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Review

Recent Progress on Acid Mine Drainage Technological Trends in South Africa: Prevention, Treatment, and Resource Recovery

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Abstract: South Africa is the home of major global mining operations, and the acid mine drainage (AMD) contribution has been attributed to abandoned mine sites and huge pyrite-bearing tailings from coal and gold mines. Determining the true economic impact and environmental liability of AMD remains difficult. Researchers have been looking into several treatment technologies over the years as a way to reduce its possible environmental impact. Different methods for active and passive remediation have been developed to treat AMD. The aim of this review was to describe the AMD-impacted environments and critically discuss the properties of AMD and current prediction and preventative methods and technologies available to treat AMD. Furthermore, this study critically analysed case studies in South Africa, gaps in AMD research, and the limitations and prospects offered by AMD. The study outlined future technological interventions aimed at a pattern shift in decreasing sludge volumes and operational costs while effectively improving the treatment of AMD. The various treatment technologies have beneficial results, but they also have related technical problems. To reduce the formation of AMD, it is recommended that more preventive methods be investigated. Moreover, there is a current need for integrated AMD treatment technologies that result in a well-rounded overall approach towards sustainability in AMD treatment. As a result, a sustainable AMD treatment strategy has been made possible due to water reuse and recovery valuable resources such sulphuric acid, rare earth elements, and metals. The cost of AMD treatment can be decreased with the use of recovered water and resources, which is essential for developing a sustainable AMD treatment process. More study is required in the future to improve the effectiveness of the various strategies used, with a focus on reducing the formation of secondary pollutants and recovery of valuable resources.

Keywords: acid mine drainage; sustainable remediation technologies; prevention and prediction; resource recovery

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Citation: Baloyi, J.; Ramdhani, N.; Mbhele, R.; Ramutshatsha-Makhwedzha, D. Recent Progress on Acid Mine Drainage Technological Trends in South Africa: Prevention, Treatment, and Resource Recovery. *Water* **2023**, *15*, 3453. https://doi.org/10.3390/w15193453

Academic Editor: Liliana Lefticariu

Received: 4 August 2023
Revised: 27 September 2023
Accepted: 28 September 2023
Published: 30 September 2023

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1. Introduction

Acid mine drainage (AMD) is a persistent pollutant as a result of current and past mining activities, which is currently one of the critical environmental challenges in South Africa and globally. AMD presents a challenge for operational and deserted mines, in shafts below ground, open holes, waste rock mounds, and powder tailings [1–4]. AMD is more serious in deserted and inactive mines, where there is no pumping occurring and the water table recoils, in contrast to active mines where the water table levels are kept to a minimum through the use of pumps [5–7]. Besides the environmental impacts, AMD also impacts sustainability, which includes environmental, community, and financial concerns. AMD has an effect on the removal of resources, which has an effect on economic activity, infrastructure, and people's livelihoods. This has a substantial effect on the guidelines of developing countries for climate change and their efforts to switch to becoming 'green' economies. New sustainable technologies, efficient management plans, and AMD treatment procedures are currently required. In South Africa, AMD, mainly from gold mine tailings dams/slime dams, is one of the most serious environmental and socio-economic challenges [8,9]. Acidity is formed when pyrite, in the gold-bearing ores, is oxidised and encounters atmospheric oxygen. This oxidation goes through several steps, as follows:

$$4FeS_2 + 15O_2 + 14H_2O \rightarrow 4Fe(OH)_3 + 8H_2SO_4 \quad (1)$$

On the left side of the above equation, pyrite is ferrous sulphide and the right has ferric oxide and sulphate. Thus, both the iron and sulphur components of pyrite have been oxidised. Acid mine drainage develops when water permeates through the zone of oxidised pyrites forming sulphuric acid, which drains out into the surrounding environment [8,10].

AMD causes significant environmental problems that are both locally and globally intractable in the near future. It takes development of effective, innovative, and affordable approaches for addressing and overcoming these issues. Mining for minerals, such as gold, copper, and nickel, has been linked to AMD issues, which could have long-term consequences for streams and biodiversity. Some metal mining effluents contain high levels of poisonous cyanides and heavy metals, which have major human health and environmental consequences [3,11,12]. To remediate AMD, many wastewater treatment technologies have been used, including neutralization [13], selective precipitation [14], use of membranes [15], exchange of ions [16], and the removal of sulphate biologically [17]. The challenge, however, is that the constituents of AMD, while hazardous, may be collected and turned into valuable materials that can be commercialized. Sulphuric acid, for example, has a major demand in the chemical and metallurgical industries [18,19]. While the need for critical minerals and erratic metals in the ground is probably set to increase going forward, there is a desire to create innovative results that blend ground earth metal reclamation with AMD remediation [20,21]. The financial advantages could then be utilized to offset the entire expense of AMD treatment.

To that purpose, the most recent research on AMD development, prevention, and treatment is summarized and critically reviewed. The central focus of this article is to evaluate studies on the prevention, prediction, impact of AMD on the water quality, management, and prospective reclamation of beneficial by-products from mine drainage. The specific objectives are to (1) briefly present the past impacts and possible future effects of AMD on the water condition in South Africa, (2) discuss the developments on the prediction and prevention of AMD, and (3) review of the main large-scale AMD treatment processes applied locally for AMD elimination and simultaneous recovery of the valuable by-products.

