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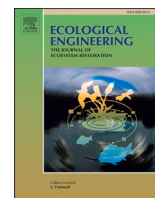
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Review

Challenges and avenues for acid mine drainage treatment, beneficiation, and valorisation in circular economy: A review

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ABSTRACT

Mining activities are notorious for their environmental impact, with acid mine drainage (AMD) being among the most significant issues. Specifically, AMD has recently been a topical issue of prime concern, primarily due to the magnitude of its environmental, ecotoxicological, and socioeconomic impacts. AMD originates from both active and abandoned mines (primarily gold and coal) and is encountered in Canada, China, Russia, South Africa, USA, and other countries with strong mining industry. Owing to its acidity, AMD contains elevated levels of dissolved (toxic) metals, metalloids, rare-earth elements, radionuclides, and sulfates. Practical and cost-effective solutions to prevent its formation are still pending, while for its treatment active (driven by frequent input of chemicals and energy) or passive (based on oxidation/reduction) technologies are typically employed with the first being more efficient in contaminants removal, however, at the expense of process complexity, cost, and materials and energy consumption. More recently, and under the circular economy concept, hybrid (combination of active and passive technologies) and particularly integrated (sequential or stepwise treatment) systems have been explored for AMD beneficiation and valorisation. These systems are costly to install and operate but are cleaner production systems since they can effectively prevent pollution and can be used for closed-loop and sustainable AMD management (e.g., zero liquid discharge (ZLD) systems). Herein, the body of knowledge on AMD treatment, beneficiation (metals/minerals recovery), valorisation (water reclamation), and life cycle assessment (LCA) is comprehensively reviewed and discussed, with focus placed on circular economy. Future research directions to introduce reuse, recycle, and resource recovery paradigms in wastewater treatment and to inspire innovation in valorising this toxic and hazardous effluent are also provided. Overall, AMD beneficiation and valorisation appears promising since the reclaimed water and the recovered minerals/metals could offset treatment costs and environmental impacts. However, the main challenges include high-cost, complexity, co-contamination in the recovered minerals, and the generation of a highly heterogeneous and mineralised sludge.

1. Introduction

The omnipresence of valuable mineral resources plays a pivotal role in economic development, with coal and gold mining being historically perceived as the backbone of the economy and the main driver of

economic growth in low- and middle-income countries (LMICs) (Berenbaum et al., 2019; Haldar, 2018; Masindi et al., 2021). Due to its high calorific value, coal has been mined for more than 140 years for power generation (Andrić et al., 2015), while gold has been mined and used (e.g., ornamental objects and jewellery) since ancient times (Neingo and Tholana, 2016). Revenue generated from mining also leads

Abbreviations: LMICs, Low- and middle-income countries; Fig, Figure; DAS, Dispersed alkaline substrate; LPD, Liters per day; R&D, Research and development; ALSX, Acid leaching-solvent extraction; BOD, Biochemical oxygen demand.

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Nomenclature*Abbreviations/acronyms*

ABC	Alkali-based-calcium
AGP	Acid generation potential
AHCL	Advanced hardpan cover liner
ALSX	Acid leaching-solvent extraction
AMD	Acid mine drainage
ANP	Acid neutralization potential
ARD	Acid rock drainage
BOD	Biochemical oxygen demand
BOF SRO	Basic oxygen furnace soda ash and reverse osmosis
CIF	Continuous ionic filtration
CSIR	Council for Scientific and Industrial Research
DAS	Dispersed alkaline substrate
DC-MFC	Dual-chamber microbial fuel cell
DCMD	Direct contact membrane distillation
EARTH	Environmental and remedial technology holdings
EC	Electrical conductivity
Eq	Equation
Fig	Figure
FGR-DAF	Flocs generator reactor - dissolved air flotation
GARD	Global acid mine drainage
HiPRO	High Recovery Precipitating Reverse Osmosis
LC	Lethal concentration
LCA	Life cycle assessment
LD	Lethal dosage
LMIC	Low- and middle-income countries
LPD	Liters per day
MASRO	Magnesite softening reverse osmosis
MASROE	Magnesite softening reverse osmosis and eutectic freeze
MD	Membrane distillation
MF	Microfiltration
MIP	Mixed-integer programming
MWW	Municipal wastewater
NF	Nanofiltration
O&M	Operation and maintenance
PCB	Printed circuit board
PHREEQC	pH-REdox-EQuilibrium in C language
PRB	Permeable reactive barriers
R&D	Research and development
RO	Reverse osmosis
TDS	Total dissolved solids
TUT MBA	Tshwane University of Technology Magnesium Barium Alkali
ZLD	Zero liquid discharge
UF	Ultrafiltration

