Review

Review of Remediation Solutions for Acid Mine Drainage Using the Modified Hill Framework

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Abstract: This paper reviews the Acid Mine Drainage (AMD) remediation potential and operational costs of twelve existing AMD remediation methods against Class 0 and Class I AMD geochemical characteristics as defined in the Modified Hill Framework. Of the twelve remediation options reviewed in this study, eleven required additional process steps either for further treatment to achieve the discharge limits or for the safe management of hazardous waste by-products. Chemical desalination showed the greatest potential with high quality treated water and operational costs between USD 0.25 and USD 0.75 per cubic meter treated. The management of the toxic metal and sulphide by-products remains a key challenge that requires further research for sustainable mine water remediation. Further development of end-to-end methods suitable for Class 0 AMD with economical operational costs is recommended in order to effectively address the ongoing environmental challenges posed by AMD globally.

Keywords: Acid Mine Drainage (AMD); geochemical classification; AMD remediation technologies; AMD remediation costs; AMD remediation efficiency; mine-water treatment

1. Introduction

Acid Mine Drainage (AMD) is a toxic wastewater stream formed when oxygenated water comes into contact with exposed mine rock surfaces containing sulphide minerals [1–3]. The formation of AMD is most prominent in abandoned mines where water accumulates in mine shafts and pits allowing exposure to sulphide minerals [4]. The resulting AMD is typically characterised by a low pH with high concentrations of heavy metals and dissolved sulphate. When left untreated, AMD streams can cause severe environmental degradation, including the contamination of natural water bodies, destroying aquatic life and toxifying natural habitats [2,5,6]. Mine sites can continue to generate AMD centuries after commercial mining operations have ceased [6,7]. The implementation of suitable technologies for the long-term remediation of AMD is, therefore, a critical task for local governments and organisations managing AMD streams [8].

Developing a general solution for abandoned mine AMD remediation has proven a difficult task due to the vast variation in the geochemical characteristics of AMD in each site [9–11] and the seasonal variation of AMD within sites, coupled with the long term decant of AMD [12,13]. The geochemical variation is influenced by weather, availability of oxygen, temperature, type of ore deposits, degree of rock fragmentation, metal sulphide exposure and biological activity amongst other factors [12–15]. The use of AMD classification frameworks has been proposed as a means to categorise the contamination potential and geochemical composition of AMD streams, while also indicating the degree of treatment required for safe environmental discharge [16–18]. Furthermore, classifying AMD streams can enable alignment and decision making between key stakeholders such as governments, mining companies, regulatory bodies and effected communities. Thisani et al.

(2020) conducted a study on the geochemical distribution of seventy-two AMD mine water sites across five continents and used the results to improve the Hill (1968) classification framework. The resulting Modified Hill Framework is a simple yet informative AMD geochemical classification method. The Modified Hill Framework classifies AMD by the degree of toxicity and indicates the level of treatment required. Class 0 of the framework representing highly acidic and concentrated AMD, while Class IV indicates oxidised and neutralised AMD safe for environmental discharge. The Modified Hill Framework also includes the dissolved concentration level of zinc as an indicator species for cytotoxic metals in AMD. Table 1 illustrates the Modified Hill Framework.

Table 1. Modified Hill Framework for Acid Mine Drainage Classification.

Class	Class Description	Thresholds		
Class 0 **	Highly concentrated and acidic mine drainage **	pH = 0.5–3 ** Acidity = 5–45 g/L **	Total Fe = 1000–12,000 mg/L **	SO_4 = 10–60 g/L ** Al = 1000–18,000 mg/L **
Class I	Acid Mine Drainage	pH = 2.0–4.5 Acidity = 0 **–15 g/L	Fe^{2+} = 0 **–10,000 mg/L Fe^{3+} = 0 mg/L	SO_4 = 0 **–20 g/L Al = 0–2000 mg/L
Class II	Partially oxidised and/or neutralised	pH = 3.5–6.6 Acidity = 0–1 g/L	Fe^{2+} = 0–500 mg/L Fe^{3+} = 0–1.000 mg/L	SO_4 = 500–10,000 mg/L Al = 0–20 mg/L
Class III	Neutral and not oxidised	pH = 6.5–8.5 Acidity = 0 mg/L	Fe^{2+} = 0–500 mg/L Fe^{3+} = 0 mg/L	SO_4 = 500–10,000 mg/L Al = 0–2000 mg/L
Class IV	Oxidised and neutralised/alkaline	pH = 6.5–8.5 Acidity = 0 mg/L	Fe^{2+} = 0 mg/L Fe^{3+} = 0 mg/L	SO_4 = 500–10,000 mg/L Al = 0 mg/L
Cytotoxic metals indicator **	Low = Zinc ≤ 1 mg/L **	Mid = Zinc ≤ 25 mg/L **	High = Zinc > 25 mg/L **	

Asterisks (**) indicate the revisions made to the original Hill (1968) framework.

AMD remediation methods can be divided into two main categories, active treatment and passive treatment methods. Active treatment methods are characterised by process inputs such as energy, chemicals, labour and automated control, whereas passive remediation technologies require minimal process inputs and are mostly self-sustaining [6,19,20]. Active treatment methods make use of various physical, chemical and biological processes and may present advantages such as metal recovery, high quality remediated water, greater flexibility to accommodate loading variation and consistency in remediation quality [21–23]. The key challenges associated with active treatment include the high operational costs and the generation of hazardous sludge that requires additional management and safe disposal [6,24,25]. These disadvantages can lead to active methods being economically unfeasible for abandoned mines in remote regions [23,26]. Passive treatment methods are typically at source constructions comprising of a combination of naturally occurring geochemical, physical and biological processes [27,28]. Passive treatment typically makes use of low-cost alkaline reagents such as limestone. Passive solutions may also make use of organic matter from sources such as wetlands to neutralise the pH causing metal precipitation and for hosting microorganisms to bioremediate contaminants [19,29]. The key advantages of passive methods include the low operational cost and minimal external intervention required for operation [30,31]. Some of the challenges associated with passive methods include the inability to increase the pH above 8.0 for acidic AMD, which limits the effectiveness for the removal of highly soluble metals such as Mn and Mg, the deterioration of the system over prolonged operation due to the depletion of reactive materials and the large land area required for surface constructions such as wetlands [23,26,30].

Global experience has shown that there cannot be a single treatment method that can be used in every single AMD scenario due to the vast variation in chemical composition and, therefore, treatment techniques need to be selected based on the specific mine water quality encountered and site-specific constraints [6,32,33]. This review was undertaken to evaluate the remediation capabilities of existing AMD treatment methods against the AMD classes defined in the Modified Hill Framework. The review further presents the

operational cost for each remediation solution in US dollars (USD) based on the existing literature. The operational costs presented are based on the remediation of AMD and do not include the costs associated with the management of waste by-products. The review categorises each treatment method using the AMD class that it can treat to safe discharge limits and the estimated operational cost range for the method.