2. Past Impact of AMD on the Condition of Water in South Africa

Mining has been the backbone of the South African economy for many years. Coal and gold mining are the major sources of AMD as sulphide-bearing materials are concentrated in geological environments containing coal and gold ore deposits [22]. Gold mining pollution is associated mainly with the release of harmful elements from tailings and other types of mining waste [23]. The infiltration of water through sulphide-containing tailings piles and ponds, surface and underground workings, waste, and development rocks leads to the leaching of large volumes of metals like zinc, nickel, lead, copper, and sulphate ions into streams and river ecosystems [24]. This results in acid mine drainage (AMD) with severe detrimental effects on the receiving water bodies. Heavy metal pollution and acid mine drainage is a very important environmental concern where waste materials containing metal-rich sulphides from mining activity have been stored or abandoned. Tailings and rock dumps are associated with surface impacts which greatly affect surface and ground water quality. The underground impacts are caused by the influx of water into the underground workings and the subsequent dewatering of the aquifer [25].

To maintain safe mining conditions, over 120 mines would have to pump out the water that had been collected in them. However, as the mines became derelict, drainage of the mine voids became less frequent, and the voids started filling up with water which resulted in AMD [26]. AMD can contaminate shallow aquifers, and if the underground mine water reaches the near surface, it starts to decant and flows down to wetlands, streams, and rivers. Since these rivers are used as a water source for agriculture, recreation, and drinking purposes, AMD potentially affects the quality of this water. The two main types of mining occurring in South Africa contribute significantly to the generation of AMD, as listed below in Table 1. The resultant water bodies affected by this AMD pollution are also shown in Table 1 together with the affected areas.

The Western, Central, and Eastern Basins have been highlighted since they have been affected the most by AMD. The mining shafts started decanting in 2002, polluting the Tweelopiespruit that drains into the Krugersdorp Game Reserve [27]. There are abandoned mines which are not operational which have become sources of AMD and water ingress into the shafts. AMD decants through three old mine shafts [28]. The volume being decanted was noted at an average of 20 ML/d, and up to 60 ML/d during the wet season. Approximately 12 ML/d is treated partially, and the remainder flows into the Tweelopiespruit. About 27 ML/d of AMD needs to be treated in this basin to maintain the water below the environmental critical level (ECL). The long-term effects of decanting into this stream result in the pollution of the Hartbeespoort Dam, the Crocodile River, and the transboundary Limpopo River [29].

Table 1. Types of mining in South Africa that predominantly contribute to AMD pollution and their areas of impact.

	Provinces	Key Areas	Water Resources Impacted	References
Gold	Northwest, Gauteng, Mpumalanga, Limpopo	Within Gauteng: Witwatersrand gold spans the Central and Eastern Basins Within the Witwatersrand Eastern Basin: Brakpan, Springs, Nigel Klerksdorp Kloof, Driefontein, Western Deep Levels	Tweelopiespruit, Hartbeespoort Dam, Crocodile River, Limpopo River, Vaal River, Klip River, Blesbokspruit, Barrage, Vaal Dam	[30–32]
Coal		Witbank, Delmas, Secunda	Boesmanspruit, Blesbokspruit, Vaal River	[33–35]

Since the quarrying of mines began, dumps containing the wasteful end-products from gold mining have become a common sight around mining towns and have been releasing contaminated water for decades. Tailings dumps flourish in the upper catchments of springs at the Blesbokspruit and Klip Rivers, where this pollution is predominant [36]. The Witwatersrand gold mines have shut down over several years, and water began to fill in the voids as the pumping of the mines had stopped. This accumulated water travelled into neighbouring mines since all the mines are connected. This process forced these neighbouring mines to take on the responsibility of pumping. A subsidy was initiated by the government to help the mines cover the costs involved in pumping the additional high volumes of water which filled up and requires treatment to an acceptable quality. This treatment involves the use of lime to increase the pH levels and the pumping of oxygen into the water to trigger the iron to oxidize and precipitate along with several other heavy metals. The precipitated iron will settle out, separate, and be discarded in dumps of tailings and the remainder of the water is diverted into local rivers. However, these discharges increased the number of pollutants previously transported via the rivers through the mining towns.

The salt levels present in the Vaal River indicate the effects of long-winded and point source pollutants stemming from the gold mines in the Central and Western Basins, which has increased to greater than twice the amount between the Barrage and Vaal Dams. This is due to the incoming water from the Klip and the Blesbokspruit Rivers (via the Sukerbos River) [37]. The water composition is not good at the Barrage which makes it necessary to periodically let go off water from the Vaal Dam to decrease the salt content for the users downriver of the Vaal. During the rainy season this is not a problem, but during a drought, this could pose a challenge, when the water upriver of the Vaal system, which is mainly for Gauteng, must be discharged for the aim of dilution [38]. The void began to fill once the last Goldfield mine shut down and stopped pumping water. Decantation of the Western Basin began in 2002. When draining of the Central Basin ceased in 2008, the water levels continued to rise at a rate of 12 m per month. Pumping at the Eastern Basin started to slow down in late 2010, and eventually stopped at the beginning of 2011. The decanted water emanating from the void is of very bad quality, as is noted from the water draining from the Western basin. The level of sulphate is typically approximately 3500 mg/L, with a pH range of 2–3. There are also elevated iron and other heavy metal concentrations present in the water. Oxidation occurs when the iron is exposed to air which leaves a bright orange stream of precipitate on the banks and beds of rivers. It is expected that the basins in Boksburg (Central) and Nigel (Eastern), where the bottommost shafts are located, will decant in approximately three years if there are no interventions put into place [39].