Symbols/notations/chemical formulas

(Fe,Ni) ₉ S ₈	Pentlandite
(Fe, Zn)S	Sphalerite
Al	Aluminum
As	Arsenic
Ba	Barium
BaCO ₃	Barium carbonate
Cr	Chromium
Ca(OH) ₂	Hydrated lime
Ca	Calcium

CaCO ₃	Limestone
Cd	Cadmium
CaMg(CO ₃) ₂	Dolomite
CaO	Quicklime
Cl	Chloride
Cu	Copper
Cu ₂ S	Villamaninite
CuFeS ₂	Chalcopyrite
CuS	Covellite
Fe	Iron
FeAsS	Arsenopyrite
FeS ₂	Pyrite
FexSx	Iron sulfides
Fe ²⁺	Ferrous iron
Fe ³⁺	Ferric iron
H ⁺	Hydrogen ion
H ₂	Hydrogen
H ₂ O	Water
H ₂ S	Hydrogen sulfide
H ₂ SO ₄	Sulfuric acid
Mg(HCO ₃) ₂	Magnesium bicarbonate
Mg(OH) ₂	Brucite
Mg	Magnesium
MgO	Periclase
Mn	Manganese
Mo	Molybdenum
MoS ₂	Molybdenite
Na	Sodium
Na ₂ CO ₃	Soda ash
NH ₃	Anhydrous ammonia
NH ₄ OH	Ammonium hydroxide
NaHS	Sodium hydrosulfide
NaOH	Sodium hydroxide (caustic soda)
Ni	Nickel
NiS	Millerite
O ₂	Oxygen
P	Phosphorus
Pb	Lead
PbS	Galena
REE	Rare earth elements
S ²⁻	Sulfide
Sb	Antimony
SO ₄ ²⁻	Sulfate
Sr	Strontium
Y	Yttrium
Zn	Zinc

Units

CO ₂ eq	Carbon dioxide equivalent
D	Day
dam ³	Cubic decametre
h	Hour
L	Litre
m ³	Cubic meter
M	Mega
kg	Kilogram
t	Tonne (metric)

to job creation, thus contributing, in the short-term at least, to social welfare (Neingo and Tholana, 2016). However, this often comes at the expense of the environment, since mining activities are notorious for their detrimental environmental impact (Omotehinse and Ako, 2019). Furthermore, by-products and waste materials, such as overburden and tailings, are also generated, which typically remain on-site long after mining activities have ceased, unless rehabilitated. Without suitable management practices, the overburden and tailings, along with the exposed mined areas and the tunnels and shafts of abandoned and/or active mines, will react with water forming basic, circumneutral, or, more often, acidic leachates (Park et al., 2019). The latter is popularly known as acid and metalliferous drainage, or acid mine drainage (AMD), or acid rock drainage (ARD) (Masindi et al., 2021; Nordstrom et al., 2015b; Tutu et al., 2008).

AMD greatly affects the receiving environment, mainly by altering the ambient pH and the dissolved concentrations of different chemical species (Masindi et al., 2017a). The minerals contained in AMD can also precipitate at the bottom of receiving waterbodies, such as streams and rivers, and affect benthic organisms (Hogsden and Harding, 2012). Of primary concern is the existence of hazardous and toxic chemical species in AMD, such as arsenic (As), chromium (Cr), iron (Fe), aluminium (Al), copper (Cu), zinc (Zn), lead (Pb), molybdenum (Mo), and nickel (Ni) (Masindi and Tekere, 2020). These contaminants can cause ecotoxicological, carcinogenic, mutagenic, and teratogenic effects on exposure (Talukdar et al., 2017). Apart from negatively affecting aquatic ecosystems, AMD also affects the quality of natural waterbodies that are intended for human consumption or for irrigation (Zhu et al., 2020).

