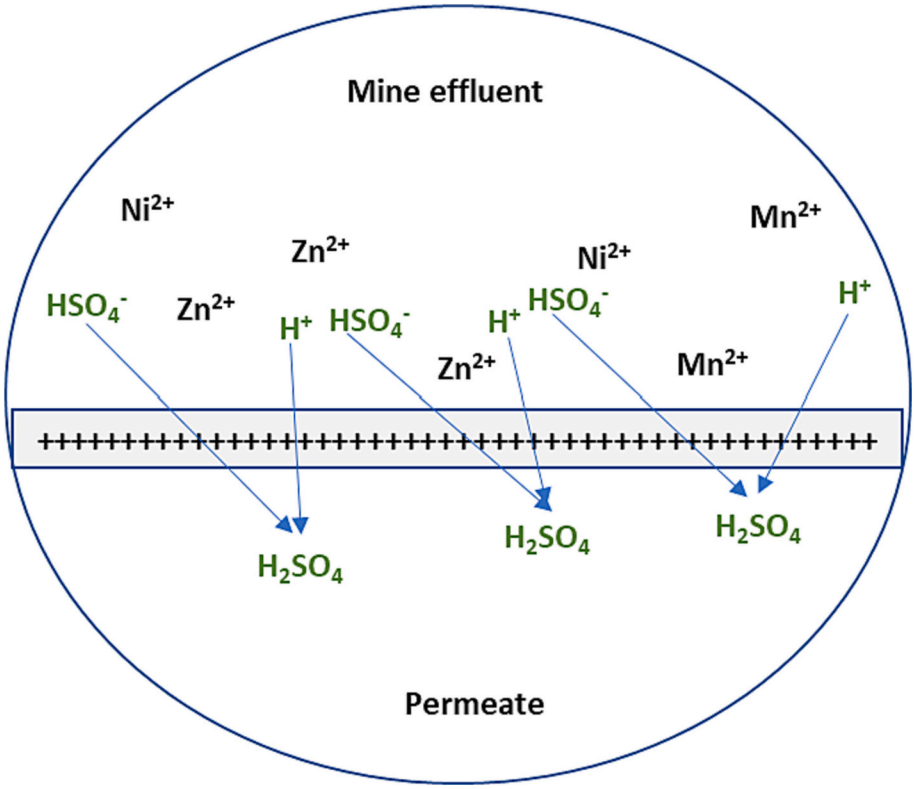


Fig. 4. Recovery of sulfuric acid using nanofiltration method.

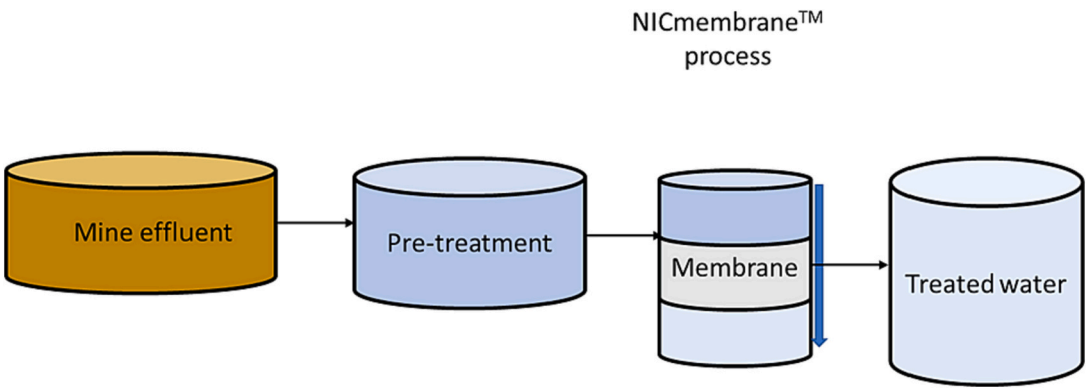


Fig. 5. Simplified schematic of a NICMembrane™ demonstration plant (Steenhoff-Snethlage, 2018).

in nanofiltration treatment of AMD from Pennsylvania, USA at pilot scale. The NF90 used had aeration and sedimentation for iron control and bag filtration. The process was operated at 10 bar using a feed flow rate of 3.5 gal per minute. About 57 % of water recovery was achieved with >98 % of TDS removal. Moreover, there was no evidence of fouling and/or scaling. Furthermore, Zhong et al. (2007) used NF (DK-4040F) made up of polyamide composite membrane for the secondary treatment of AMD from a copper mine in China. More than 90 % of metals were removed and the total conductivity was decreased by 48 %. However, water recovery was assumed to be between 13 and 15 %. Lopez et al. (2018) also used nanofiltration membranes for the recovery of metals and sulfuric acid from solutions representing AMD. Semi-aromatic polyamide (NF270) and sulfonated polyethersulfone (HydraCoRe 70pHT) membranes were compared at different trans-membrane pressures. More than 90 % and 60 % metal rejections were observed for NF270 and HydraCoRe 70pHT, respectively. Moreover, NF270 has a higher capacity for concentrating sulfate ions and metals than HydraCoRe 70pHT. The ability of NF270 and TriSep TS 80 polyamide thin film composite NF membranes to retain cations when filtering AMD from a copper mine in Western Australia was evaluated by Mullett et al. (2014). The isoelectric point (IEP) and feed pH tests were considered. The results indicated that the retention of cations was high when the feed solution pH was less than the IEP of the membranes. Overall, TS 80 performed better than NF270 since the feed pH of the cations and sulfur did not significantly affect the rejection performance which was >90 %, >95 % and >78 % for metals, sulfate and chloride, respectively at 70 % water recovery.

5.3.2. Application of reverse osmosis technologies

There are a number of reverse osmosis technologies which been applied in large scale. Most of the technologies presented below have patented and commercialised.

5.3.2.1. HIPRO®. High Recovery Precipitating Reverse osmosis (HiPRO®) is a multiple stage ultrafiltration technology that uses reverse osmosis (RO) membrane systems (Van Rooyen et al., 2021). It was implemented at the eMalahleni Water Reclamation Plant in South Africa. There are three stages and each is associated with a pre-treatment, clarifier, ultrafiltration and RO steps. In stage one pre-treatment step, iron and manganese are removed using ozonation so that RO can be possible. During RO, the aqueous phase becomes supersaturated with sulfates and antiscalant is added to keep sulfates in solution thus, preventing precipitation onto the membrane. In each stage, potable water is produced through the RO process and released whilst the remaining reject becomes the feed for the following stage. The pre-treatment steps in stage two and three use lime to increase the pH of the reject received so that salts (mainly gypsum and magnesium hydroxide) can precipitate (Karakatsanis and Cogho, 2010). Ultrafiltration is used to remove all the remaining solids so that only the liquid passes through to the RO step which is the final step of the process.

Gypsum and sludge are collected during the clarification steps and are disposed of after dewatering. However, some of the sludge and brine are disposed of without further treatment. More than 95 % of sulfate can be recovered using this process (Van Rooyen et al., 2021). Some of the great things about this process include the production of high quality drinking water and the removal of monovalent ions from water (Karakatsanis and Cogho, 2010).

5.3.2.2. SPARRO. Since lime is normally used for the neutralisation of AMD, the resulting solution is usually high in calcium. Thus, there is therefore a need for a technology that can deal with both AMD and high calcium concentration as it can desalinate solutions with calcium and sulfate content. One such technology is Slurry Precipitation and Recycle Reverse Osmosis (SPARRO®) process. In this process, there is a slurry suspension tank where all the precipitation products can form and retained so that membrane surfaces are protected (Pulles et al., 1992). It was designed to overcome some shortcomings of seeded systems including high energy consumption, high calcium sulfate recirculation rate and a poor calcium sulfate seed and brine mass balance control system (Juby et al., 2008). This technology uses seed crystals to precipitate calcium sulfate and other scaling compounds from the reverse osmosis concentrate. This ensures that these compounds do not precipitate on the RO membrane surfaces, but on the seeds (Juby et al., 2008). More than 90 % water recovery can be achieved, however high operating costs may result due to the chemicals used and large footprint (Semblante et al., 2018).