These points of decantation are created on the assumption that the water is spontaneously flowing via the cavities and that mine quarries are the only gaps to these voids. However, this may not be true. For example, in the Western Basin, it was found that water was draining from a borehole on a farm, and thereafter from a longstanding mine quarry which may have or may not have been linked up to the central void. Many decant points can arise if the flow rate through the void is not enough to allow for the inflow.

3. Possible Future Impacts of AMD on the State of Water in South Africa

The Olifants River catchment is in a state of deterioration [40]. There was an idea to connect a plant to treat the high number of pollutants in Brugspruit which is close to Witbank, but the idea had limited efficacy [40]. The main purpose was to address the pH challenge and it did not impact the salt concentrations in the water. A plant to treat water was commissioned in the area (eMalahleni Water Reclamation Plant) which operates by reverse osmosis [41]. This plant showed the potential to treat highly polluted water to an acceptable standard for drinking purposes. The setback is that this water costs more than (approximately triple) the water that is distributed to this area from the Vaal River by the local water agency [42]. This plant, although beneficial in producing drinking water for the public, has a limitation of not being beneficial for the complete improvement of rivers that are polluted in the area [28]. The state of the water of the Olifants River will remain in a state of deterioration in the future [43]. This is due to the massive quantities of coal that are found in the Olifants Catchment, which are not mined [44], thereby leading to a rise in pollutants in the future.

Coal mining was occurring for a vast number of years in the upper catchment of the Vaal River [45]. These mines are mostly deep and are still actively operated and overseen. Nevertheless, it was found that there is a high inflow of applications for permits for new mines in that catchment. Funds were set aside by the government in the past to tackle the issue of decanting from the gold mines in Witwatersrand, which will involve draining as well as the straightforward operational treatment being reinitiated (lime being added and iron being removed) in the goldfields which are presently affected [46]. This will aid in stopping the unrestrained draining of the basins (Western, Central, and Eastern) [47]. However, as much as this intervention will vastly help to improve the Western Basin, it will not affect the state of the water of the Vaal River. Instead, it will take the system back to its original state at a time when the mines were still being pumped and treated and water was drained from the mine cavities.

Several different technologies have been developed for desalination of contaminated water from local mines. Only one of these has been commercialised and is being implemented, reverse osmosis treatment technology (used at Witbank), which has shown that although this type of reverse osmosis treatment may help overcome the challenges, it is not feasible [42]. It is possible that although most of the suggested treatment methods are appropriate for remedying water with contaminants at the point source (e.g., pumped from old mines), it is unlikely that polluted water from diffuse sources such as waste dumps, could be treated as well [48]. For gold mines, the water accumulated in the cavity can most often be accessed and treated as a point source. Coal mining is more composite however, and it may not be possible to avoid uninhibited draining of AMD from restored opencut mines [49]. Thus, the state of water in these regions should be anticipated to decline.

Since the treatment techniques used for prevention do not need to be continuous (for maintenance as well), they are more viable than conventional treatment techniques. However, most preventative treatments, such passivation and microencapsulation, are still under experimentation and focus on pure pyrite systems [50] The results were encouraging for microencapsulation techniques; however, this is restricted to batch, single-metal systems. Therefore, it is unclear if they may be used for waste rocks and mine tailings that contain a variety of minerals such silicates and aluminosilicates in complex environmental conditions. This suggests that continuous tests should be conducted to determine the efficacy of various microencapsulation techniques using real or synthetic tailings, including waste rocks high in pyrite [51]. It is important to thoroughly examine the long-term stability of treated waste rocks and tailings under environmental conditions, such as drying–wetting cycles. Utilizing or recycling mine waste for use in building and geopolymer materials is another choice to consider while attempting to limit the generation of AMD. Finding value in and managing mine waste has become incredibly important [52].

4. AMD Prevention

The complexity of the treatment system that is required to guarantee that effluent standards will be satisfied depends on a number of variables [53]. These include the chemical properties of the AMD, the volume of water that needs to be treated, the local climate, the topography, the properties of the sludge, and the anticipated lifespan of the plant [53]. Various treatment techniques have been developed and can be categorised as either 'abiotic' or 'biotic', the former of which does not rely on biological activities while the latter does [5].

Passive treatments involve the passage of mine water through a controlled environment, rather than a receiving water body, where naturally occurring geochemical and biological reactions take place and improve the mine water quality [10,54,55]. Examples of passive abiotic treatment include anoxic limestone drains, open limestone channels, limestone leach beds, slag leach beds, diversion wells, limestone sand, and oxidation channels [12]. These materials all generate alkalinity which help to neutralise the AMD and raise the pH, while at the same time oxidising and precipitating out metals. Passive biotic treatments, on the other hand, include wetlands and bioreactors where natural biological processes work, either in aerobic or anaerobic conditions, to neutralise the AMD and precipitate the hazardous concentrations of contaminants (e.g., metals) over time [12,56]. In line with this, Ramla and Sheridan [57] proved the efficacy of utilizing indigenous South African grass as a suitable organic substrate for sulphate-reducing bacteria to reduce sulphate to sulphides during the passive biotic treatment of AMD. In this experiment, Hyparrhenia hirta grass supplemented with soil containing microbes produced the best outcomes.