2. Evaluation of AMD Treatment Options against the Modified Hill Framework

2.1. Chemical Treatment

The most commonly applied method for the primary remediation of AMD is a chemical treatment method known as neutralisation that uses lime ($Ca(OH)_2$) or limestone ($CaCO_3$) to neutralise the pH, resulting in the precipitation of heavy metals as metal oxides/hydroxides [6,34]. Chemical neutralisation may also incorporate aeration to oxidise reduced metals such as Fe^{2+} to Fe^{3+}, leading to increased precipitation [23]. An increased pH and the availability of calcium from calcium containing neutralising reagents enables some removal of high SO_4 concentrations (>1500 mg/L) through the crystallisation of gypsum ($CaSO_4 \cdot 2H_2O$) [6,35]. The major benefits of chemical treatment methods include the process scalability and versatility that enables the handling of any acidity loading and heavy metal concentrations, while being easy to operate and maintain [23,26,36].

2.1.1. Conventional Neutralisation

Conventional neutralisation processes consist of reaction tanks with stirrers for alkaline chemical dosing and reaction with the AMD [23]. The reaction tank is typically followed by a sedimentation process where the suspended solids are allowed to settle forming a hazardous Low-Density Sludge (LDS) of between 2–5% solids [37,38]. The supernatant quality contains low concentrations of heavy metals and neutralised pH [35]. However, conventional neutralisation is not effective at removing SO_4 [39]. The management of the LDS, the limited effectiveness of AMD remediation resulting in additional processing requirements and the continuous dosing of chemicals are key challenges associated with conventional neutralisation methods [26,30,40]. Conventional neutralisation and other neutralisation methods present opportunities for selective metal precipitation for metal recovery based on the solubility differences among metal compounds [21,41,42].

2.1.2. High Density Sludge

High Density Sludge (HDS) treatment is an improved neutralisation treatment method widely implemented globally for the primary treatment of AMD [30,34,35]. The HDS treatment process recycles and thickens the sludge with flocculation processes to form a hazardous high-density sludge of between 25–30% solids [23,34,43]. The sludge recycling promotes greater solids precipitation by providing a surface for heterogeneous nucleation to catalyse precipitation [44,45]. The HDS process utilises lime more efficiently and the sludge generated is especially high in gypsum [39,46]. The increased sludge density lowers the operational costs for sludge management, lowers the total footprint of the treatment works and produces an improved remediation quality over conventional neutralisation treatment [43,47]. It has been estimated that each megalitre of AMD produces 20 tons of toxic HDS [45,48]. The management of the toxic sludge generated by HDS processes remains a key challenge [45]. Figure 1 shows a simplified process flow diagram of the HDS process.

2.1.3. Chemical Desalination

HDS processes are still limited to low treated water quality, which has prompted further advancements in chemical treatment technologies through the development and deployment of chemical desalination processes such as the CSIR Alkaline–Barium–Calcium (ABC) process and the Magnesium–Barium–Hydroxide (MBO) process [5,32,49]. In the chemical desalination process, the pH is increased sequentially using alkaline reagents and calcium sulphide compounds leading to neutralisation and metal precipitation as metal

sulphides and metal hydroxides. Lime or $Mg(OH)_2$ and barium-containing compounds, typically $BaCO_3$ or $Ba(OH)_2$, are then dosed to raise the pH above 10 with adequate retention time allowed for the reaction, resulting in the precipitation of highly soluble metals Mg, Mn, Ni and Cd as metal hydroxides and the precipitation of SO_4 as $BaSO_4$ [5,50,51]. A variation is the use of calcium sulphide (CaS) with lime to increase the pH to above 10 and the precipitation of metals as hydroxides and sulphides [32,49,52]. A high degree of SO_4 removal is achievable in this process due to the very low solubility of $BaSO_4$ in water, typically below 5 mg/L [53,54]. The pH is then dropped to below 8 through CO_2 dosing before effluent discharge. The advantages of chemical desalination include the high treated water quality, the use of readily available and affordable chemicals and the potential for chemical recover from the sludge for reuse to lower the chemical costs [32,49,50]. The challenges associated with chemical desalination include the use of toxic soluble compounds of Ba and the sludge generation that requires additional processing at an added cost [30]. Figure 2 shows a simplified process flow diagram of a chemical desalination process.
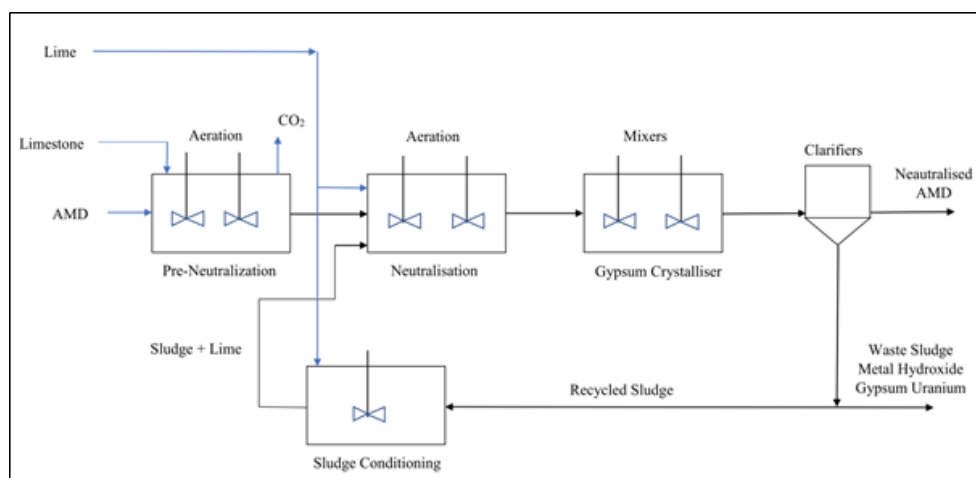

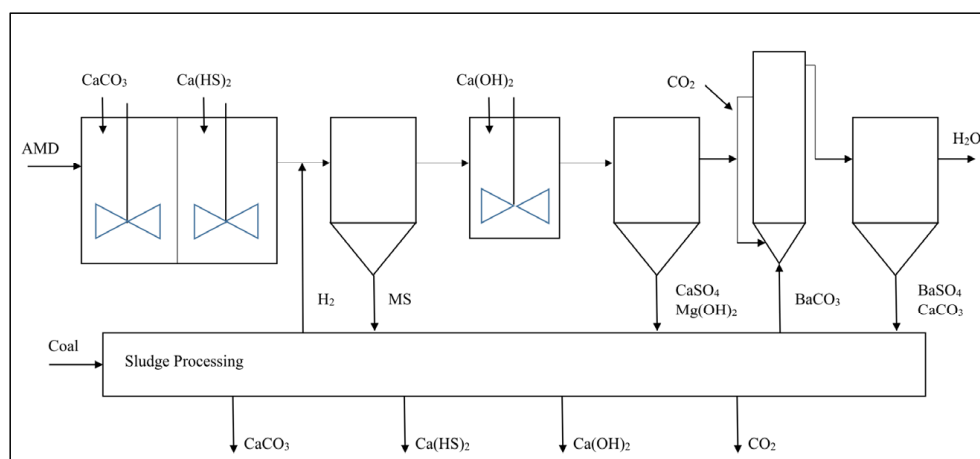**Figure 1.** Process Flow Diagram of HDS Treatment Process [49].