5.3.2.3. Forward osmosis-reverse osmosis osmotic dilution process. Choi et al. (2017) constructed a pilot plant for wastewater reclamation using RO process over five months. Two membrane process systems viz forward osmosis - reverse osmosis (FO-RO) dilution and conventional seawater reverse osmosis (SWRO) processes were used. Integrating FO-RO dilution reduced the RO fouling potential by FO treatment of wastewater. The wastewater was collected from Samcheok Coal-fired Power Plant, Korea and the seawater was collected from the East Sea in Korea. The FO system was made up of the polyamide FO membrane with four modules installed in parallel. Both systems had multiple-component configurations with SW30HR-380 RO membranes with three modules installed in series. The permeate water recovery rate in the FO-RO and SWRO systems was fixed at 55 % and 35 %, respectively. The total energy consumption was found to be 15 % less than that of typical seawater desalination by RO. The FO-RO was operated at 55 % recovery whilst rejecting 856,800 g of salts per day. The SWRO was operated at 35 % recovery with the rejection rate of 857,398 g per day (Choi et al., 2017).

5.3.2.4. MAXH2O desalter. IDE technologies which is a company that provides cheap and efficient solutions to treat high sulfate

concentrations in mine water impacted water has developed a technology called MAXH2O Desalter which uses RO (IDE Technologies, 2021; Wu et al., 2021a). It has a RO system which is integrated with a salt precipitation unit that removes salts such as calcium sulfate so that the required sulfate concentration can be obtained without compromising the system. During the process, there is a recirculation of the treated water at high shear velocity through the RO system as well as the recirculation of brine which reduces the build-up of salt concentration near the RO membrane wall. The technology also has a seeded fluidised bed reactor where the brine from RO is pumped to maintain a state of fluidisation. The seeds adsorb and precipitate the salts and the anti-scalant and produces salt-coated crystals which sink to the bottom of the bed. The lower part of the bed is discharged when the maximum limit is reached. The obtained crystals can be used as animal food additives, soil neutralisation and among other applications or disposed. The developed technology has been found to overcome scaling and biofouling. The technology was tested using mine water with high concentration of sulfates and the total sulfate recovery was >85 % with sulfate calcium sulfate saturation index of >1000 %. However, in practice, the saturation index is kept between 200 and 400 % by continuous precipitation. This technology overcomes the challenge of failing to meet the required sulfate concentrations by most current technologies whilst remaining economically viable.

5.3.2.5. RO concentrate treatment. Pervov et al. (2023) developed a technology that can reduce operational costs by reaching high recoveries in RO facilities. This can be achieved by developing a process that produces small amount of concentrate which can be evaporated to produce dry salty sediment that can be discarded in a special landfill site. This will avoid using other processes that deal with the RO produced concentrate e.g., zero liquid discharge. Thus, the technology focuses on the reduction and withdrawal of RO concentrate flow whilst producing a dewatered sludge. This is achieved by mixing the concentrate with wet sludge which is then dewatered and withdrawn. During the process, there is reduction of calcium as a result of calcium carbonate deposition on the seed crystals in the circulation mode. The technology further separates the concentrate into two streams containing monovalent and divalent ions from the feed.

5.4. Freeze crystallisation

AMD usually contains high concentrations of salts which are sometimes not considered during AMD treatment. Moreover, most of the AMD water treatment technologies including the recently developed, produce large amounts of inorganic brines and concentrates which are released into the environment (Randall and Nathoo, 2015). Brine, which is simply water impregnated with salts, has negative effects in the environment and thus, it has to be treated. Freeze crystallisation processes can be used to remove salt from water. This method can recover salt whilst producing pure quality water. During this process, water containing solutes (including contaminants) is cooled below its freezing point and forms ice crystals which floats whilst the solutes remain in solution (Randall and Nathoo, 2015). The solutes in solution can crystallize out but not always as the solution can be in the metastable state. However, there are Freeze Crystallisation techniques that can ensure that the solutes crystallize out. HybridICE™ is one of the freeze crystallisation technologies tested in pilot scale for the treatment of wastewaters (Mtombeni et al., 2013). It has been found to produce high-purity ice crystals without using fresh water to wash the crystals (Mtombeni et al., 2013). A static concentrator known as the HybridICE Filter Module (HIF) is used to separate suspended ice crystals from the concentrated brine slurry in order to recover the crystals as pure water (Adeniyil et al., 2013). The technology has been found to be cost effective due to its simplicity, low energy demand and the high possibility of producing cold energy from the process. The results from the pilot plant indicated that about 96 % salt removal was achieved (Mtombeni et al., 2013).

Eutectic Freeze Crystallisation is a type of Freeze crystallisation process where the supersaturated solution formed from cooling crystallizes out at eutectic temperature thus, the salt and water crystallize simultaneously (Fig. 6) (Lewis et al., 2010). The ice crystals are easily separated from the main solution since the solutes sink to the bottom of the crystalliser due to their high density. The obtained frozen water can be washed and melted to produce water that is nearly pure (Randall and Nathoo, 2015).

Hauensuo (2017) studied the application of eutectic freeze crystallisation on the treatment of mine wastewater. The study focused on bioleach mine in Sotkamo, Finland. The results of the study showed that the eutectic freeze crystallisation process was selective and the crystallised components showed high purity. Most of the sulfate in the water was removed by forming potassium sulfate. Lewis et al. (2008) found that Eutectic freeze crystallisation can be used for the treatment of brines and concentrates which are known to be challenging and somewhat impossible to treat with some of the well-known treatment techniques. The authors found that pure water and individual salts could be recovered in high quantities. Randall et al. (2011) used eutectic freeze crystallisation process for the treatment of aqueous solutions from a water reclamation plant in South Africa. The authors found that the brines can be reduced by 97 % and lead to the overall water recovery of >99 % with pure calcium and sodium sulfate salts production (>95 % purity).

5.5. Precipitation technologies

Precipitation technologies have been found to be cost-effective for treating mine wasters with high sulfate and metal content (Zahedi and Mirmohammadi, 2022). Most of the chemical precipitation treatment technologies use lime since it is cost-effective and efficient (Ndlovu, 2017). The technologies presented below have been patented and applied in pilot scale.

5.5.1. Alkali-barium-calcium (ABC) desalination process

The alkali-barium-calcium (ABC) desalination process is an integrated lime/limestone process which is widely used to treat AMD since it can neutralise and remove elements and sulfate from wastewater (Motaung et al., 2008). Due to the high concentration of sulfate in AMD, technologies that can particularly remove sulfate from wastewater are

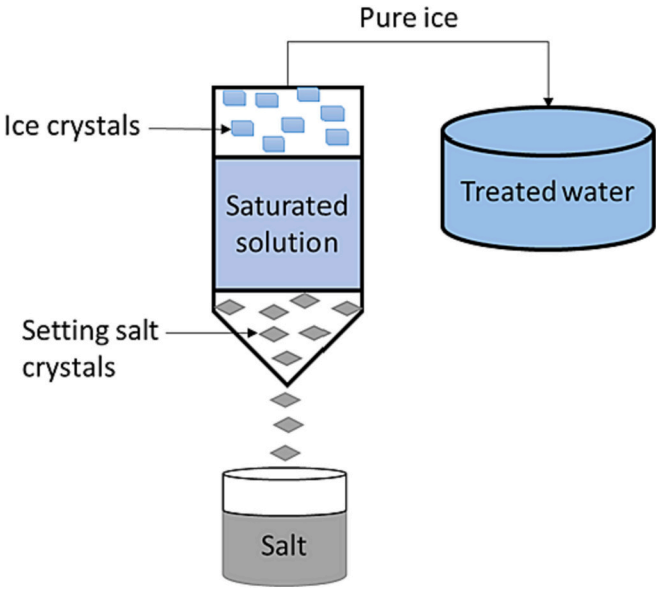


Fig. 6. Eutectic freeze crystallisation technique (Hubbe et al., 2018).

very crucial (Mulopo and Motaung, 2014). The ABC process is one of the few technologies that can remove sulfate with minimal waste. It uses three stages to treat AMD. In stage one, limestone is used to neutralise AMD where, some of the elements are removed. Lime is also added in stage one to partially remove sulfate as gypsum. In stage two, the residual sulfate is removed using barium carbonate ($BaCO_3$) and the resulting barium sulfate ($BaSO_4$) has low solubility (Eq. (3)). Moreover, $CaCO_3$ is precipitated using CO_2 stripping or $Ca(OH)_2$ dosing (Akinwekomi et al., 2017). Sludge processing occurs in stage 3 where, heat is used to convert the barium sulfate-rich sludge (from stage 2) to barium sulfide (BaS). $BaCO_3$ is regenerated by hydrolysing BaS with hot water and carbonation with CO_2.

$$Ca^{2+}(aq) + SO_4{}^{2-}(aq) + BaCO_3(s)\ BaSO_4(s) + CaCO_3(s) \quad (3)$$

This process results in good quality water that can be used for drinking. Minimal sludge waste is also produced. However, the process can be costly due to energy requirements and operational costs (Mulopo and Motaung, 2014). A pilot plant was constructed for the ABC desalination process at a gold mine shaft in Western Basin, South Africa (De Beer et al., 2010). The results indicated that the sulfate concentration was reduced from 4500 mg L^{-1} to 1250 mg L^{-1} through gypsum crystallisation at the pre-treatment stage. Finally, the $BaCO_3$ treatment reduced the sulfate concentration to 100 mg L^{-1} which is ideal for potable water. Some metals were removed through precipitation either as hydroxides or sulfides and the alkalinity of the water was reduced from 1000 mg L^{-1} to 110 mg L^{-1}. Mulopo and Motaung, 2014 also investigated the ABC desalination process at a pilot plant in Harmony gold mine shaft in the Western Basin, west of Johannesburg, South Africa. The results are indicated in Table 3. More than 90 % of sulfate was removed by the process including other elements as indicated in Table 3. However, some elements including Cl, K and Na were not efficiently treated by the process.