Both passive biotic and abiotic AMD treatments require relatively little resource input, tend to be more useful for AMD flows of less than 2 to 5 ML/d with low acidity, e.g., <800 mg/L as $CaCO_3$, and which require little metal and sulphate removal [54,58,59]. However, in comparison to active treatments, passive treatments need larger areas of land and additional time to neutralise AMD and precipitate the contaminants. Thus, passive options are more applicable when AMD treatment needs to be accomplished at closed mine sites with low AMD flows as they are a potentially lower-cost, longer-term sustainable option. An example of one that has been studied in some detail is the passive system set up to treat AMD mine seepage from a long-abandoned mine near the town of Red Oak in eastern Oklahoma [56]. The benefits of passive systems are their self-sufficiency, infrequent maintenance requirements, and extremely low operating and capital expenses. However, the quality of the resultant effluent is poorer than that produced by active treatment systems [54].

Unlike passive treatments which depend mostly on naturally occurring reactions, active AMD treatment is performed in a constructed plant where processes are controlled and sustained via the continuous input of resources [60]. It involves the utilisation of alkaline substances to increase the pH of the drainage and precipitate heavy and toxic metals from the AMD [55]. In line with this, the operating and capital costs of sustaining effective and efficient functioning of the plant can be high as it requires a continuous supply of chemicals, electrical and mechanical power sources, and the employment of operations and maintenance staff [60]. Therefore, active AMD treatment is more suited for application at operational mine sites where the necessary resources are more readily available [61]. Additionally, the kind of neutralizing agent utilized affects the effectiveness, cost, and potential environmental effects of using an active treatment system [62]. The selection of the neutralizing agent is based on the chemical composition of the AMD, site-specific conditions, and expected outcomes with the understanding that some level of cost and benefit trade-off will be required. For instance, sodium hydroxide is more effective in AMD treatment than lime but is approximately 1.5 times more expensive and must be handled in line with specific health and safety requirements due to its hazardous nature [62]. Likewise, anhydrous ammonia requires safe handling and if excessively used can spur nitrification or denitrification in receiving water bodies [62]. In some cases, the split treatment of AMD may yield the most desirable results, e.g., using lime and limestone [63].

The benefits of active AMD treatment, however, can be considered as great advantages over passive treatment techniques [64]. These include that active treatment can be applied to all AMD flow rates, it is fast and effective, it produces good quality effluent with a potential for cost recovery via the sale of the resulting water, metals, and by-products and involves a lower cost in the handling and disposal of generated sludge [10,60,62,65]

Traditional abiotic active treatment of AMD is characterised by the use of alkaline chemicals to neutralise acids, deactivate metals, and precipitate salts [66,67]. Calcium-, sodium-, ammonium-, or magnesium-based chemicals that have been, and are, used to neutralise AMD include calcium carbonate (limestone, $CaCO_3$), calcium hydroxide (slaked lime, $Ca(OH)_2$), calcium oxide (lime or quick/burnt lime, CaO), sodium hydroxide (caustic soda, NaOH), sodium carbonate (soda ash, Na_2CO_3), ammonium hydroxide (NH_4OH), and magnesium hydroxide ($Mg(OH)_2$) [68]. Using calcium hydroxide, calcium oxide, or calcium carbonate can result in large amounts of sludge that retain water as calcium will bond with sulphates and then precipitate out of solution, together with the metals (usually as hydroxides), at higher pH levels [69]. Recycling this sludge is difficult, leaving disposal to landfill or sludge dams as the main option for handling this waste [69]. Furthermore, some metal hydroxides are amphoteric, which presents the probability for dissolution of potentially harmful chemicals from the sludge both during and after disposal [70]. Thus, the sludge disposed at landfill sites or sludge dams would need to be properly managed and regularly monitored to ensure that no long-term negative environmental impacts occur [69]. In addition, landfills and sludge dams can occupy large areas of land, especially when considerable amounts of sludge are produced and disposed of [69]. Neutralising chemicals such as magnesium hydroxide, ammonium hydroxide, and sodium hydroxide have proven to be comparatively more useful as they tend to precipitate metals (e.g., as hydroxides) while leaving the sulphate in solution. This sulphate can subsequently be treated to produce gypsum, which may be valuable in other economic and industrial sectors.

An area of rising importance in the active abiotic treatment of AMD is the use of waste by-products to treat other wastes. An example of this is the use of calcium-containing waste such as dust from cement and lime kilns to neutralise AMD and precipitate metals. Another example is the use of coal combustion by-products to partially treat AMD. Coal-based by-products generally are very good adsorbents because they have a high surface area, microporous structure, and high surface reactivity [71]. However, these by-products seem to be best at removing trace concentrations of more toxic metals such as radioactive thorium, uranium, radium, and lead. Kaur et al. [67] also describes the use of an alkaline waste material from the alumina refining industry as a possible alternative neutralising material. The costs to obtain and use one waste to treat another is often much lower, making AMD neutralisation and other treatment processes (e.g., metal extraction) potentially much cheaper. However, the feasibility of these types of options will vary depending on the type, availability, and location of the various wastes as well as the properties of the AMD. In addition, it must be noted that calcium-based neutralisation chemicals tend to produce considerable quantities of waste sludge which would still need to be dealt with if cement waste or lime kiln dust is used. Other examples of good absorbents that have shown potential to treat AMD include bauxite and naturally occurring bentonite clay. Bentonite (primarily aluminium phyllosilicate) has been used to neutralise AMD and remove metals. However, the suitability of these type of adsorbents needs to be investigated on a large, long-term scale to prove that they can work as well as the current technologies whilst also being more sustainable and cost-effective.