Figure 2. Process Flow Diagram of Chemical Desalination ABC Process [47].

2.1.4. Evaluation of Chemical Treatment Methods

The chemical treatment methods were evaluated for their remediation capabilities of pH, acidity, total Fe, Al, SO_4, Zn, Mg and Mn at the 80% distribution limit and their operational costs from the existing literature. Chemical desalination is capable of fully remediating AMD at the 80% distribution limit with the scalability to accommodate most AMD flowrates and becomes more economical at higher flows as a function of economies of scale [5,50,52]. Conventional neutralisation and the HDS process can effectively reduce

acidity and Fe, Al, Mn and Zn concentrations, but are limited in SO_4 and Mg reduction [35,37,46]. The estimated operating costs exclude the additional sludge handling costs, which are site specific. Table 2 shows the evaluation matrix of the discussed chemical treatment methods. From these observations, chemical desalination is the most promising chemical treatment method available for global AMD remediation.

Table 2. Evaluation matrix for chemical treatment methods.

Criteria	Conventional Neutralisation	High Density Sludge	Chemical Desalination
pH neutralisation of acidic AMD pH = 2.0, acidity = 2000 mg/L	Effective Treated pH > 8	Effective Treated pH > 8	Effective Treated pH > 8
Removal of Total Fe removal Fe = 2800 mg/L	Effective Treated Fe < 1 mg/L	Effective Treated Fe < 1 mg/L	Effective Treated Fe < 1 mg/L
Removal of Al removal Al = 500 mg/L	Effective Treated Al < 1 mg/L	Effective Treated Al < 1 mg/L	Effective Treated Al < 1 mg/L
Removal of SO_4 removal SO_4 = 8000 mg/L	Limited Treated SO_4 > 2500 mg/L	Limited Treated SO_4 > 1900 mg/L	Effective Treated SO_4 < 200 mg/L
Removal of Zn removal Zn = 210 mg/L	Effective Treated Zn < 1 mg/L	Effective Treated Zn < 1 mg/L	Effective Treated Zn < 1 mg/L
Removal of Mn removal Mn = 120 mg/L	Effective Treated Mn < 5 mg/L	Effective Treated Mn < 5 mg/L	Effective Treated Mn < 5 mg/L
Removal of Mg removal Mg = 300 mg/L	Ineffective at changing Mg concentration	Ineffective at changing Mg concentration	Effective Treated Mg < 50 mg/L
Estimate operational costs	USD 1–0.5	USD 1–0.5	USD 0.25–0.75
References	[30,32,55]	[30,45,46,55]	[5,30,32,50]

2.2. Wetlands

Wetlands are large ecosystems with intense biogeochemical activity that play an important role in water treatment [56]. The remediation of AMD using wetlands is the most researched and implemented passive AMD treatment method globally due to the appealing self-sufficiency, pollution free treatment process and minimal maintenance requirements [6,19,20,57]. Wetlands make use of chemical, microbiological, phytoextraction and rizhofiltration processes for the remediation of AMD through the precipitation of metal hydroxides, Biological Sulphate Reduction (BSR), metal sulphide precipitation and direct uptake by living plants [19,57,58]. The vegetation growth, most commonly *Typha* and *Phragmites,* on the submerged substrate of wetlands offers a continuous supply of carbon and energy for the microbiological community [59–61]. Wetlands are often preceded by limestone channels to neutralise the pH of AMD and aid in the metal precipitation in the wetland [62].

The application of wetland is limited to low acidity, low fluctuation of chemical composition and low flow rate AMD sources [63,64]. The key challenges toward the implementation of wetland solutions are the large land area requirements to treat high flows of AMD, the reduction in performance overtime due to the metallic sludge accumulation leading to refurbishment requirements and the high investment costs associated with the construction of wetland systems [64–66]. There are two types of wetlands used for the remediation of AMD, namely aerobic wetlands and anaerobic wetlands. Figure 3A,B shows a simplified process flow of an anaerobic wetlands and aerobic wetlands, respectively.

2.2.1. Aerobic Wetlands

Aerobic wetlands (AeWs) are the most basic passive treatment technique available and are suitable for treating net alkaline AMD with high concentrations of Fe [67]. AeWs oxidise Fe^{2+} and Mn^{2+} to a lesser extent, while allowing a sufficient hydraulic retention

time for the settling of the metal hydroxides [27]. AeWs can also remove metals such as arsenic through co-precipitation due to the adsorption onto positively charged Fe^{3+} that may result in the formation of scorodite ($FeAsO_4$) [6]. The oxidation of ferrous iron is net acid generating and, thus, AeWs are mainly suited for the remediation of net alkaline AMD or for final stage AMD treatment [11,68]. AeWs comprise of shallow basins holding water depths of between 10 to 30 cm and the surface flow of AMD to maintain the oxidising conditions [67]. Plants and other vegetation play an important role in AeWs' performance by regulating and diversifying water flows for optimal surface area utilisation, preventing flow channelling that can lead to reduced hydraulic retention times and stabilising the ferric iron precipitants [6,67]. The limited remediation capability coupled with the remediation application limited to only net-alkaline AMD sources makes the use of AeWs as a stand-alone solution unsuitable for the vast majority of AMD sources [11].
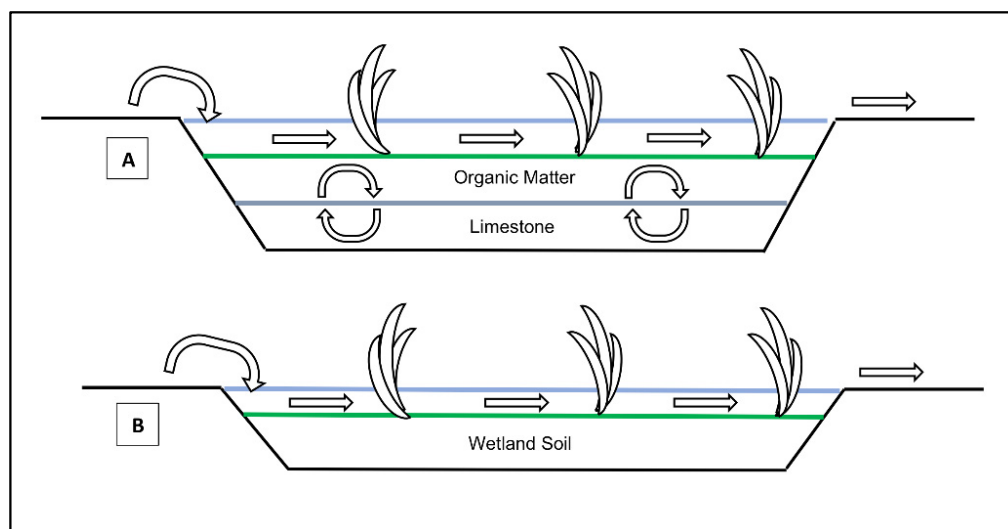
Figure 3. Process flow diagram of (**A**) anaerobic wetlands and (**B**) aerobic wetlands [64].