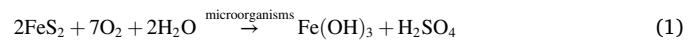
Therefore, preventing the formation of AMD, or effectively treating it, can safeguard human health and the environment. AMD treatment is typically based on active (driven by frequent input of chemicals, energy, and equipment) or passive (based on oxidation or reduction) technologies. However, these technologies have variable efficacies in contaminants removal and also produce sludge and/or brines which can cause secondary contamination if not properly managed (Kefeni et al., 2017b). Hybrid and integrated technologies have also been recently introduced for AMD management. These technologies can be used for AMD's beneficiation (metals/minerals recovery) and/or valorisation (water reclamation) (Masindi et al., 2019a), since AMD often contains high concentrations of Fe and sulfate (SO_4^{2-}) (Akinwekomi et al., 2020), along rare earth elements (REE) and yttrium (Y) (Ayora et al., 2016).

However, even though industrial AMD treatment systems have been routinely used for more than two decades (Jarvis and Younger, 1999), these often suffer from subpar performance and failures while beneficiation and valorisation opportunities have not been fully explored. To this end, here the state-of-the-art on AMD generation, abatement, and treatment is distilled and critically reviewed. This paper contains eight sections. Section 1 introduces the reader to the topic and section 2 discusses AMD's formation pathways and its environmental and ecotoxicological impacts. Section 3 summarises AMD abatement techniques, while section 4 describes the mechanisms underlying contaminants removal and minerals synthesis and recovery. Section 5 provides insight on AMD treatment methods and section 6 summarises the body of knowledge on life cycle assessment (LCA) on AMD treatment and prevention. In section 7, avenues for AMD valorisation and beneficiation are discussed, and finally, in section 8, concluding remarks and future research directions are provided.

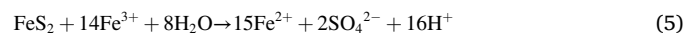
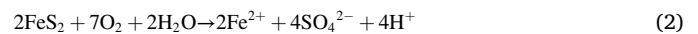
2. AMD formation pathways and toxicity

Although pyrite (FeS_2) and arsenopyrite (FeAsS) are mainly responsible for AMD formation, the weathering of other sulfide-rich or sulfide-bearing minerals such iron sulfides (Fe_xS_x), pentlandite ((Fe, Ni) $_9\text{S}_8$), chalcopyrite (CuFeS_2), villamaninite (Cu_2S), covellite (CuS), molybdenite (MoS_2), sphalerite ((Fe, Zn)S), millerite (NiS), and Galena (PbS) also contribute to AMD formation (Simate and Ndlovu, 2014; Tabelin et al., 2017). These minerals are typically encountered in

organic rich (reducing) sediments (e.g., in coal deposits) (Akinwekomi et al., 2017; Tabelin et al., 2017), or rock altered by sulfur-rich hydrothermal fluids e.g., volcanogenic sulfide ore producing metals such as copper (Simate and Ndlovu, 2014), gold (Masindi et al., 2015a) and zinc (Galjak et al., 2020). When they become exposed to the atmosphere, typically through mining activities (e.g., surface or deep mines, waste piles, and tailings) they oxidize and AMD is formed, with iron sulfide minerals (typically pyrite) being the main culprit behind AMD formation and other sulfide minerals only contributing to a limited extent (Nordstrom et al., 2015b). The main mechanism for AMD formation from pyrite in the presence of air (oxygen – O_2), water (H_2O), and microorganisms (catalyst), is shown in Eq. (1) (Masindi et al., 2015a):



Specifically, the reaction shown in eq. (1) is catalysed by certain microorganisms, particularly Fe-based (e.g., acidithiobacillus (acidic) and thiobacillus (basic pH)) and S-based bacteria (e.g., sulfobolus (acidic) and desulfovibrio (circum-neutral-basic)) (Nordstrom et al., 2015b; Tabelin et al., 2017). The inter-dependent chemical reactions that govern AMD formation are described in Eqs. (2) to (5) (Simate and Ndlovu, 2014).