5.5.2. High density sludge

In search for affordable ways of treating AMD, the use of high density sludge (HDS) was discovered. It is used to neutralise mine wastewater and to precipitate elements. Sludge containing some of the settled solids from the active lime treatment process of AMD is recycled and taken back to the process so that it can be mixed with fresh lime before it is contacted with AMD (Mackie and Walsh, 2015). However, not all of the produced sludge is used, some of it is disposed of as waste. When the mine effluent is mixed with lime/limestone and recycled sludge, neutralisation is reached. The resulting product is then added into a lime reactor with aggressive aeration and high shear agitation. From the lime reactor, the discharge goes into a flocculation tank which leads to precipitation which is finally sent into a clarifier where the treated effluent will be separated from the sludge. The HDS process takes place between pH 9 and 9.5 since most elements precipitate at this pH (Murdock et al., 1994).

During the HDS process, elements in their hydroxide forms and gypsum are precipitated. If the wastewater contains high amount of sulfate, gypsum that exceeds its solubility will be produced hence, precipitate with the sludge (Murdock et al., 1994).

Dinu et al. (2014) and Stefanescu et al. (2015) investigated the performance of HDS pilot plant installed at Ilba-Alunis Asecare mine-shaft in Romania for the treatment of AMD. The HDS pilot plant had three reaction steps including automated pH control, flocculation and sludge recycling. The results of the study indicated that the HDS process was suitable for sludge densification even though the sludge settling rate was low. Moreover, the resulting effluent had fine precipitate solids due to high concentration of metals. Thus, a further filtering step was required. Lourenco and Curtis (2021) investigated the influence of high density sludge AMD chemical treatment plant on the surrounding water along the Blesbokspruit Wetland, South Africa. The Eastern Basin chemical AMD treatment plant was installed in Springs, South Africa for the treatment of AMD using HDS. The main purpose of the installation was to mitigate the decanting of AMD solution from an abundant mine into a nearby wetland. The HDS treatment process was found to increase the downstream concentration of sulfate, chloride, magnesium and sodium. Only iron, ammonia and phosphate were reduced. Thus, a secondary treatment is necessary.

Table 3
Chemical composition of feed and ABC process treated AMD from Harmony gold mine shaft in South Africa (Mulopo and Motaung, 2014).

Parameter	Feed (mg L^{-1})	Treated (mg L^{-1})
Al^{3+}	6	0
Ca^{2+}	559	30
Fe^{2+}	1100	0
Fe^{3+}	200	0
Mn^{2+}	174	1
Ni^{2+}	18	0
SO_4^{2-}	4510	250

5.5.3. SAVMIN

SAVMIN® is a technology developed by Mintek, South Africa. It has been created to treat highly acidic mine-impacted water. The main target of this technology is to remove metals from water and to reduce the high sulfate content (to <200 mg L^{-1}) that may be present in AMD (Dlamini et al., 2019). It has four stages and is capable of treating 20 ML of wastewater per day. In the first stage, lime is added to raise the pH so that gypsum and hydroxides of the elements can precipitate out and be separated from the process as waste. In the second stage, lime is added to the remaining solution in stage one so as to further raise the pH. Moreover, aluminium hydroxide is added so that it can mix with the gypsum saturated water. This then produces calcium aluminium sulfate salt known as ettringite which is separated from the solution. In stage three, the solution is treated with carbon dioxide to lower the pH which leads to the formation of calcium carbonate which is collected as a by-product. The remaining solution in stage three is the final treated water. In stage four, sulfuric acid and make up aluminium sulfate are added to the ettringite slurry from stage two where the pH is decreased and aluminium hydroxide as well as gypsum precipitate. Aluminium hydroxide is then separated from gypsum so that it can be recycled to ettringite formation.

This technology operates in ambient temperature and pressure thus, the energy consumption is very low. The drawback of SAVMIN® is that large amount of sludge is produced and it includes hydroxides of the elements and gypsum, which can be costly to manage (Ndlovu, 2017). The waste is discarded in lined hazardous waste disposal site. Also, the calcium carbonate produced can be re-used in the process whilst some is discarded as waste. The treated water from this process is potable and suitable for use in agriculture, domestic and industrial consumption.

5.5.4. Cost effective sulfate removal (CESR) process

The Cost Effective Sulfate Removal (CESR) process was developed in Europe in the 1990s for the treatment of industrial wastewaters associated with mining and mineral processing (Reinsel, 1999). Thus, it has been widely used for the treatment of AMD. In the CESR process, a proprietary powdered reagent is used to reduce the sulfate concentration to <100 mg L^{-1}. The addition of lime prior to the treatment process is so that some of the metals and sulfate could be removed through precipitation. Hydrated lime is used to reduce the concentration of sulfate when it is >8000 mg L^{-1}. However, if the concentration is <8000 mg L^{-1}, hydrated lime is used to increase the pH to 10.5 in order to remove metals as hydroxides and to precipitate gypsum. The powdered reagent is added to the wastewater which has been treated with lime to produce an insoluble alumina-sulfate compound (ettringite). During this process, a polishing effect results, which allows for the precipitation of metals including those which are challenging to remove from wastewaters. Moreover, ions including boron, fluoride, nitrate and chloride are also removed by the ettringite. All the constituents removed

cannot be leached hence the waste produced is not hazardous. The process is also environmentally friendly since the chemicals used can be precipitated. The resulting treated water meets the drinking water standards for metals, sulfate and constituents. Because of the efficiency of the CESR process, >20 treatment plants in Europe have been utilising the process at 350 gpm flow rate. The process was also used in Montana (USA) (Grey et al., 2018). Sample water from Berkeley Pit acid mine site with an initial sulfate concentration of 8700 mg L^{-1}, was used. The resulting effluent had a sulfate concentration of 56 mg L^{-1} and the treatment cost was determined to be $0.79 per cubic meters of wastewater.

5.5.5. Ettringite precipitation 1

Zahedi and Mirmohammadi (2022) developed an ettringite process to remove sulfate and to recovere $Al(OH)_3$ from an industrial wastewater. The process has five steps and the first step involves mixing the wastewater with $Ca(OH)_2$ to raise the pH to 12–12.5. The resulting slurry is stirred and allowed to settle so that the supernatant can be separated. During this step, magnesium is precipitated as $Mg(OH)_2$. In step 2, $Al(OH)_3$ is added in order to increase the concentration of Al which allows for the ettringite precipitation of sulfate ions at pH 12–12.5. The ettringite precipitate settled and the sulfate free supernatant was separated. The ettringite precipitate was mixed with sulfuric acid in step 3, to lower the pH to 4–5 in order to recover $Al(OH)_3$ which was reused in the ettringite precipitation step after several rinsing cycles. After settling, the supernatant was separated and used as the gypsum crystallisation step feed. Step 4 is known as the crystallisation step where the sulfates in the rinsing water are precipitated as gypsum at ambient temperature. The supernatant was separated after gypsum settlement. In step 5, the supernatant from the ettringite precipitate with no sulfate concentration was carbonised by blowing CO_2 using a porous bubbler and calcium in the form of $CaCO_3$ was precipitated. This resulted in sulfate-free treated water and the metals (Mg and Ca) were reduced by >75 %. The concentration of Al in the treated water met the drinking water standard.