2.2.2. Anaerobic Wetlands

Anaerobic wetlands (AnWs) offer superior overall AMD remediation performance in comparison to aerobic wetlands. The remediation mechanisms involved in AnWs include biological sulphate reduction, the formation and precipitation of metal sulphides and the formation of carbonate alkalinity [27,31]. AnWs comprise of permeable layers of limestone, organic substrates, plants and other vegetation with water depths of greater than 30 cm to maintain anaerobic conditions [27]. The limestone layer generates alkalinity as a bicarbonate (HCO_3^-) through the dissolution of $CaCO_3$, while the organic substrate layer provides a nutrient and energy supply for the consortium of microbial life [31,67]. One of the main microorganisms involved in AnWs' remediation are Sulphate Reducing Prokaryotes (SRP), which reduce SO_4 to Hydrogen Sulphide (H_2S) gas or dissolved sulphide and generating alkalinity as HCO_3^-. The resulting H_2S can react with dissolved metals causing precipitation as metal sulphides, while concurrently the dissolution of limestone results in an increased pH and further metal precipitation as metal hydroxides [69]. The precipitation of metals as metal sulphide is advantageous over metal hydroxide precipitation due to most metal sulphides being less soluble than metal hydroxides and having a smaller bulk volume, leading to sludge being more compact, which can extend the productive life of AnWs [69–71].

2.2.3. Evaluation of Wetlands

The wetlands were evaluated for their remediation capabilities of pH, acidity, total Fe, Al, SO_4, Zn, Mg and Mn at the 80% distribution limit and their operational costs from the existing literature. Both wetland methods are not suitable for the acidity level and dissolved solids concentrations present at the 80% distribution limit and, therefore,

additional pre-treatment processing steps would be required in order to consider these methods. Table 3 shows the evaluation matrix for AnWs and AeWs.

Table 3. Evaluation matrix for chemical treatment methods.

| Criteria	Aerobic Wetlands	Anaerobic Wetlands
pH neutralisation of acidic AMD pH = 2.0, acidity = 2000 mg/L	Ineffective Limited to treating net alkaline AMD	Ineffective at treating pH of 2 Limited to treating pH > 4.5
Removal of Total Fe removal Fe = 2800 mg/L	Effective Fe < 1 mg/L	Ineffective at changing concentration Pre-treatment required
Removal of Al removal Al = 500 mg/L	Ineffective at changing concentration Pre-treatment required	Ineffective at changing concentration Pre-treatment required
Removal of SO_4 removal SO_4 = 8000 mg/L	Ineffective at changing concentration Pre-treatment required	Ineffective at changing concentration Pre-treatment required
Removal of Zn removal Zn = 210 mg/L	Ineffective at changing concentration Pre-treatment required	Ineffective at changing concentration Pre-treatment required
Removal of Mn removal Mn = 120 mg/L	Ineffective at changing concentration Pre-treatment required	Ineffective at changing concentration Pre-treatment required
Removal of Mg removal Mg = 300 mg/L	Ineffective at changing concentration Pre-treatment required	Ineffective at changing concentration Pre-treatment required
Estimate operational costs	No direct cost	No direct cost
References	[11,23,30]	[11,31,64]

2.3. Membrane Treatment

Membrane technologies include ultrafiltration, nanofiltration, reverse osmosis and membrane distillation amongst others and are generally operated as secondary treatment systems typically following HDS treatment [72,73]. A major benefit of secondary membrane treatment is the high treated water quality of up to potable water standards and the high water recovery rates, which can exceed 90% [74,75]. In addition, membrane treatment methods offer flexibility, scalability and compatibility with various primary remediation processes [42]. The high operational and maintenance costs of membrane technologies due to the high pressures required for membrane treatment and the inevitable scaling of membranes are the leading challenges associated with membrane treatment adoption for AMD remediation [30,32].

2.3.1. Membrane Desalination

Membrane desalination using Reverse Osmosis (RO) processes typically comprise of Ultrafiltration (UF) membranes and RO membranes [76,77]. In some AMD applications, Nanofiltration (NF) may also be used [76]. UF and NF membranes consist of fine pores, of approximately 0.01 and 0.001 microns, respectively, that can trap fine suspended solids and microorganisms in the solution, while allowing the filtered water to pass [78]. RO membranes are driven by positive hydrostatic pressure across a semi-permeable membrane where water molecules and some ions may pass through the membrane, while the remaining are retained in the membrane and discharged as a concentrated brine stream [72]. To increase the water recovery rate across an RO system, the concentrate stream from one RO stage can be fed into a second stage RO for further desalination and increased water recovery, as illustrated in Figure 4 [72]. RO systems have high energy requirements due to the high operating pressures, while also requiring consistent antiscalant dosing to the influent to minimise the effects of membrane scaling, thereby promoting membrane lifespan and economic viability [79,80]. In addition, periodic Cleaning-In-Place (CIP) procedures with warm water and CIP chemicals are necessary to mitigate membrane fouling and the higher the influent Total Dissolved Solids (TDS) is, the more frequent the CIP requirements for