Where, Eq. (2) shows the oxidation of the sulfide (S^{2-}), when exposed to air (O_2) and water (H_2O), to sulfate (SO_4^{2-}), acidity (H^+), and ferrous iron (Fe^{2+}). The latter is then oxidised to ferric iron (Fe^{3+}) and hydrogen (H_2) (Eq. (3)). In Eq. (4) the hydrolysis of Fe^{3+} , in the presence of H_2O , is shown, which leads to the formation of ferric oxy-hydroxide minerals (e.g., $4\text{Fe}(\text{OH})_3$) in sediments, popularly known as 'yellow-boys' (Fig. 1), and H^+ . Finally, Eq. (5) shows how the oxidation of additional FeS_2 by Fe^{3+} occurs. These reactions usually occur spontaneously and are catalysed by microorganisms that obtain energy by oxidation reactions (Eq. (1)). The net effect of these reactions is the release of H^+ , which lowers the pH and maintains Fe^{3+} and metals/minerals solubility in the AMD matrix (Simate and Ndlovu, 2014). These reactions affect the physicochemical characteristics of the receiving water matrices, as well as of the bottom sediments, leading to changes in the pH, composition, and colour, among others (Fig. 1).

As soon as AMD is formed, it finds its way to receiving waterbodies mainly through: i) flooding of mines (following the cessation of groundwater pumping when mines are abandoned), ii) open surface mining activities, iii) seepage from mine residue deposits, and iv) mine water losses (Johnson and Hallberg, 2005; Park et al., 2019). To reduce the flow of water into mining areas and curtail AMD releases to the environment various abatement techniques can be employed, such as mine waste storage and improved mine closure practices (Parbhakar-Fox and Lottermoser, 2015), as discussed in section 3.

2.1. Physicochemical characteristics

AMD's physicochemical characteristics are typically distinctive at a spatial level, since these are traced back to the host minerals and underlying geology, which, along with the local water quality, climatic conditions, and bacterial concentrations will dictate the final type of mine drainage to be formed (Simate and Ndlovu, 2014; Zhu et al., 2020).

2.1.1. Physical and aesthetic properties

Colour is a distinctive physical property of AMD and this can range from blackish to greenish to bluish to reddish and can even be colourless,



Fig. 1. AMD from underground shafts in Witwatersrand basin, South Africa. In (A) and (B) the effluent is initially colourless with the yellow-boys being visible, and in (C) after oxidation by atmospheric air the effluent gradually turns red.

principally depending on the dissolved chemical species (Warren, 2011). Typically, AMD is reddish in colour, which is mainly linked to the oxidation of Fe^{2+} to Fe^{3+} (Tutu et al., 2008). The greenish colour is linked to Cu presence (Simate and Ndlovu, 2014), bluish colour is attributed to Fe^{2+} , while white or black colour suggest the predominance of Al^{3+} or manganese (Mn), respectively (Gooneratne et al., 2011). Furthermore, in streams with pH in the range 5 to 6.5, ferric iron-rich sediments can be formed, which are orange-yellow in colour (the yellow-boys shown Fig. 1) (Amos et al., 2015; Zhao et al., 2012). Moreover, if AMD reaches freshwater bodies, SO_4^{2-} will impart a salty or bitter taste to water, hardness and total dissolved solids (TDS) will also be affected leading to unpleasant taste, while skin dryness on exposure, scum in water, and high soap consumption could be also observed (EPA, 2017; SABS, 2015; WHO, 2017).