5.5.6. Ettringite precipitation 2

Nevatalo et al. (2014) patented a precipitation process of treating AMD. The process begins by mixing AMD with a calcium compound to produce a sludge containing gypsum. The sludge is then mixed with another calcium compound and aluminium compound to produce a sludge with ettringite and gypsum. In both stages of sludge productions, the gypsum is removed if the amount of gypsum products is >10 % by weight otherwise, the process proceeds to the separation step where the ettringite and gypsum are separated from the liquid, thus, producing the first solution which is mixed with a carbonating agent in order to neutralise the solution. During this step, the calcium precipitates as $Ca(OH)_2$ and separated. The separation leads to solution with reduced sulfate, calcium and other metals. More than 90 % of sulfate and metals can be removed when the appropriate aluminium chemical is used.

5.6. Ion exchange

Different ion exchange resins have been used to remove metals and the removal can be done directly without any treatment to the AMD since most of them are present mainly as cations, hence cationic resins have been mostly used (Felipe et al., 2021). Ion exchange has been found to be an environmentally friendly technology. Companies such as DuPont, Dow Chemical Co. and Mintek have developed ion exchange resins to target specific elements (Steck and Yeager, 1980; Green et al., 2002; Williams et al., 2015). Moreover, these resins are now commercially available and have been used in large scale.

Most commercialised techniques for the recovery of metals from mine influenced water (MIW) are effective when the concentrations are higher (i.e., >1 g L^{-1}) (Moreira et al., 2021). However, there is a need for techniques that can recover important metals in low concentrations. Ion exchange is a hydrometallurgical technique that has been found to be effective on the recovery of metals at very low concentrations (ppm levels) (Sole et al., 2018). Ion exchange is a reaction where, there is reversible interchange between ions in solution (aqueous phase) which may be cationic or anionic and ions in the exchange resins phase) which may be cationic or anionic (e.g. Eq. (4)) (Choi et al., 2020). Ion exchange resins are characterised by presence of unique functional groups that imparts specific binding characteristics. In order, for the resin to be considered as effective, it must be selective for the analyte of interest which is in the presence of other competing species (Dąbrowski et al., 2004). This can be achieved by incorporating a specific functional group that has high affinity for the analyte of interest whilst exhibiting low affinity for the other species.

$$R^{-}A^{+} + M^{+}\ R^{-} - M^{+} + A^{+} \quad (4)$$

where the cation A^+ in the functional group of cationic resin exchange with the cation M^+ in the solution.

Ion exchange can potentially treat poorly clarified liquors or even pulps, thus eliminating costly solid/liquid separations (Berrios et al., 2013). The main advantages of the exchange technique are its high selectivity, high adsorption capacity, the lack of sludge production and the recovery of valuable metals (Dlamini et al., 2019). Ion exchange resins are made of a polymeric structure and tailored for selectivity of various metals with various acidic or chelating functional groups (Felipe et al., 2021). Chelating resins which normally have higher selectivity are highly preferred over acid resins due to the stabilities of metal complex at appropriate pH. Chelating resins have functional groups which act as donor atoms (Lewis base) for cations (Silva et al., 2018). These functional groups are required for metal complexation coordinate bond interaction or electrostatic interactions (Silva et al., 2018). The most common functional groups of the chelating resins include oxygen, nitrogen, sulfur and phosphorus (Sengupta and SenGupta, 2002). This is because base metals i.e., transition metals surround themselves with electron donating molecules or ions. These molecules or ions donate their electron pairs to the metal to form coordination bonds.

Batch and column processes have been used in an industrial scale (Shaidan et al., 2012). During a batch process, the resins and the electrolyte solution are mixed in a tank for a particular amount of time which is indicated by equilibrium (Wheaton and Anderson, 1958). After the reaction time, the resins are separated from the solution using filtration. However, due to the limitation of the resin capacity in batch mode, column is normally used in the industrial scale since its only limitation is the selectivity of the resin. For higher efficiency, a multi-column can be used where, the output solution of one column is the input solution of the next column and so on (Botelho Junior et al., 2019). There are three modes in which the column can be operated and these are; up flow, down flow and counter flow.

The advantages of chelating resins applications which include high capacity for the metal species of interest, fast reaction kinetics, efficient elution, and service life durability have made them suitable for the recovery of metals from mine impacted water (Van Deventer, 2011; Ulloa et al., 2020).

Nascimento et al. (2004) used a commercialised macro-porous strong base ion exchange resin, Amberlite Ira 93 for the recovery of uranium from AMD at Caldas Uranium Mining and Extraction plant, Brazil. The resin was first converted to the chloride form using HCl. The best recovery results were obtained when the AMD was pre-treated with lime for the removal of iron. Over 94 % of uranium was removed using ion exchange and NaCl was used for uranium elution from the resin. More than 98 % of commercial grade uranium was eluted.

Gypsum-continuous ion exchange (GYP-CIX) is a continuous fluidised bed ion exchange process that has been designed to remove calcium and sulfate from wastewaters (Robertson and Rohrs, 1995). Fluid seed beds are used in the loading sections so that unfiltered wastewater can be used. A pilot plant was designed for this process and is

represented in Fig. 7 (Robertson et al., 1994). In the first stage of the process, the feed water is introduced into the cation loading section to remove cations. The resulting solution is pumped into the anion loading section for the removal of anions and finally, product water at neutral pH results. The process leads to high quality calcium and sulfate precipitates which can be reused. The cation and anion resins can be regenerated by lime and sulfuric acid, respectively. However, this generates large amount of sludge which requires a disposal approach which is often costly. The pilot plant was operated for one year without interruption and the results showed that >80 % of sulfate was removed. Other anions (PO_4^{3-}, NO_3^-, F^- and Cl^-) and cations (Ca^{2+}, Mg^{2+}, Na^+ and K^+) were also significantly removed including radioactive elements such as U and Ra. About 90 % of water recovery was achieved by the plant and it was determined that a low resin requirement is required (i.e., 5–10 % per year) (Robertson and Rohrs, 1995).

6. Combinations of AMD treatment technologies

Wastewater resulting from mines, whether closed, abandoned or operating should be carefully treated to avoid serious environmental, economic, societal and health consequences (Simate and Ndlovu, 2014). Since the AMD wastewater can be of different qualities, methods of treatment should be carefully selected. Moreover, because there is no "one-size-fits-all" solution to water treatment that suits every need for the mining industry, some of the technologies can be combined to sufficiently treat AMD (Taylor et al., 2005). Also, the resulting treated water from one technology can be the influent to the other technology so that it can achieve what the previous technology could not. Thus, suitable AMD treatment technologies can be combined to achieve what they could not when used in isolation. This phenomenon has gained attention in recent years and has been found to be more effective since the acid, water and elements i.e. metals can be recovered in one system involving the different technologies (Simate and Ndlovu, 2014). The combined technologies are applied in a step-wise fashion where the treated AMD from one technology becomes an influent for the next technology in line so that high treatment efficiency is obtained (Simate and Ndlovu, 2014). The following scenarios focus on AMD treatment technologies that can be combined based on the quality of AMD to be treated and the final desired products (Taylor et al., 2005).

6.1. Scenario 1

A technology like SAVMIN® can be used to treat AMD with high sulfate content. Beyond removing sulfate from AMD, it can also recover >95 % of water, remove metals through neutralisation with lime and produce a non-saline effluent stream (Naidoo et al., 2018). The limitation of the SAVMIN® technology is that it does not address the monovalent ions and not all of the metals are removed, which is why another technology such as the ion exchange can be combined with the SAVMIN process (Fig. 8) (Naidoo et al., 2018). The combination of these technologies will also be important for AMD with high concentrations of REEs and concentrations of other valuable metals such as Mn, Ni, Co and Zn which can be recovered using suitable ion exchange resins (Puyol et al., 2017). The combination of these technologies is based on a study which was conducted in South Africa, where acid mine water was oxidised with H_2O_2 and precipitated using lime which was followed by the application of ion exchange to reduce the concentration of cations (Feng et al., 2000). The addition of lime increased pH and significantly reduced the concentrations of sulfate by >70 % due to the precipitation of an insoluble salt i.e., ettringite ($3CaO{\cdot}3CaSO_4{\cdot}Al_2O_3{\cdot}31H_2O$). However, the remain sulfate concentration was still high (>1000 mg L^{-1}). Thus, SAVMIN which is more efficient can be used instead of the oxidation and precipitation processes, to ensure that the sulfate concentration in the mine water is very low. The remaining metals from the SAVMIN process will be removed by ion exchange. The resulting effluent from this combination can be used for irrigation. The metals in AMD can