membrane longevity are [81]. The described operational requirements contribute to the high operational costs associated with RO processes.
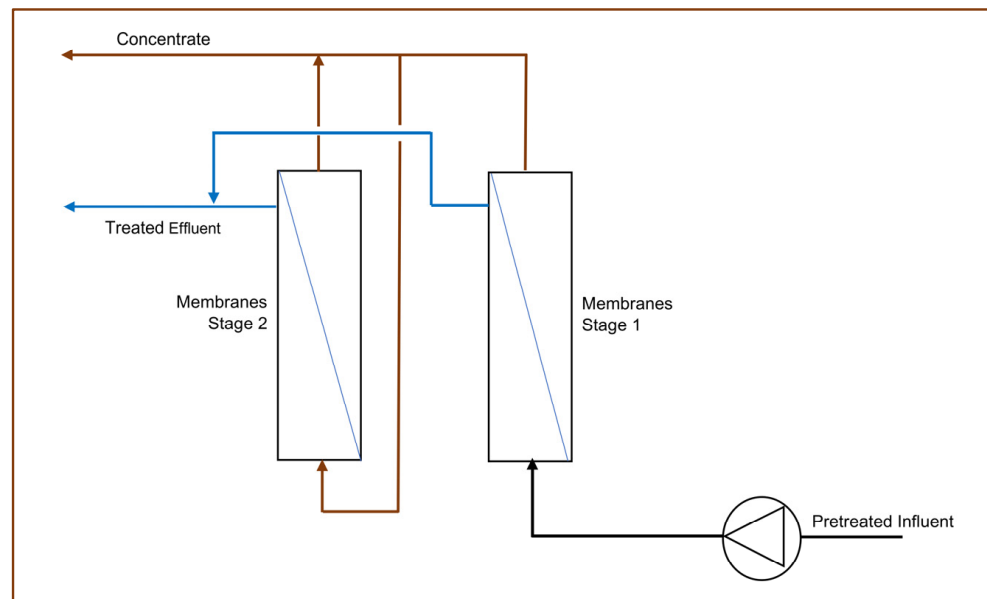
Figure 4. Process flow diagram of 2-stage reverse osmosis.

RO processes with pre-treatment, typically HDS, have proven to be highly efficient at AMD remediation at a large commercial scale [75,77]. The eMalahleni Water Reclamation Plant in the Mpumalanga province of South Africa treats 30 megalitres per day of AMD to potable water quality with the TDS below 450 mg/L and over 98% recovery [75,77]. The process combines HDS pre-treatment with a multistage UF and RO process and is formally known as the Keyplan High Recovery Precipitating Reverse Osmosis (HiPRO®) process [77]. However, due to the high operational and capital investment costs, this treatment option may only be viable for active mines where the costs can be absorbed by the mining operations and the treated potable water can be supplied to the local water supply grid to generate revenue, as is the case with the eMalahleni Water Reclamation Plant [75,77]. At a laboratory scale, AMD treatment using UF followed by NF and RO has been applied with promising outcomes [76]. However, the membrane longevity of this kind of approach at an industrial scale for the global 80% distribution would be a limitation. Figure 4 shows a simplified process flow of a two-stage RO process.

2.3.2. Membrane Distillation

Membrane Distillation (MD) is a thermal driven physical separation process where a hot saline stream is separated into a cold purified stream and a concentrate stream using the vapour pressure difference induced by the differential temperature across the membrane, which results in the separation of dissolved ions from water [73,82]. The process consists of hydrophobic microporous membranes where water vapour molecules are passed through the membrane from the higher vapour pressure side to the low vapour pressure side [42]. MD is effective at the separation and rejection of ions, macromolecules and non-volatile organics present in AMD and can operate at lower pressures then RO systems that translate to energy cost and equipment cost reductions [83,84]. However, additional energy is required for heating the influent AMD and cooling the treated permeate to maintain the differential temperature across the membrane that induces the vapour pressure gradient [82,84]. The feed temperatures can range from as low as 30 °C to as high as 90 °C [82,83]. MD technologies have various operating principals, which include direct contact, air gap and vacuum, amongst others, all of which have varying degrees of effectiveness [85,86]. Figure 5 shows a simplified process flow diagram of the simplest MD operating principal, namely direct contact [86].
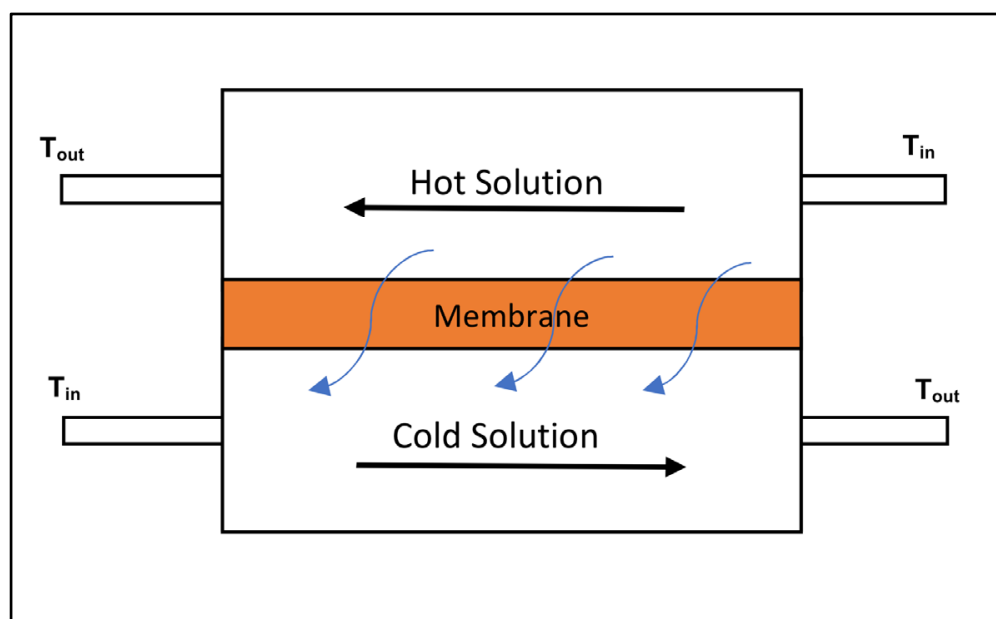
Figure 5. Process flow diagram of direct contact membrane distillation [86].

MD processes have lower equipment costs and lower pre-treatment requirements in comparison to RO processes; however, the technology has yet to be successfully scaled and commercialised for AMD desalination [82,85,87]. Theoretically, membrane distillation methods can achieve a 99.9% rejection of ions [88]. Another advantage of MD processes over desalination membranes is the minimal role the membrane has in the physical separation and the larger pore size that results in less membrane fouling [83]. However, high precipitant deposits have been observed on membrane surface, leading to the partial clogging of membrane pores when operating a submerged direct contact distillation membrane at a bench scale with a model AMD solution [84]. The biggest challenges associated with MD are the scalability of the process, the low permeate yield achieved in non-batch mode applications, the high energy requirements of the process per cubic meter of treated water and the high operational costs associated [83,88,89]. In addition, it has been estimated that MD can be more expensive to operate than RO at a commercial scale [88].