2.1.2. Chemical properties

Various classification frameworks for mine drainage have been proposed. For example, the global acid mine drainage (GARD) guide uses the pH and TDS to classify mine drainage as acid, neutral, or saline (Thisani et al., 2020). However, a simpler classification is based solely on the pH, where mine drainage is classified as acid (<6), circumneutral (6 to 9), or basic (>9) (Madzivire et al., 2011; Nordstrom et al., 2015a). The pH influences the chemical composition of the mine drainage, with metals and sulfate predominating the acidic pH (AMD) and base metals (nonferrous) the basic pH (Langmuir, 1997). Circumneutral drainage can contain metals, oxyanions (e.g., sulfate and metalloids), and base metals (Nordstrom et al., 2015b; Spellman et al., 2020b). Metals concentration in basic and circumneutral drainage are much lower than in AMD, since metals precipitate with increasing pH values. This also suggests that metals can be precipitated and removed from AMD by increasing its pH (Masindi et al., 2018b). For context, the mean chemical concentrations of acid and circumneutral drainage are listed in Table 1.

As shown in Table 1, beneficiation opportunities, i.e., metals/minerals recovery, are available for both effluents and particularly for AMD. However, radionuclides might also be present in drainage that originates from gold mining, but these are rarely found in coal mine drainage (Tutu et al., 2008). As shown in Table 1, circumneutral drainage has elevated

Table 1

The mean chemical composition of acid (AMD) and circumneutral mine drainage.

pH/ element	Types of mine drainage		References
	AMD	Circumneutral	
mg L ⁻¹ (ppm)			
pH	2 - 4	5.5 - 9.5	(Madzivire et al., 2011; Madzivire et al., 2010; Maree et al., 2004a; Maree et al., 2004c; Maree et al., 1999; Masindi et al., 2017a; Masindi et al., 2021; Masindi et al., 2016; Masindi et al., 2017b; Masindi et al., 2019a; Park et al., 2019; Pope and Trumm, 2015)
Al	75 - 500	0.016	
Fe	500 - 8000	0.074	
Mn	10 - 100	2.5	
SO_4^{2-}	1000 - 80000	4600	
Cu	0.4 - 10	-	
Ni	0.21 - 10	0.21	
Pb	0.10 - 10	-	
Zn	0.1 - 20	0.16	
Ca	50 - 450	540	
Mg	50 - 400	860	

levels calcium (Ca) and magnesium (Mg), primarily due to the dissolution of carbonate minerals in the host rock during its formation, along with SO_4^{2-} . Due to the elevated levels of Mg and Ca, water hardness is high. As such, the elements that could possibly be recovered from circumneutral drainage include SO_4^{2-} and possibly Mg and Ca, depending on their concentration, typically by means of softening techniques such as soda ash treatment (Masindi et al., 2019a). On the other hand, AMD has a greater potential for minerals recovery and synthesis, due to elevated levels of Fe, Al, Mn, and SO_4^{2-} (Table 1). For example, Fe and SO_4^{2-} based minerals can be synthesized and recovered from coal AMD (Akinwekomi et al., 2020; Akinwekomi et al., 2017).

2.2. Environmental and eco-toxicological impacts

Mine drainage, and particularly AMD, is responsible for a number of negative social, economic, and environmental impacts (Chalkley et al., 2019; Netto et al., 2013; Talukdar et al., 2016). Contaminants, and primarily toxic metals contained in AMD can cause teratogenic,

carcinogenic, and mutagenic effects to living organisms on exposure (Balistrieri et al., 2007; Chu et al., 2018; Ivask et al., 2015; Jooste and Thirion, 1999), impacting organisms that reside in different environments i.e., soil, water, and air (Luo et al., 2020; Sarmiento et al., 2011; Waters and Webster-Brown, 2013).

Eco-toxicological studies and assays have reported various conditions that are associated with elements present in AMD (Netto et al., 2013), with toxicity typically measured using the lethal concentration (e.g., LC₁₀, LC₂₀, LC₅₀, and LC₁₀₀) and lethal dosage (e.g. LD₁₀, LD₂₀, LD₅₀, and LD₁₀₀) on pot assays (Netto et al., 2013), aquarium studies (Dutta et al., 2020), and toxicological tests (Talukdar et al., 2016).