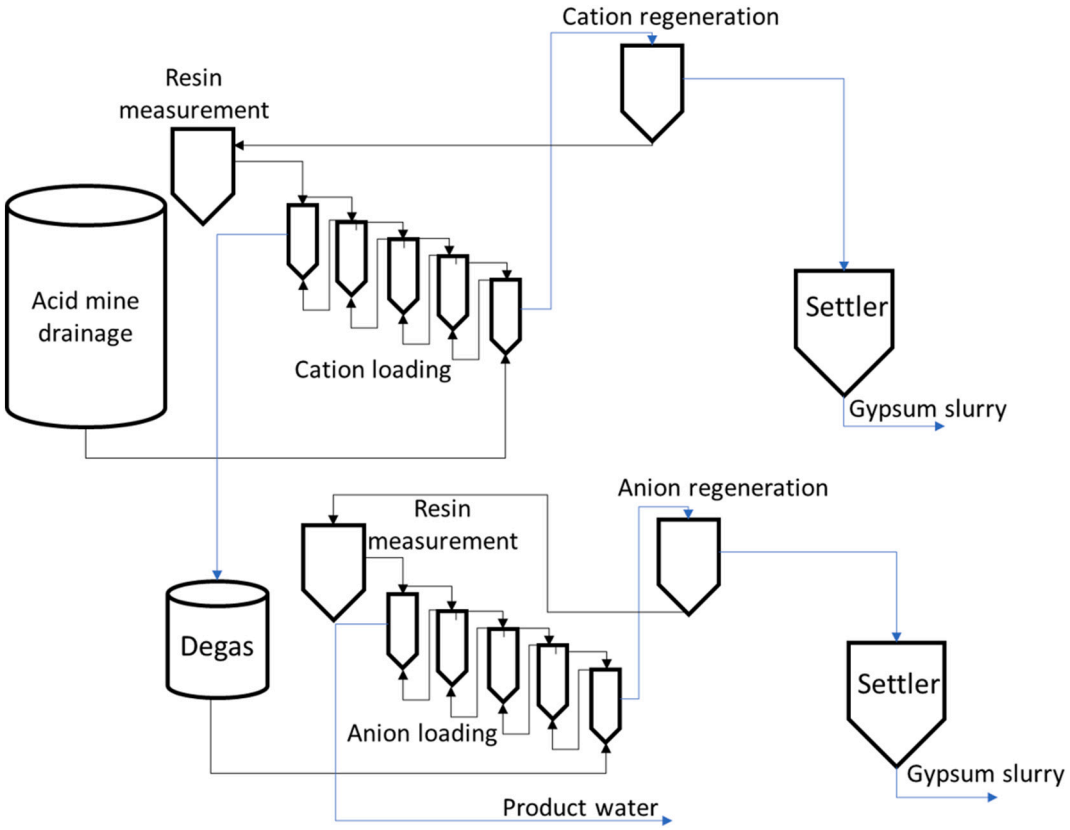


Fig. 7. Gypsum-continuous ion exchange process (Robertson et al., 1994; Robertson and Rohrs, 1995)

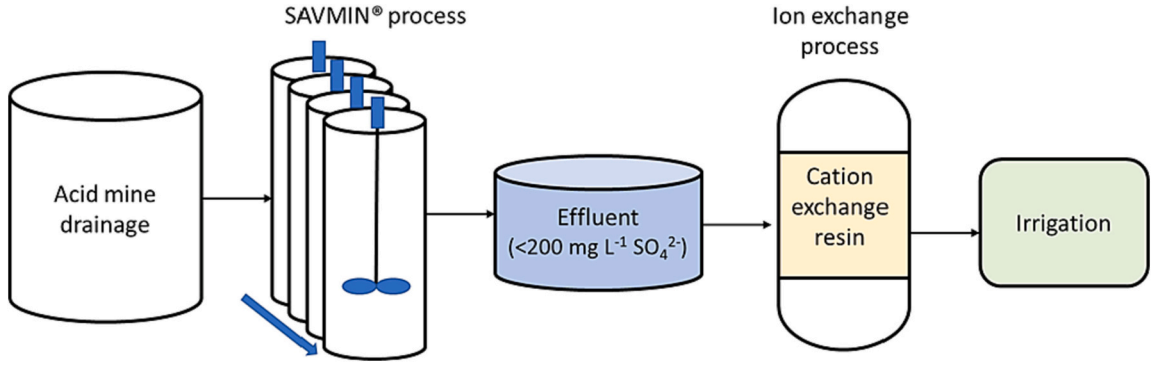


Fig. 8. Combination of the ion exchange and SAVMIN® processes for the treatment of AMD.

also be removed using extraction however, the whole process including installation and apparatus is too expensive, hence ion exchange is a suitable alternative (Dąbrowski et al., 2004).

6.2. Scenario 2

The final produced water from Scenario 1 can be treated further using ultrafiltration i.e., NICmembrane™, in order to potable water (of drinking quality) (Fig. 9) (Petterson, 2018). The application of the membrane after other treatment technologies will prevent the usual membrane challenges when applied to AMD. These challenges include abrasion and breakage of the membrane material due to solid particles as well as membrane fouling (Agboola, 2019). The combination of these technologies can be used in regions where very large amount of water is used during mineral processing, resulting in large amount of AMD. This can also be used in regions where there is high volume of AMD from closed and abandoned mines especially in arid and semi-arid regions faced with water scarcity. Consequently, some of the SAVMIN® treated water can be used for irrigation whilst some goes through the final process of ultrafiltration where pure water will be produced. The main challenge with this combination is that the quantity of treated water from SAVMIN might be high, which means that the feed flow to the ion exchnage and the memebrane processes will require control, so that suitable flow rate is used in order to get the best results of the processes (Nasir et al., 2016; Dagde, 2018).

6.3. Scenario 3

In the case where AMD has high concentration of sulfates and no valuable elements, the SAVMIN® technology can be used followed by the NICmembrane™ process so that potable water can be produced (Steenhoff-Snethlage, 2018). However, if potable water is not a necessity, the SAVMIN® technology on its own will be sufficient to produce water that can be used for irrigation (Dlamini et al., 2019). The main issue with the first three scenarios is that there is high residual waste produced (e.g., gypsum, Mg, Fe etc.), which must be disposed (Dlamini et al., 2019). To prevent the negative environmental impact of the combined technologies, some of the waste can be separated and considered as resource potential e.g., gypsum can be used as a soil conditioner or cement (Prasad, 2016).

6.4. Scenario 4

Alternative to using the SAVMIN® process, technologies that use sulfate reducing bacteria (i.e., CloSURE or BioSURE) can be used to treat AMD or mine water with high sulfate concentration and metal content (Fig. 9) (Foucher et al., 2001). These technologies are sustainable and cheaper than conventional technologies, and will therefore be highly suitable for closed and abandoned mines in struggling and developing countries (Foucher et al., 2001). The SRB technologies have also been found to sufficiently remove >90 % of sulfates from mine drainages with over 2000 mg L^{-1} of sulfate concentration (Rose, 2013). When combined with other technologies, CloSURE or BioSURE can begin the treatment process as they will ensure that most of the sulfates and some of the elements are removed from the solution (Luptáková et al., 2016). A study by Poinapen (2012) used the BioSURE process to remove sulfate from AMD and found that the 1800 mg L^{-1} initial sulfate concentration in AMD was reduced to 143 mg L^{-1} after the process. Maree et al. (2000) also used the BioSURE process treat AMD and the results indicated that the effluent had 123 mg L^{-1} of sulfate which was initially 1672 mg L^{-1}. du Preez (2021) indicated that the CloSURE™ process small scale plant was set-up in Mintek premises in South Africa to treat AMD using raw mine water which was collected from a coal mine site in Mpumalanga Province, South Africa. The average volumetric sulfate reduction rate of 2.05 mol m^{-3} d^{-1} (87 %) was achieved within 90 days. The results from these studies also showed that some metals were removed, however, some including magnesium, calcium and sodium were still in high concentrations. Overall, the effluents from these studies were suitable