2.3.3. Evaluation of Membrane Treatment

The membrane treatment methods were evaluated for their remediation capabilities of pH, acidity, total Fe, Al, SO_4, Zn, Mg and Mn at the 80% distribution limit and their operational costs from the existing literature. Desalination was evaluated for the proven HiPRO process, which is highly efficient at AMD remediation. Due to the high energy requirements for both membrane technologies, the operational costs are dependent on the energy tariffs in the installed region [76]. Both methods can achieve a high remediation quality; however, MD is limited to low flow rates. Table 4 shows the evaluation matrix of the discussed membrane treatment methods.

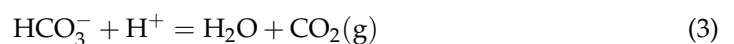**Table 4.** Evaluation matrix for membrane treatment methods.

Criteria	Membrane Desalination	Reverse Osmosis-HiPRO Process
pH neutralisation of acidic AMD pH = 2.0, acidity = 2000 mg/L	Ineffective at pH correction Pre-treatment required	Effective Treated pH between 7–8
Removal of Total Fe removal Fe = 2800 mg/L	Effective Treated Fe < 0.3 mg/L	Effective Treated Fe < 0.3 mg/L
Removal of Al removal Al = 500 mg/L	Effective Treated Al < 0.3 mg/L	Effective Treated Al < 0.3 mg/L

Criteria	Membrane Desalination	Reverse Osmosis-HiPRO Process
Removal of SO_4 removal SO_4 = 8000 mg/L	Effective Treated SO_4 < 250 mg/L	Effective Treated SO_4 < 250 mg/L
Removal of Zn removal Zn = 210 mg/L	Effective Treated Zn < 5 mg/L	Effective Treated Zn < 5 mg/L
Removal of Mn removal Mn = 120 mg/L	Effective Treated Mn < 0.4 mg/L	Effective Treated Mn < 0.4 mg/L
Removal of Mg removal Mg = 300 mg/L	Effective Treated Mg < 120 mg/L	Effective Treated Mg < 120 mg/L
Estimate operational costs	Cost ≥ USD 1	Cost ≥ USD 1
References	[88,89]	[30,32,76,90]

2.4. Biological Treatment

Biological treatment is a secondary remediation process that makes use of the naturally occurring sulphur cycle to reduce SO_4 to H_2S gas [91,92]. The process consists of a consortium of interdependent microorganisms, the most notable of which being Sulphate Reducing Prokaryotes (SRP), primarily represented by the genera of *Desulfovibrio*, *Desulfobacter*, *Desulfomicrobium* and *Desulfootomaculum* [19]. SRP oxidise simple organic matter using SO_4 as a terminal electron acceptor, resulting in Hydrogen Sulphide (H_2S) gas and bicarbonate ions, as indicated in Equation (1) [69,93]. The resulting H_2S reacts with the dissolved metals and precipitates the metals from the solution as insoluble metal sulphides, as indicated in Equation (2) [40,94]. The bicarbonate ion from Equation (1) reacts with the protons (H^+) to form CO_2 and water, thereby removing acidity from the solution, as indicated in Equation (3) [59,95].

$$3SO_4^{2-} + 2CH_3CHOHCOOH = 3H_2S + 6HCO_3^- \tag{1}$$

$$H_2S + Me^{2+} = MeS(s) + 2H^+ \tag{2}$$

$$HCO_3^- + H^+ = H_2O + CO_2(g) \tag{3}$$
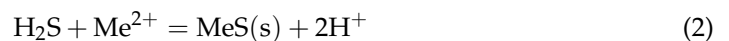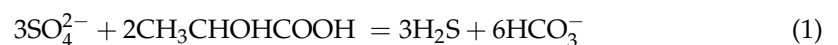
2.4.1. Active Anaerobic Digestion

Active biological treatment using SRP is a promising AMD remediation method with the advantage of combining SO_4, metals and acidity removal into a single Anaerobic Digestor (AD) process with minimal sludge generation [59]. Influential design and operational factors for successful AD biological sulphate reduction include consistency in SO_4 loading rates, pH stability, minimal temperature variance, maintaining a balance between alkalinity and volatile fatty acids, hydraulic retention time, the availability of nutrients nitrogen, phosphorus and potassium (N, P, K), the reactor design, the organic source available for SO_4 reduction and the ability of the SRP to outcompete methanogens for the available organic substrates [43,62,95,96]. Active AD treatment processes typically comprise of automated temperature regulation at mesophilic temperatures of between 35–39 °C, automated flow regulation and automated alkaline dosing to neutralise the acidity generated by the acidogenic microorganisms [30,32].

A major constraint to the AD design for SO_4 reduction is the relatively poor adhesion capability of SRP [96], leading to the research into various AD configurations [7,20,22,40,62,69,94,97,98]. The configurations investigated in the existing literature include the recycling sludge bed reactor [7,62], up-flow anaerobic sludge blanket [40], moving-bed biofilm reactor [97], fixed bed AD [99] and fluidised bed AD [69]. Another great challenge associated with the active biological treatment for AMD remediation is the sourcing of a stable supply of a suitable organic matter source and management of the supply chain over many years [30,32]. This has resulted in the investigation and use

of various organic wastewater streams including primary sewage sludge [7,40,62,100], brewery effluent [27,97] and biowaste [93] for co-treatment with AMD.

The challenges associated with the implementation of active biological treatment for AMD remediation include the high operational complexity, the costs and logistics for a stable supply of a suitable organic matter source, the high process sensitivity to change and the toxicity of high concentrations of heavy metals and high acidity to the biology that necessitates pre-treating [62,94–96].

2.4.2. Passive Bioreactors

Passive bioreactors employ the same anaerobic biological SO_4-reducing principals as active AD remediation without the automated temperature control, alkaline dosing for pH stabilisation and flow rate regulation. Bioreactors employ various organic and alkaline materials and can remove dissolved concentrations of heavy metals and SO_4 from AMD with limited acidity and low total dissolved solids [27,101]. The performance of different organic matter sources is highly varied and can be influenced by the availability of nutrients, microbial diversity, the utilisation of organic materials and the ease of material biodegradation [62,96,101]. Simple organic matter sources are prone to rapid utilisation, which can lead to high performance in the short-term but is not good for long-term AMD remediation [101].

Bioreactors mainly rely on biological SO_4 reduction and pH neutralisation, while alkaline materials also generate alkalinity for pH neutralisation [102]. Metals are typically removed as a result of pH correction and are precipitated as hydroxides, sulphides or carbonates and may also be absorbed in solids present in the bioreactor [69,103]. One of the biggest challenges associated with passive bioreactors is the progressive biological degradation of the organic substrate and its effects on the hydraulics and overall performance of the bioreactor [104]. Additionally, bioreactors are limited to low AMD flow rates [27]. The ambient operating temperatures of bioreactors result in lower microbial activity and longer retention times required for SO_4 reduction when compared with active AD systems under mesophilic conditions [62]. The treatment performance of bioreactors is highly variable and can be influenced by factors such as variation in heavy metals, SO_4 and acidity loading rates, ambient temperature, AMD flow rates, organic substrate source, degree of carbon utilisation and microbial diversity amongst others [69,101,102,104].