Exposure to toxicants contained in AMD can alter plants' metabolic activities, homeostasis, and induce toxic symptoms, as well as destabilise the salt balance (Vardhan et al., 2019; Yadav, 2010). Acidity released from AMD also lowers the pH of receiving ecosystems, which increases the mortality of some organisms such as fish and plants and/or forces species to migrate, if possible (Oberholster et al., 2013). Not only this, but (heavy) metals, metalloids, anions, and even radionuclides in AMD can also bio-accumulate in living organisms and eventually bio-magnify through the food chain causing severe challenges in different trophic levels (Simate and Ndlovu, 2014; Vardhan et al., 2019; Vigneri et al., 2017; Yuan et al., 2019). As such, AMD has also been associated with skin lesions, hyperpigmentation, cancer, upper respiratory, pulmonary, gastrointestinal and cardiovascular failure, along with nerve damage and multiple organ failure (EPA, 2017; SABS, 2015; WHO, 2017). Finally, mine drainage from both active/operating and particularly abandoned/derelict mines threatens groundwater, which is a valuable freshwater resource for drinking and agricultural use (Tomiyama and Igarashi, 2022).

Therefore, to safeguard human health and the environment, there is a pressing need to limit and manage the large AMD quantities produced year-round. To this end, various AMD abatement and treatment techniques have been employed, while more recently AMD valorization/beneficiation has also been proposed to reduce treatment cost and environmental impacts.

3. AMD prevention and abatement

Several techniques have been proposed to prevent the formation of AMD, focusing both on operating and abandoned mine sites. Their primary function is to limit the exposure of mine areas and tailings to oxygen, water, and/or microorganisms (Eq. 1) (Hughes and Gray, 2011; Kefeni et al., 2017b; Park et al., 2019; Sahoo et al., 2013). Specifically, over the past two decades AMD formation has been prevented through the elimination of at least one of those factors, mainly using the techniques that are discussed below.

3.1. Utilisation of covering materials

This technique makes use of waterproof materials, such as clays (e.g., bentonite) or plastics, to cover the exposed areas/tailings and prevent their oxidation. To improve waterproofing, layers of different materials can be used, and by doing so not only water but also oxygen ingress can be limited (Kefeni et al., 2017b; Pozo-Antonio et al., 2014; Sahoo et al., 2013). However, cover materials are prone to failures. They can react with the encapsulated materials, or their leachates, and are also exposed to the environment, which leads to their degradation and reduces their waterproofing effectiveness. As such, thick and/or multiple layers of covering materials are required to ensure the sealing of the covered material (AMD), which increases cost and imposes on the sustainability of this technique (Park et al., 2019; Zipper and Skousen, 2014).

3.2. Stabilization using alkaline materials

This approach involves the addition of alkaline materials to tailings,

voids, and other geological settings that are conducive for the formation of AMD. These materials hinder the production of acidic and metalliferous drainage since they raise the pH and lead to the precipitation of metals. In most cases, brucite (Mg(OH)₂), periclase (MgO), quicklime/hydrated lime (CaO/Ca(OH)₂), limestone (CaCO₃), dolomite (CaMg(CO₃)₂), and soda ash (Na₂CO₃) are used (Skousen, 2014; Tripathy, 2014; Watten et al., 2005). The use of fly ash (Gitari et al., 2008), alkaline tailings (Kastyuchik et al., 2016; Masindi, 2016), and alkaline waste (Ouakibi et al., 2013) has also been proposed. This technique is simple to apply and effective for AMD abatement; however, its main drawback is that the neutralization potential reduces over time, hence, in the long-term AMD will form (Tripathy, 2014). Static tests for acid generation potential (AGP) and acid neutralization potential (ANP) are typically carried out to overcome this problem (Skousen et al., 2019; Xenidis et al., 2002). Geochemical modelling (e.g., the pH-REdox-EQuilibrium (in C language) (PHREEQC)) has also been explored to model the time-related generation of AMD from mine wastes (Simunika et al., 2013) and its leaching and attenuation reactions (Hanna et al., 2016).