Active biotic AMD treatment involves the use of off-line sulfidogenic bioreactors where the hydrogen sulphide produced by sulphate-reducing bacteria (SRB) is used both to add alkalinity to neutralise the acidic waste streams and to precipitate metals as insoluble sulphide precipitates, which may then be recovered and reprocessed [5,72]. Table 2 below summarizes the various technologies used in the treatment of AMD.

Table 2. List of the AMD treatment methods [12,66].

Active/Passive	Biotic/Abiotic	Treatment Methods	Advantages
Passive	Abiotic	• Anoxic limestone drains • Open limestone channels • Limestone leach beds • Slag leach beds • Diversion wells • Limestone sand • Oxidation channels	• Self-sustaining. • Needs sporadic maintenance. • Very low operating and capital costs.
	Biotic	• Wetlands • Bioreactors	• Applicable at closed mine sites with low AMD flows with potentially lower costs and longer-term sustainability.
Active	Abiotic	• Use of alkaline chemicals such as calcium, sodium, calcium hydroxide (slaked lime), calcium carbonate (limestone), calcium oxide (lime or quick/burnt lime), sodium carbonate (soda ash), sodium hydroxide (caustic soda), magnesium hydroxide, and ammonium hydroxide	• Waste by-products can be used to treat other wastes. • Can be applied to all AMD flow rates. • Fast and effective. • Produces good quality effluent. • Cost recovery via the sale of resulting water, metals, and by-products. • Has a lower cost in the handling and disposal of generated sludge. • Suitable for operational mine sites where the necessary resources are readily available.
	Biotic	• Off-line sulfidogenic bioreactors	• Can be applied to all AMD flow rates. • Fast and effective. • Produces good quality effluent. • Potential for cost recovery via the sale of resulting water, metals, and by-products. • Lower cost in handling and disposal of generated sludge.

5. AMD Impact

High quantities of dissolved metals and acid constitute AMD, which is extremely harmful to groundwater, streams, and rivers. AMD also damages ecosystems, corrodes infrastructure, and poses a number of environmental challenges for aquatic life. This often results in contaminated water supplies to areas where freshwater is not easily accessible [73].

For AMD pollutants to affect humans, they need to be exposed and several AMD pollutants are dangerous to humans [74]. According to Orlović-Leko et al. [75], heavy metals have an adverse effect on both people and the environment, and they can linger for a very long time in natural ecosystems where they build up at higher and higher levels of the food chain. This will lead to acute and chronic diseases where metabolic functioning is disrupted by accumulation of heavy metals in vital organs and glands [65].

The water from AMD inflicts terrible damage since it starts out clear and quickly turns brilliant orange when iron oxides and hydroxides precipitate due to the high acidity levels. By becoming embedded on the river, stream, or ocean bed, this fine precipitate, known as ochre, cements substrates that serve as a food supply for benthic creatures, which eventually go extinct [74], affecting the higher levels of the food chain. Because of these indirect effects, AMD still has an effect on people and wildlife further downstream even if the acidity and heavy metals are reduced.

Heavy metals also contaminate soil which poses serious environmental issues where plant growth is affected by oxidative stress [8]. According to Li et al. [76], this causes cellular damage and disturbs homeostasis, which affects the physiology and morphology of plants. Calcium and magnesium are unavailable to plants as well as nitrogen, phosphorus, and potassium when the pH of the soil is low. At a low pH, soil particles also release aluminium, iron, and manganese, enhancing their toxicity. Furthermore, low soil pH affects how well plants use nutrients, establish roots, and tolerate drought by reducing the activity of soil organisms that break down organic materials. According to Jiao et al. [9], heavy metals are accumulated by aquatic creatures like fish both directly from tainted water and indirectly through the food chain. Since they are highly persistent and poisonous at trace levels, cadmium, copper, lead, and zinc have the potential to cause severe oxidative stress in aquatic organisms [77]. While chronic exposure can cause mortality or stunted growth, limited reproduction, malformations, or lesions, acute exposure to them can directly kill organisms. As a result, aquatic organisms' typical physiological processes—including ion exchange with the water and respiration—are influenced by the pH of the water.

6. Current Treatment Technologies and Resource Recovery

Many studies and investigations have been conducted by academics to treat AMD. The following treatment techniques are frequently applied: neutralization [78], precipitation [79], and sedimentation [80]; nevertheless, additional techniques such anaerobic bioreactors [81], sorption [82], coagulation [83], flocculation [84], and crystallization [85] may also be employed. Although these effluents are typically treated, these techniques may not be sufficient to treat the effluent characteristics to fulfil standards for discharge and/or reuse; high levels of chemical product consumption can produce significant amounts of sludge polluted with metal [9]. The membrane separation process has emerged in the treatment of AMD with an ability to have salts and metals retained from aqueous media using membrane separation methods, particularly reverse osmosis (RO), membrane distillation (MD), forward osmosis (FO), and nanofiltration (NF). The NF process is a third option between RO and UF that can retain multivalent ions and dissolved compounds with molar masses between 200 and 1000 g/mol [86]. Numerous studies have demonstrated the effectiveness of NF as a secondary or tertiary treatment system [87,88]. This is due to its low consumption of power, high efficiency, and ease of operation. Meanwhile, RO technology has been reported as a promising AMD option for producing high-quality water while minimizing the discharge.