2.4.3. Evaluation of Active Biological Treatment

The biological treatment methods were evaluated for their remediation capabilities of pH, acidity and the total Fe, Al, SO_4, Zn, Mg and Mn at the 80% distribution limit from the existing literature. Both remediation methods are ineffective at remediating AMD at the 80% distribution limit due to the high acidity loading and high TDS [7,62,69]. Therefore, pre-treatment is a requirement for both methods. The high costs associated with the sourcing of suitable organic matter and the supply chain management involved for AD remediation are dependent on various factors that are site specific. Table 5 shows the evaluation matrix of the discussed biological treatment methods.
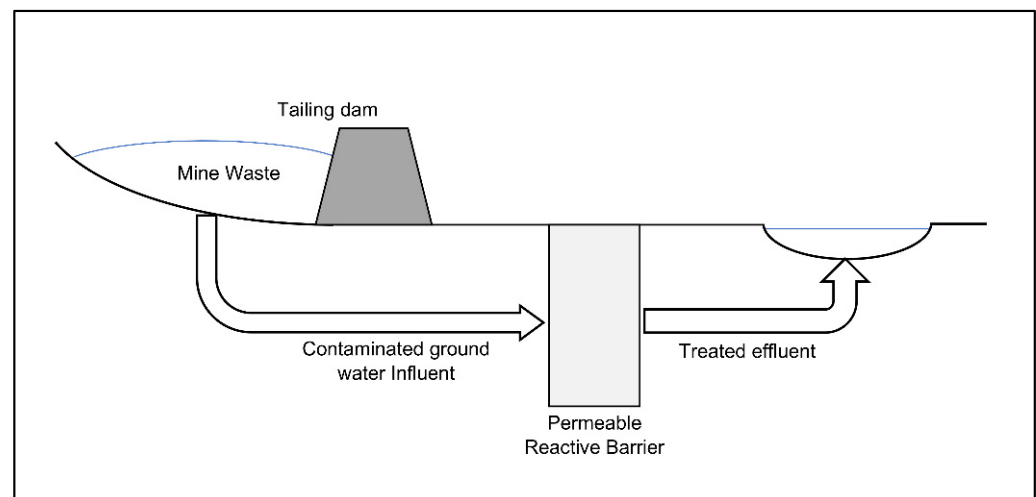
2.5. Permeable Reactive Barriers

Permeable Reactive Barriers (PRBs) are an emerging AMD treatment method characterised by their high porosity and various material compositions that can include alkaline materials such as concrete, cement, fly ash and lime as well as various organic materials and electron donor materials such as zero-valent iron [105–109]. PRBs can be installed underground, directly in the flow path of contaminated groundwater and used as in situ systems, which eliminates the costs associated with the pumping and distribution of mine water to treatment works [25,110]. Other advantages of PRB remediation include the low operational cost and the minimal operational supervision required [25,30,111]. However, the effectiveness of a PRB system for AMD remediation is limited by the depletion of the chemical components of the reactive barrier, armouring of the reactive surface and the

physical clogging of the porous network with time [62,110,112]. In addition, PRBs have high construction and installation costs with the trenching for in situ PRBs accounts for an estimated 70% of the total capital investment [110,113]. PRBs employ mechanisms of adsorption, precipitation and biological degradation for remediation [114]. Various PRB materials have been investigated and used for AMD remediation, this section focuses on the three main materials used, namely zero-valent iron, pervious concrete and organic substrates. Figure 6 shows a simplified process flow diagram of an in situ PRB.

Table 5. Evaluation matrix for biological treatment methods.

Criteria	Anaerobic Digestor	Passive Bioreactors
pH neutralisation of acidic AMD $pH = 2.0$, acidity = 2000 mg/L	Ineffective at low pH Pre-treatment neutralisation required	Ineffective at low pH Pre-treatment neutralisation required
Removal of Total Fe removal $Fe = 2800$ mg/L	Ineffective at changing concentration Pre-treatment neutralisation required	Ineffective at changing concentration Pre-treatment neutralisation required
Removal of Al removal $Al = 500$ mg/L	Ineffective at changing concentration Pre-treatment neutralisation required	Ineffective at changing concentration Pre-treatment neutralisation required
Removal of SO_4 removal $SO_4 = 8000$ mg/L	Effective Treated $SO_4 < 200$ mg/L	Effective with high retentions times Treated $SO_4 < 1000$ mg/L
Removal of Zn removal $Zn = 210$ mg/L	Ineffective at changing concentration Pre-treatment neutralisation required	Ineffective at changing concentration Pre-treatment neutralisation required
Removal of Mn removal $Mn = 120$ mg/L	Ineffective at changing concentration Pre-treatment neutralisation required	Ineffective at changing concentration Pre-treatment neutralisation required
Removal of Mg removal $Mg = 300$ mg/L	Ineffective at changing concentration Pre-treatment neutralisation required	Ineffective at changing concentration Pre-treatment neutralisation required
Operational costs	Cost $\geq$ USD 1 per m^3	No direct cost
References	[7,40,62,97]	[59,69,100]

Figure 6. Process flow diagram of in situ permeable reactive barriers for contaminated groundwater [23].

2.5.1. Zero-Valent Iron

Zero-Valent Iron (ZVI) is the most widely used reactive material for groundwater remediation PRBs [115,116]. ZVI treats AMD through the reduction in heavy metals and co-precipitation with secondary precipitants [117]. ZVI has been found to be effective at removing arsenic, heavy metals and nitrate [108]. ZVI is typically used as one of numerous reactive materials in an AMD PRB's composition. Some tested PRB designs have included ZVI, organic substrates and limestone with the aim of using ZVI to reduce heavy metals and to enhance the reducing conditions for an anaerobic biological sulphate reduction

with SRP [118]. In other experiments, ZVI has been used with silica sand and the effective removal of Al, Mn, Ni, Zn, Co and Cu was achieved, while the reduction in SO_4 was limited [115,119]. Ekolu and Bitandi (2018) found that ZVI-based PRBs are not as effective at raising the AMD pH as pervious concrete PRBs, and pervious concrete PRBs have a higher overall contaminant removal efficiency. As is the case with all reactive barriers, clogging and armouring are key challenges associated with the use of ZVI PRBs [110,112].

2.5.2. Pervious Concrete

Pervious concrete PRBs neutralise AMD acidity through the dissolution of the portlandite ($Ca(OH)_2$) found in the concrete mix, resulting in metal precipitation as metal hydroxides and SO_4 reduction through gypsum crystallisation [107,110,120,121]. Pervious concrete PRBs have been shown to be effective in the removal of metals Al, Fe, Mn, Co and Ni, while other metals such as Ca, Mg and K may increase in concentration following treatment through the leaching of the metals from the concrete [115,122,123].