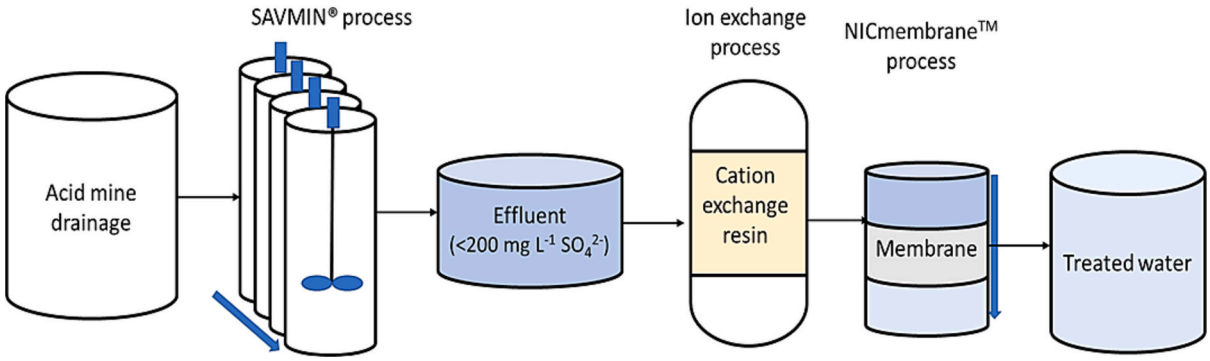


Fig. 9. Combination of the ion exchange, SAVMIN® and NICmembrane processes for the treatment of AMD.

for irrigation activities or release into the environment (Van Zyl et al., 2001; du Preez, 2021). To obtain potable water, the effluents from the SRB technologies can be further treated with other technologies e.g., NICmembrane™ (Fig. 10). If there is a need to recover valuable metals from the AMD, the ion exchange process can be added in the combination.

6.5. Scenario 5

Some wastewaters have very high concentration of both metals and salts, some of which are very toxic and would require serious attention (Utgikar et al., 2002). The combination of reverse osmosis technology and freeze crystallisation will be very imperative in such a situation. Fig. 10 shows the combination of the HIPRO® process and eutectic freeze crystallisation technologies. The HIPRO® technology treating >20 ML day^{-1} of AMD has been applied on a commercial scale to treat AMD from coal mines in South Africa to potable standards (Steyn et al., 2021; Broadhurst et al., 2022). Anglo coal together with its partner BHP Billiton developed a plant for the HIPRO® process which has been successful in removing >99 % of salts and metals in order to achieve 99.7 % water (potable) recovery rate (Gunter and Naidu, 2008). The water is delivered to a local eMalahleni municipality in Mpumalanga province, South Africa, in order to prevent the dewatering of underground workings so that mining can continue whilst protecting the environment (Gunter and Naidu, 2008). As shown in Fig. 11, the HIPRO® process involves the ultrafiltration and reverse osmosis processes to ensure that potable water is produced (Naidoo et al., 2018). However, there is large amount of sodium sulfate brine which results from the reverse osmosis process, and would require further treatment or special disposal at waste disposal facilities, which can be very expensive (Mulopo, 2015). The high salinity of the brine makes it unsuitable for use in irrigation, hence the importance of freeze crystallisation, which will ensure that the salts are separated from the solution (Lewis et al., 2010; Naidoo et al., 2018). Freeze crystallisation processes have been found to have the potential of reaching a near zero waste by producing salts including pure salts and potable water from high salinity brines (Randall and Nathoo, 2015). Freeze crystallisation technologies have been used to treat brines from reverse osmosis processes. Eutectic freeze crystallisation has been used in complex hypersaline brines that are typical of reverse osmosis retentates in South Africa, and was found to be efficient and successful, which is why the combination is vital in highly concentrated AMD (Lewis et al., 2010). A paper by Randall et al. (2011) described a case study where eutectic freeze crystallisation was used to treat brine from the HIPRO® process in South Africa. About 97 % of the brine was recovered as pure water and salts such as calcium sulfate and sodium sulfate were obtained in their pure forms (>96 %). The study concluded that the overall, >99 % of the brine was converted to viable products. Moreover, the HIPRO® process also produce solid waste, most of which is gypsum. The gypsum processes developed by the Council for Scientific and Industrial Research (CSIR) in South Africa were added to the combination of the technologies so that they can convert the gypsum waste into valuable products such as calcium carbonate and magnesium carbonate in order to offset the high costs associated with gypsum disposal. This will ensure that the waste produced is minimal which lessens the effect on the environment (Mulopo, 2015). The disadvantage of the combination of the HIPRO® process with freeze crystallisation technologies is the capital cost resulting from the freeze crystallisation processes, however, these can be overcome as the technology develops (Rane and Padiya, 2011). Also, the reverse osmosis process from the HIPRO® technology will require constant monitoring and long-term treatment (Chen et al., 2021).

6.6. Scenario 6

In case the AMD is highly concentrated with sulfates, an additional technology can be added in the above description (scenario 5) to significantly reduce the concentration of sulfates. The incorporation of SAVMIN® or other technologies that are designed to treat mine waters with high sulfate concentration can be used (Fig. 12) (Zahedi and Mirmohammadi, 2022). The sulfate removing technology will have to be used first so that the HIPRO® process perform very well to produce potable water (Steyn et al., 2021; Broadhurst et al., 2022). If the SAVMIN® process is used, the pre-treatment and the addition of lime steps in the HIPRO® process will not be necessary. Thus, leading to a cheaper HIPRO® process. Besides using the HIPRO® process which may be expensive, the conventional reverse osmosis process can be used instead

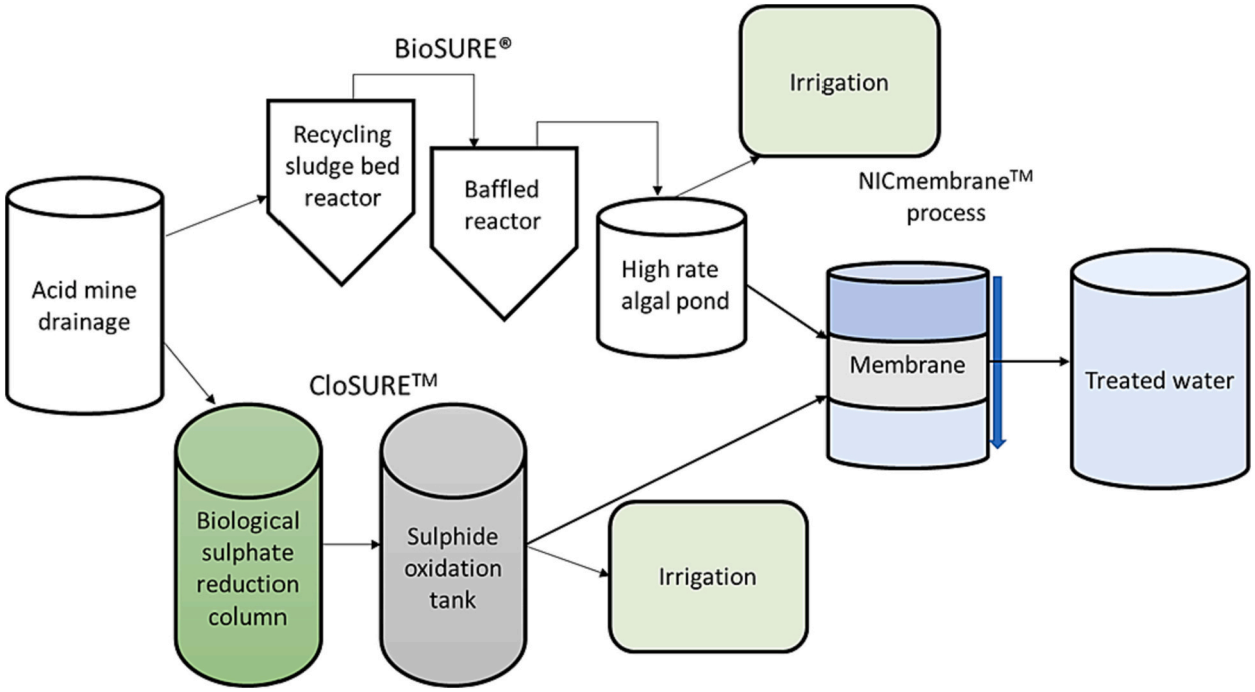


Fig. 10. Combination of the sulfate reducing bacteria technologies and the NICmembrane™ process for the treatment of AMD.