For example, Andalaf et al. [89] developed an AMD treatment process to treat and predict the behaviour of AMD; an NF pilot-scale system with two different membranes (NF270 and NF90, France) was used. All ions were rejected with a high rejection rate (100%); however, fouling was found at a water recovery rate of 75%. Wadekar et al. [90] compared ceramic and polymeric nanofiltration membranes in the treatment of abandoned coal mine drainage. The AMD was sampled from a site in Pennsylvania and was treated with NF (ceramic and polymeric NF270 membranes, pressure: 35 bar) which was pretreated with aeration and microfiltration. Over 96% of multivalent ions were rejected by NF270. Approximately 55 to 67% of ceramic membrane rejections resulted from its use. Membrane fouling occurred with a water recovery rate of 75%. Masindi et al. [91] looked into recovering drinking water from acid mine drainage from a South African coal mine, and they found that a reverse osmosis device can successfully prepare the water. The drinking water produced by this procedure had a pH of about 6.5 and a metal removal rate of about 100%, which met the SANS 241 criteria for drinking water quality. With the use of forward osmosis, acid mine drainage can be concentrated in a way that promotes the growth of enrichment sludges and the subsequent selective metal precipitation. León-Venegas et al. [92] studies the potential for water and metal recovery from acid mine drainage from the Iberian Pyrite Belt, Southwestern Spain, by combining hybrid membrane processes with selective metal precipitation. Forward osmosis, reverse osmosis, and osmotically assisted reverse osmosis mixed with selective metal precipitation was used to treat AMD, obtaining high water recovery and an enriched metal sludge. They reported that two steps of FO using draw solutions based on sodium chloride could recover about 80% of the water from AMD. Moreover, selective metal precipitation can be used to produce sludges rich in Fe, Al, Cu, Zn, and Mn from AMD. Asif et al. [93] showed the efficacy of a direct contact (DC)-MD system for the treatment of AMD. They found that the DCMD achieved 100% removal of AMD and produced high-quality effluent. However, the permeate flux was reduced by 76% due to membrane fouling induced by membrane scaling, and this flux reduction was based on the metal content as well as the presence of bulk organics in the feed water.

This section reviews some examples of South African AMD treatment projects that are either commercially developed and in operation, in the pilot stage, or under evaluation. The alkali–barium–calcium (ABC) method, developed by the Council for Scientific and Industrial Research (CSIR) in South Africa, consists of three phases. The first stage is the addition of lime and calcium sulphide to remove metals and acids. The second stage involves treating most of the remaining water with barium carbonate to remove the remaining sulphate as barium sulphate. The barium sulphate and some sludge wastes are reduced in a coal-fired kiln to recover some of the alkaline compounds used for neutralisation as well as barium and calcium, some of which can be recycled back into the treatment process [94–96]. This method is a potentially cost-effective treatment for AMD because of the potential reuse opportunities from recycling [97]. However, it still produces a significant amount of waste sludge that must be disposed of. In addition, this process also involves high capital and operating costs, especially with running a coal-fired kiln.

A possible modification/improvement to the above is the Tshwane University of Technology's magnesium–barium–alkali (MBA) treatment process [98]. This process uses barium hydroxide for two purposes, that is, to precipitate and remove sulphate as barium sulphate and to precipitate and remove magnesium as magnesium hydroxide.

The CSIR recently developed and patented a sustainable AMD treatment technology called Magnesite–Softeners–Reverse-Osmosis–Eutectic (MASRO) Freeze Crystallisation that uses magnesite slurry to neutralise the AMD and precipitate metal hydroxides [99]. The advantage of using magnesite is that most of the gypsum, which usually forms a large portion of AMD sludge if it is first treated with a calcium compound like lime as a neutralising agent, does not precipitate with the metal hydroxide sludge. This allows for the possibility of easily concentrating and treating the metal hydroxide sludge to remove more of the valuable metals. Following this step, a lime slurry is added to the AMD, and this precipitates a gypsum sludge (70% gypsum) containing brucite (i.e., $Mg(OH)_2$). Next, the AMD is treated with a soda ash slurry to recover any residual calcium (65% as calcium carbonate) and magnesium and finally, the remaining water is treated using RO to improve its quality so it can be fit for human consumption. Masindi et al. [100] published results of a pilot plant of the MASROE process which was designed and built to treat 20 kL of AMD per day. They also calculated that the direct field costs to treat the AMD by this process was ZAR 65.60/kL.