The key design considerations for pervious concrete PRBs include the type of cement, the aggregate size, the aggregate material and the selection of extender materials [107,109,120]. The clogging of the porous surface, the armouring of the reactive surface and the depletion of the reactive materials lead to the reduction in efficiency of the pervious concrete PRBs over a prolonged operation [110,112].

2.5.3. Organic Matter

Organic matter mixed with alkaline reagents have been piloted as PRB reactive materials for AMD remediation [114,118,124,125]. Organic matter PRBs use physicochemical and biological sulphate reducing mechanisms for the remediation of AMD [124,125]. Organic substrate PRBs operate very similarly to bioreactors, with the biggest differences being the process configuration. Various studies have been conducted to evaluate the use of organic waste materials such as saw dust, vegetable composites, manure, kraft pulp mill alkaline residue and sewage sludge amongst others as reactive material for the biological remediation of AMD in PRBs [114,118,124,125]. Similar to bioreactors, the remediation performance of different organic substrates is highly variable [114,118,124,125]. The progressive biological degradation of the organic substrate limits organic matter PRBs to short term remediation with high replacement costs [104]. The availability of nutrients N, P and K in the organic matter source can promote microbial growth and enzymic activity [96,124]. The microorganisms involved are sensitive to process variations, including acidity, SO_4 and heavy metal loading [96,114]. For effective bioremediation in PRBs, high retention times are required [124,125], which increases the capital investment costs [110,113]. With a sufficient hydraulic retention time, low heavy metal concentrations and low acidity, organic matter PRBs can effectively reduce SO_4 and precipitate trace metals [114,118,124,125].

2.5.4. Evaluation of PRB Methods

The PRB treatment methods were evaluated for their remediation capabilities of pH, acidity and the total Fe, Al, SO_4, Zn, Mg and Mn at the 80% distribution limit from the existing literature. Due to the high acidity and high concentrations of heavy metals at the 80% distribution limit, ZVI and pervious concrete PRBs require high retention times to effectively raise the pH, precipitate heavy metals and precipitate some dissolved SO_4 as gypsum. The metal concentrations are toxic to the consortium of microorganisms involved in biological sulphate reduction and, thus, the use of organic matter PRBs is not suitable without pre-treatment. Table 6 shows the evaluation matrix of the discussed PRB treatment methods.

Table 6. Evaluation matrix for PRB methods.

Criteria	Zero-Valent Iron PRB with Alkaline Reagent	Pervious Concrete PRB	Organic Matter with Alkaline Reagent
pH neutralisation of acidic AMD pH = 2.0, acidity = 2000 mg/L	Effective Treated pH > 7	Effective Treated pH > 8	Not effective Pre-treatment required
Removal of Total Fe removal Fe = 2800 mg/L	Effective Treated Fe < 10 mg/L	Effective Treated Fe < 10 mg/L	Not effective Pre-treatment required
Removal of Al removal Al = 500 mg/L	Effective Treated Al < 10 mg/L	Effective Treated Al < 10 mg/L	Not effective Pre-treatment required
Removal of SO_4 removal SO_4 = 8000 mg/L	Limited Treated SO_4 > 2500 mg/L Post treatment required	Limited Treated SO_4 > 2500 mg/L Post treatment required	Effective at very long retentions. Treated SO_4 > 250 mg/L
Removal of Zn removal Zn = 210 mg/L	Effective Zn < 10 mg/L	Effective Zn < 10 mg/L	Not effective Pre-treatment required
Removal of Mn removal Mn = 120 mg/L	Not effective Pre-treatment required	Effective Mn < 10 mg/L	Not effective Pre-treatment required
Removal of Mg removal Mg = 300 mg/L	Not effective Pre-treatment or post treatment required	Not effective, Concentrations can increase	Not effective Pre-treatment required
Operational costs	Cost < USD 0.5 per m^3	Cost < USD 0.5 per m^3	Cost < USD 0.5 per m^3
References	[108,115,126,127]	[107,109,115,120]	[124,125]

3. Future Prospective and Research Needs

In this paper, twelve remediation options were evaluated for AMD remediation capabilities and economic viability based on operational costs. For eleven of the twelve remediation options reviewed, additional process steps were required either for further treatment to achieve the discharge limits or for the safe management of hazardous waste by-products. The one exception was the HiPRO process, which also has the highest operational cost and is not economically viable for use on abandoned mines. In this review, chemical desalination methods were found to have the greatest potential for suitability for Class 0 and Class 1 AMD as a result of their medium to high operational costs (between USD 0.25–0.75 per cubic meter of AMD influent) and the high treated water quality achievable [5,30,49,50,127]. Chemical desalination also presents advantages, such as great flexibility, scalability, capability for chemical recovery from sludge and end-to-end treatment from raw AMD to discharge quality [26,36,49,50]. However, limited research is available on industrial scale AMD remediation using chemical desalination and there is a need for the further development of this potential solution to improve the economic viability for abandoned mines [30].

The management of the hazardous by-products present in the sludge stream remains a major challenge for sustainable AMD remediation. Management practices commonly involve the concentration of the heavy metal and sulphide precipitants into a high-density sludge stream for dewatering and disposal. More sustainable management approaches integrate mineral recovery into the process design to enable the reuse of the by-products. Chemical treatment technologies currently present the greatest opportunity for mineral recovery through the selective precipitation of ions due to the changes in the solubility of heavy metals at different pH and dissolved oxygen concentrations [30,32]. Further research is required to evaluate the effectiveness of by-product management practices.

A greater research focus is required toward the development of end-to-end remediation methods that can be scaled up for abandoned mine AMD flow rates. The current remediation options offer trade-offs between operational cost, treated water quality, longevity, scalability, influent quality treatable, capital costs and the land surface area required. These trade-offs are most distinctly visible between the passive and active treatment options

where operational cost benefits are weighed against remediation efficiency. Opportunities exist to combine active and passive methods to develop end-to-end AMD remediation with lower operational costs while maintaining process reliability and treated water consistency. Further research is required in this area, with a focus on the scalability and versatility to accommodate the Class 0 and Class 1 AMD remediation at economical costs.

4. Conclusions

This study was conducted to evaluate the AMD remediation capabilities of the existing technologies against the modified Hill Framework. Twelve remediation options were evaluated, of which chemical desalination showed great promise with high quality treated water, operational costs between USD 0.25–0.75 per cubic meter of AMD influent, with opportunities to generate revenue streams from process by-products and gaps available for further development and commercial scaling. Further development of end-to-end remediation methods suitable for Class 0 AMD at economical operational cost is required in order to effectively address the ongoing environmental challenges posed by AMD globally.

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