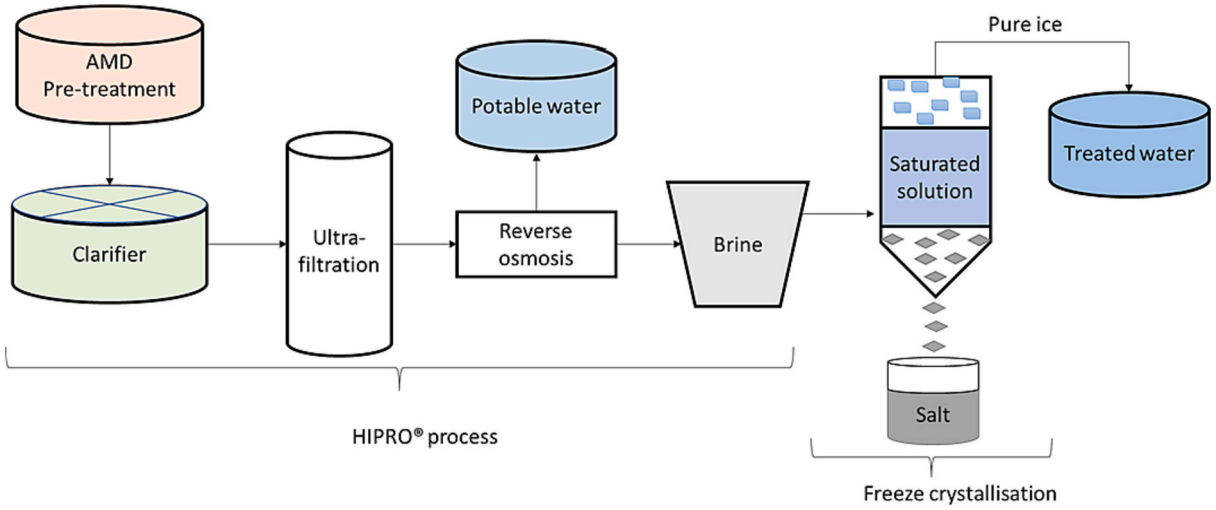


Fig. 11. Combination of the HIPRO® and freeze crystallisation processes for the treatment of AMD.

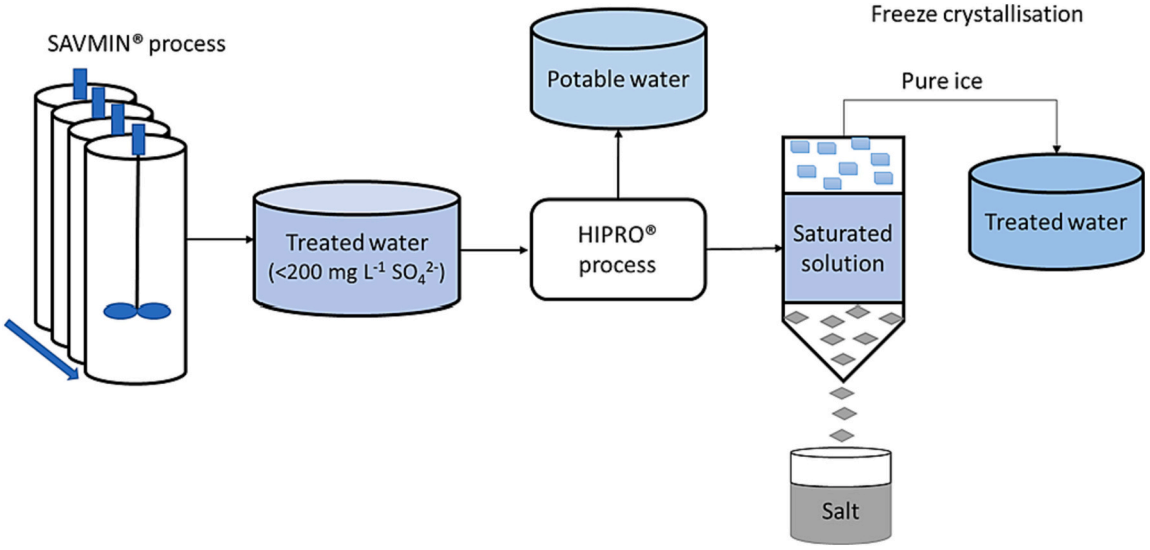


Fig. 12. Combination of the SAVMIN®, HIPRO® and freeze crystallisation processes for the treatment of AMD.

since the water coming from the sulfate removing technology is in a state that is suitable for reverse osmosis (Anis et al., 2019). The freeze crystallisation technology would still be important, since brine will be produced from the reverse osmosis process (Mulopo, 2015). Ion exchange method can be added as part of the combined technologies, if there is a need to recover valuable elements (e.g., combination 1, Fig. 12) (Simate and Ndlovu, 2014; Masindi et al., 2022). The potable water resulting from these processes can be transferred to nearby municipalities so that it can be used for drinking and domestic purposes (Gunter and Naidu, 2008).

6.7. Other possible combinations

Other possible combinations are shown in Fig. 12. The selectivity of the technologies will be based on the composition of the AMD and the targeted end use of the treated water. As indicated before, solutions with high sulfate content will require sulfate removal focused technologies such as SAVMIN® and sulfate reducing bacteria processes (CloSURE and BioSURE). These processes can be combined with conventional reverse osmosis processes to produce potable water as indicated in Combination 1 (Fig. 13) (Van Houten et al., 1994; Subramani and Jacangelo, 2014). In case of high quantity of brine, the freeze crystallisation technologies can be added to further produce potable water and pure salts (Randall et al., 2011). All those processes leading to brine production can be combined with freeze crystallisation so that minimal waste is produced. Moreover, some of the waste produced can be re-introduced into a running treatment system to be treated further and so as to reduce waste production. Though these technologies are from different places, they can still be combined to treat different qualities of AMD with different composition and quantity. Some of the technologies do not have to be incorporated in their entirety. For example, some of the steps in the HIPRO® technology can be eliminated by incorporating the membrane from the NICmembrane™ technology, thus, creating a cheaper and a more efficient process. A study by Jin et al. (2020) combined nanofiltration and ettringite precipitation processes to treat sulfate-rich brine (Combination 5). The study showed that starting with nanofiltration would results in sulfate-concentrated effluent which can be treated using the ettringite precipitation process to produce water that can be reused. But, the study indicated that a highly concentrated brine leads to the final treated water with high sulfate concentration and pH, which are higher than the discharge limits. The authors then proposed that an additional step i.e., blending of the supernatant after precipitation can be used in order to