However, if the by-products such as metal hydroxides could be treated to produce iron pigment, gypsum, and lime which could be purified to be sellable, then Masindi et al. [100] calculated that the sale of these products could perhaps yield an overall saving of approximately ZAR 9.00/kL (it is also noted that the possible sale of treated water did not appear to be accounted for and this may further increase these potential savings). Thus, presuming the above, the direct field costs could possibly be reduced to approximately ZAR 56.60/kL (i.e., a ±14% cost saving). However, it must be noted that AMD transportation costs were deemed negligible as it was assumed that the plant would be close to the AMD source. The treatment costs were also calculated presuming that the plant would operate for most of the year (i.e., 95% of the time, 24 h/day) and that electrical power and cleaning water were the only required utilities. The treatment costs also excluded any operational labour. In addition, the study did not account for any costs of processing the sludges to prepare iron pigments, gypsum, or calcium carbonate of marketable quality. Therefore, it is possible that the costs not accounted for could lead to less of a cost saving than predicted. Until further research is conducted, these additional costs might potentially increase the overall treatment costs to greater than ZAR 65.60/kL. However, one of the key findings from this research was the fact that setting up a process to effectively claim by-products for potential resale is possible if the process is planned and correctly constructed. And, the removal and resale of valuable components of the sludge such as metals, that would otherwise be potentially toxic if left in the sludge, leads to better environmental protection and potentially has less requirements for newly mined resources.

The SAVMIN process, developed by Mintek, involves five stages of AMD treatment. Firstly, lime is added to precipitate metals [101,102]. Secondly, using gypsum seed, all the remaining gypsum is removed. Thirdly, aluminium hydroxide is added to the remaining AMD water, and this produces ettringite (a calcium–aluminium sulphate mineral) which removes any remaining dissolved calcium and sulphate. Fourthly, the ettringite is removed and remixed with sulphuric acid which causes decomposition into aluminium hydroxide (which is recycled back into the process) and gypsum (some of which is recycled back into the process). Finally, in the fifth stage, the remaining water from stage four is treated with carbon dioxide to lower the pH and remove calcite by precipitation. The advantages of the SAVMIN process include high-quality by-products such as metal hydroxides, gypsum, and calcite which can potentially be resold to enhance the economic feasibility of this treatment [17,101]. However, again, it too produces significant amounts of waste sludge that would need to be disposed of.

A process developed in the United States of America is the slurry precipitation and recycling reverse osmosis (SPARRO) process. SPARRO uses membrane desalination to treat AMD and produce water at variable recoveries depending on, for instance, its chemical properties. Membrane fouling is and remains a challenge with this process and thus developing membranes to improve their performance may increase the economic feasibility of SPARRO to treat AMD [72].

Another process developed in South Africa is gypsum–continuous ion exchange (GYP-CIX). It is a continuous fluidised bed ion exchange process designed to remove calcium and sulphate from gypsum-saturated waters such as AMD [103]. During the first stage of the GYP-CIX process, cations can be removed from the AMD by cation exchange resins. After cation removal, anions are then removed by anion exchange resins. When required, the anion exchange resin is regenerated by lime while the cation exchange resin is regenerated by sulphuric acid. The advantages of this process include calcium and sulphate precipitates of relatively high quality that have the potential to be reused [104]. The use of inexpensive chemicals and efficient water recoveries are further benefits. However, as would seem to be customary, a significant amount of sludge is generated during the renewal of the ion exchange resins, and this typically necessitates an expensive disposal method.

THIOPAQ is a biotechnological approach to AMD treatment [105,106]. It involves two stages, the first being the addition of hydrogen gas to the AMD to produce sulphide from sulphate which precipitates out metal sulphides. Any excess hydrogen sulphide produced is oxidised to elemental sulphur in the second stage using sulphide-oxidising bacteria. The hydrogen gas used in the first stage of the THIOPAQ process is generated using ethanol and butanol. Recently, however, these chemicals have become quite expensive which has reduced the attractiveness of this approach.

The Rhodes BioSURE process, developed at Rhodes University in South Africa, is a biological treatment used to remove acid from AMD using waste such as sewage sludge or organic wastes [107–109]. While using these wastes will make treatment cheaper, they are also a potentially limiting reagent if a sufficient stock of them is not available. However, the advantages are re-use of sewage sludge or organic waste which would result in lower landfill loads and costs. Interestingly, this process was used by the East Rand Water Care Company in Grootvlei [72].

High-pressure reverse osmosis (HiPRO) was developed by Nafasi Water, then known as Aveng Water, in South Africa and applied at the eMalahleni treatment plant. The recovery of water was generally very good while brine and solid waste were also produced [110]. By-products from the solid waste that could potentially be sold were various purities of calcium sulphate and metal sulphates. However, the main challenge was the treatment/disposal of the remaining waste sludge and brine.

Luo et al. [111] provide details of a treatment that they successfully used to recover metals and produce hydrogen gas using microbial electrolysis cells to treat AMD. This electrolysis technology is one of the more promising methods to be developed. Through microbiologically assisted electrolysis, these researchers were able to remove copper, nickel, and iron from simulated AMD solutions while concurrently producing hydrogen to potentially offset some of the energy inputs during treatment.

Nleya et al. [64] published their research about the possible production of sulphuric acid from AMD. They summarised that, although not currently economically profitable, methods such as freeze crystallisation and acid retardation may potentially be the most promising technologies for acid recovery. These authors also indicated that the investigation and possible use of lower cost energy sources would assist to make these alternative treatments of AMD more economically viable.

The research details of staged electrochemical treatment in a laboratory to neutralise the pH and remove metals from AMD have recently been published by Brewster et al. [66]. Briefly, an electrochemical system was set up and a current applied between a cathode and anode. This caused the pH of the cathodic solution to increase while the anolyte solution pH decreased. The anions, including sulphate, were drawn across a membrane from the cathodic solution into the anolyte solution because of the decrease in pH. The increasing pH in the cathodic solution caused the dissolved metals to concurrently precipitate out at specific pH endpoints. The results indicate that metals like aluminium, iron, manganese, zinc, nickel, lead, and others were successfully removed. The advantages of this approach include the use of virtually no chemicals and the production of lower sludge volumes. Other advantages, as indicated by these authors, are the co-precipitation of most AMD metals in a controllable manner which will assist with possible recovery and recycling and the possible recovery and sale of sulphuric acid. The significant disadvantages include high initial capital costs and membrane fouling. Figure 1 summarises the current technologies used for AMD treatment.

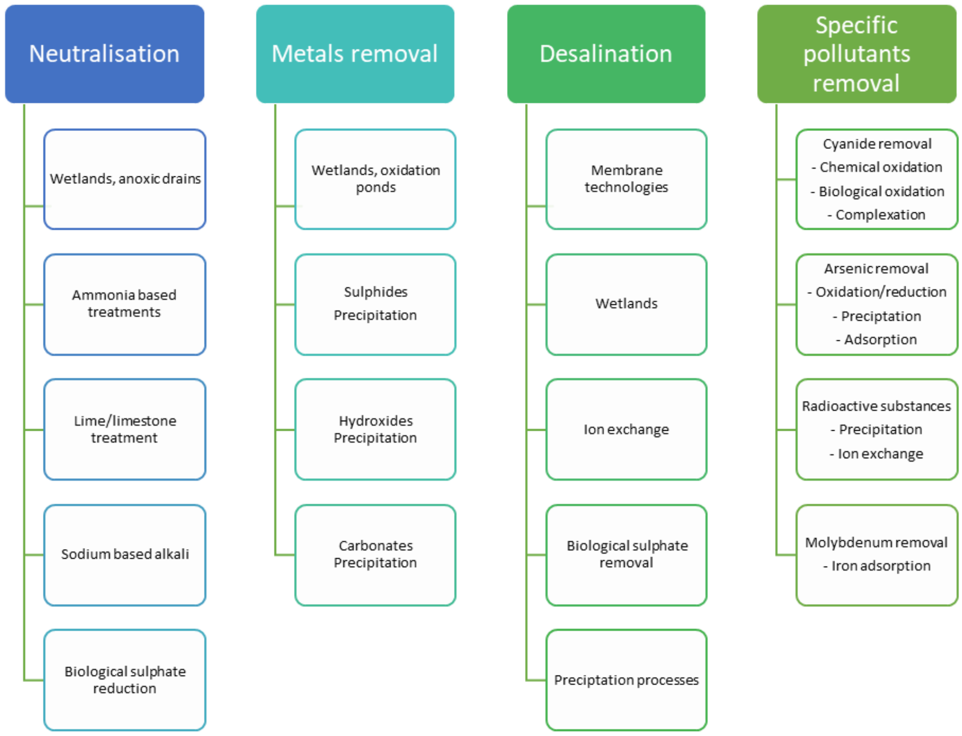

Figure 1. Current technologies used for AMD treatment.

7. Conclusions and Future Prospects

AMD, in South Africa and the rest of the world, is the cause of serious environmental and social concern and requires urgent attention. There are several active, passive, abiotic, and biotic treatments that have been investigated and, in some instances, implemented. The most widely used AMD treatment throughout the world is active chemical neutralisation which has been modified recently to be more efficient by continuously recycling most of the sludge, using fewer neutralisation chemicals, and producing less waste sludge. However, chemical neutralisation remains very expensive and produces significant amounts of potentially toxic waste. Various active and passive treatments have been investigated and generally look promising at the lab bench scale and a few at pilot plant scale. However, none of the recent techniques to treat large volumes of AMD have been implemented and proven to work at full scale for an extended period. Thus, the next step remains full-scale implementation and successful, long-term operation. Often, however, constraints like costly initial capital expenses are a factor that needs to be overcome. Besides these challenges, there is the real potential to produce by-products that could be sold to offset some of the initial capital costs and the on-going operational treatment costs. Some new technologies have successfully reclaimed metals, sulphur-based products such as gypsum and sulphuric acid, and other alkali chemicals including calcium carbonate. These alternative treatments offer several advantages including the requirement for less new materials because of recycling and a reduction in the amount and treatment and disposal costs of potentially toxic sludge waste. However, the emphasis must now be on full-scale implementation of the most promising techniques that recover and sell viable, marketable by-products to significantly offset the cost of traditional AMD treatment technologies and promote the circular economy. Future research is needed to increase the efficiency of all the methods used, with a focus on reducing the generation of secondary pollutants and recovering valuable resources.

Author Contributions: Conceptualization, J.B. and N.R.; formal analysis, J.B. and N.R.; investigation, J.B., D.R.-M., and N.R.; resources, R.M.; writing—original draft preparation, J.B. and N.R.; writing—review and editing, J.B., R.M., D.R.-M., and N.R.; visualization, J.B. and N.R.; supervision,

R.M. and J.B.; project administration, R.M. and J.B. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Data Availability Statement: No new data were created or analysed in this study. Data sharing is not applicable to this article.

Acknowledgments: The authors acknowledge the support received for this study from Council for Scientific and Industrial Research (CSIR).

Conflicts of Interest: The authors declare no conflict of interest.

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