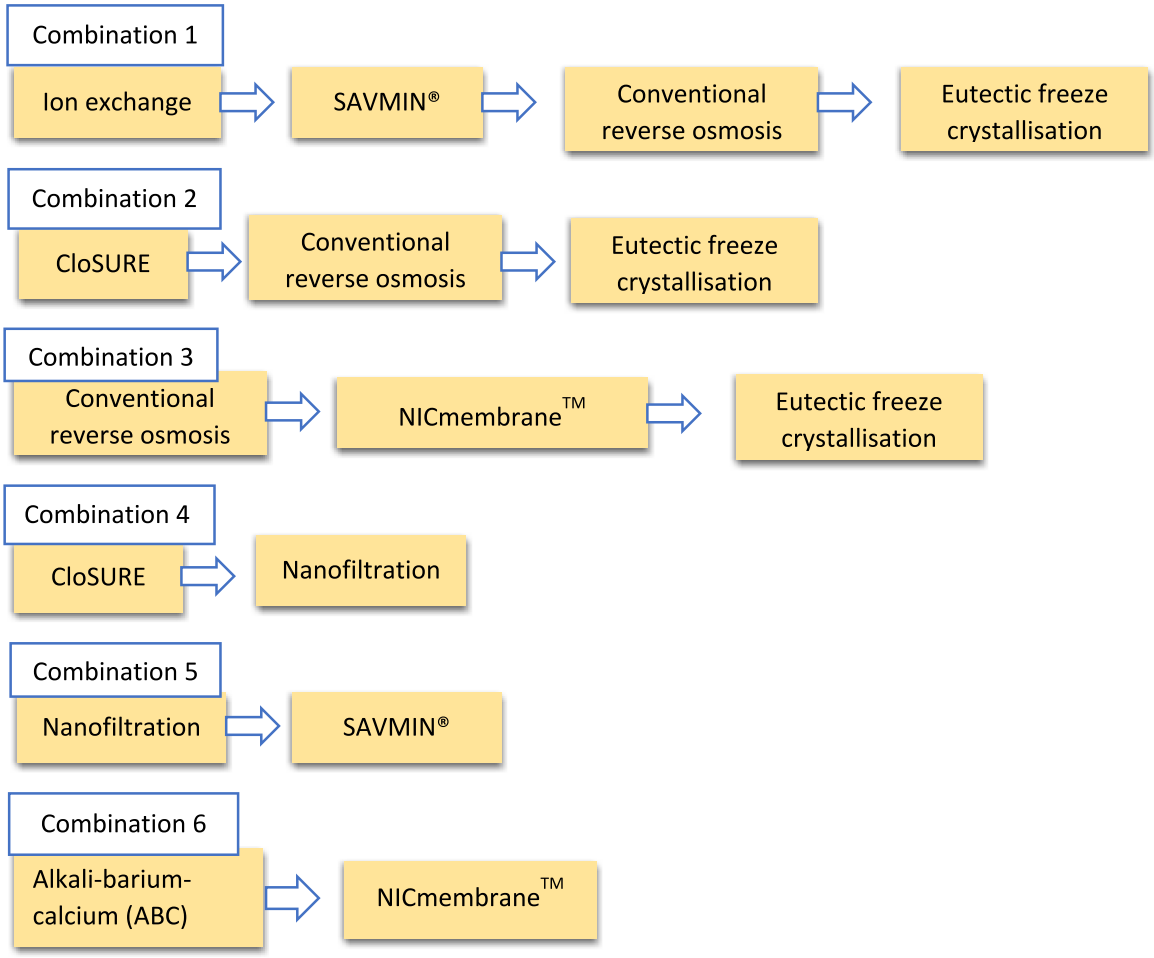


Fig. 13. Additional combinations of mine water treatment technologies.

satisfy the discharge limit for sulfate (Jin et al., 2020). Which is why, the use of nanofiltration with an efficient process such as SAVMIN will be beneficial as the produced water will be suitable for irrigation (Naidoo et al., 2018). The Alkali-barium-calcium (ABC) desalination process can remove acid, sulfate and metals from AMD to produce water whose quality can be further improved by adding another technology i.e., NICmembrane™ technology to produce drinkable water especially in water constrained countries (Combination 6) (Motaung et al., 2008; Steenhoff-Snethlage, 2018).

AMD has an overwhelming impact on the economics of mining operations and local governments and/or municipalities. The choice of the best combination can be based on a number of factors, including the quality and quantity of the AMD to be treated per day, the desired quality of effluent and other products, costs of the technologies, accessibility of materials to build process plants and necessary land surface area (Johnson and Hallberg, 2005; Taylor et al., 2005). The larger the quantity of AMD, the higher the treatment cost. South Africa, Australia, Canada and the Unites States are known as the four largest mining nations in the world and their combined AMD treatment cost is predicted to exceed USD 32–72 billion (Cozzolino et al., 2018). The treatment of AMD from closed and abandoned mine sites is estimated to be higher (Harries, 1997). The combinations of the technologies as indicated in this review, will produce effluents that can be used for irrigation or domestic purposes (Steyn et al., 2021; Broadhurst et al., 2022). Thus, reclaiming drinking water from the treatment of AMD can help reduce the consumption of water from natural resources, which are already limited and critical in other areas (Simate and Ndlovu, 2014; Al-Hwaiti et al., 2016). Other processes as indicated, can produce pure salts and other valuable minerals such as goethite and gypsum, which can be purchased to suitable industries in order to offset the treatment running costs (Masindi et al., 2018; Shingwenyana et al., 2021). Also, since considerable amounts of REEs have been detected in AMD, their recovery can also help offset the treatment costs (Ayora et al., 2016). Therefore, the combined AMD treatment technologies which can produce potable water and other products that can be sold for monetary benefits, are economically viable (Adiansyah et al., 2017). Some of the technologies require less human intervention, use less energy and cheap resources which is important for developing and struggling countries (Magowo et al., 2020).

7. Future prospects and recommendations

1. The above combinations have not been tested experimentally thus, they are selected and combined based on the quality of water that can result with each technology or process. However, calibrated computational modelling can be used to optimise the alignment of the technologies using a designed application/software. For this to be possible, the model application must be able to accurately simulate the quality of water that can be obtained with each technology considered, based on the type or quality of water in contact with the selected technology. Thus, the modelling should allow for the quality of the AMD (including pH, concentration of metals and non-metals etc.) to be specified. The application can be connected with a software called parameter estimation (PEST) which can be used to successfully calibrate the model (Doherty, 2004). If the actual quality of treated specified AMD by these technologies is known, it can be

added to the model and PEST will help with running the model many times until the results obtained are the same as those obtained during industrial application. When this is achieved, there will be confidence that the model is successfully calibrated and thus, other results will be accurately determined. Thus, the model must have an input file where the parameters of the water under study are specified so that the quality of water from each technology can be specified. Moreover, the designs can be tweaked based on the predicted water chemistry from each technology. This way, many different scenarios can be simulated, and the best possible outcomes can be configured. The modelling application can also have a cost centre, where the cost of each application is considered so that the overall cost of the combined technologies is known.

2. There is still a need to come up with cheap and efficient adsorbents that can be applied in large scale for the treatment of AMD. Re-usable adsorption systems that can be combined with other technologies or methods are still required. This should include the management of the exhausted adsorbents as well as the financial implications.
3. Researchers from different parts of the world can collaborate with the local governments and/or municipalities to construct small pilot plants using the different technologies as specified in this review to determine their applicability in large scale as well as the economic implications. Some of the technologies and methods specified can be improved to achieve better AMD treatment.
4. Since some of the technologies are mainly known in areas where they are developed, a computer program incorporating the AMD treatment technologies around the world can be developed and made available to the public so that institutions looking for AMD treatment technologies can have options. The application should include the quality of AMD that can be treated by each technology and the quality of resulting water. Moreover, the application should include names of the developers and contact details for easy access.
5. Some of the technologies included in this review are costly to maintain, there is therefore a need to come up with cheaper technologies that can treat AMD to the similar quality as these technologies.
6. Not all combinations were considered with the technologies included in this manuscript hence other suitable combinations can be analysed based on the quality of the effluent which can be a potential influent of the other technology.
1. Researchers can consider other areas which are not normally considered but are concentrated with closed, abandoned and operating mines, since research has indicated that local communities are negatively affected. The studies should include the assessment of mining impact on the environment and living organisms including humans. The health data of the people living in such areas should be compared with that of other communities in order to determine the health impact of AMD. The data can be used to urge the government and local municipalities to act hastily on the treatment of AMD to prevent future disasters.

8. Conclusion

There are different technologies that have been developed and applied in the treatment of AMD. The technologies are unique and can lead to different qualities of treated water which can be used for drinking, irrigation, industrial usage or release into the environment, depending on quality. However, most of the technologies focus on the treatment of one AMD product (e.g., removal of sulfate) and often lead to large amounts of secondary waste products which are expensive to manage and dispose. Studies have indicated that combining treatment technologies can lead to the treatment of more AMD products, improve effluent quality and reduce the waste footprint. Thus, combined technologies have the advantage of eliminating shortcomings of individual techniques whilst leveraging effective aspects. The main aim of AMD treatment has generally been to remediate the water and reduce environmental pollution which has adverse effects on the ecosystem. However, there has recently been a realisation to derive value from AMD, since valuable elements such as rare earth elements, zinc, lithium, copper, silver, nickel and cobalt have been detected in AMD. These varied or differentiated targeting of entities in polluted water has been shown to be achievable by using the treatment methods in tandem, with their configuration optimally conducted. In some cases, it has been possible to show that the configuration can be manipulated such that only a few of the steps of an individual method could be used in tandem with a few of another and so on, implying that it may not necessarily always be required that wholesome original methods be used in all instances. There is a need to create a form of application which can select the best combinations of technologies. All the AMD treatment technologies around the world can be added into the application with their front-end engineering design packages, including the cost of treatment. It has been possible to achieve some insights into the treatment methods and how best their alignment can be achieved by using computational simulations of the processes. Overall, this review showcased different AMD treatment technologies with varying readiness levels and the best possible combinations were addressed.

CRediT authorship contribution statement

Alseno Kagiso Mosai: Writing – review & editing, Writing – original draft, Visualization, Validation, Resources, Methodology, Investigation, Data curation, Conceptualization. **Gebhu Ndlovu:** Writing – review & editing, Supervision, Resources. **Hlanganani Tutu:** Writing – review & editing, Validation, Funding acquisition, Conceptualization.

Declaration of competing interest

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

Data availability

No data was used for the research described in the article.

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