3.3. Passivation or microencapsulation

Hydrophobic coating materials can be used to prevent the reactive mineral fractions from oxidizing. Organic materials and their derivatives, such as DETA, sodium oleate, phospholipids, and humic substances, are typically used towards that end, owing to their high hydrophobicity (Moodley et al., 2018; Park et al., 2019). However, similarly to covering materials, exposure to environmental conditions, substances emitted from the encapsulated material, along with (micro) organisms can degrade microencapsulation rendering it unable to isolate the covered material from the air, water, and/or bacteria, thus eventually leading to AMD formation (Villain et al., 2013).

3.4. Bactericides application

This abatement technique is based on the use of substances that aim to impede the biological activity of the bacterial communities that are harbored in tailings and mining voids, which catalyze the formation of AMD. The main focus is to limit the bacteria that oxidize sulfur, thus prevent sulfate production and hamper AMD formation (Kim et al., 1999). For example, in a case study in China the application of the bactericides Triclosan, Kathon (isothiazolinones), and sodium dodecyl sulfate in coal AMD effectively inhibited the oxidation of Fe²⁺, thus preventing the build up of acidity and increasing the oxidation-reduction potential (Hu et al., 2020). However, the main drawbacks of bactericides application is that these can be toxic to living organisms, while environmental conditions can limit their activity, thus proper assessment before their application and frequent monitoring of their levels and top-up after their application is required (Park et al., 2019).

3.5. Water ingress control

This concept is based on minimizing the exposure of the mined areas to water, typically by pumping water from voids and opencast trenches. This technique has been widely explored in South Africa but has a limitation of defaulting during heavy rainfall. Furthermore, groundwater intrusion can also be a problem. As a result, water ingress control could be considered costly, due to the rising cost of water pumping which is required to prevent its interaction with exposed minerals. This increases treatment cost and other operation requirements thereof (Tripathy, 2014).

3.6. Backfilling of voids and opencast trenches

Mining activities entail the removal of minerals from different strata and lithologies, thus leaving voids and trenches exposed to water and

air. To prevent AMD formation, voids and trenches can be filled with the extracted topsoil-rocks (overburden material) or preferably with alkaline materials. In this regard, Villain et al. (2013) studied the effects of backfilling and sealing of waste rocks at the Kimheden open-pit mine, northern Sweden and highlighted that this technique is effective and viable to preserve water quality. A mixed-integer programming (MIP) model has been used to optimise the placement of waste rocks into waste dumps (Vaziri et al., 2021). In this regard, Gitari et al. (2008) evaluated, through column studies, the use of coal fly ash for mine void backfilling and it was identified that the pH increases and inorganic contaminants are attenuated. Nonetheless, the exposed mined void volumes are typically very large and therefore their backfilling might not be feasible.

3.7. Abatement techniques for long-term sustainability

Overall, the aforementioned abatement techniques have been found promising in preventing AMD formation. However, they are, in general, expensive, while regular operation and maintenance (O&M) is required, preferably by specialised personnel. In addition, complexity, along with the need for long-term monitoring, typically limits their applicability. Therefore, given the large number of operating and particularly abandoned mines globally (> 6,000 in South Africa, >50,000 in Australia, >2,000 in UK, and >500,000 abandoned or closed mines in USA (Thi-sani et al., 2020), while only in Shanxi province, China 8,780 coal mines have been abandoned during the last two decades (Wang et al., 2021)), and the large volumes of AMD produced year-round (e.g., ~400,000 megalitres (ML or dam³) each year only in the western parts of South Africa (Masindi, 2016)) it is highlighted that robust AMD treatment techniques are also required. To achieve this, different AMD treatment technologies, which are based on distinct mechanisms (section 4) for contaminants removal or even for minerals synthesis and recovery (section 5) have been developed.

4. Mechanisms for contaminants removal and minerals synthesis/recovery

The attenuation and removal of chemical species from AMD can be achieved through different mechanisms, which typically include: i) precipitation, ii) adsorption, iii) filtration, and iv) bio/phyto-remediation. A brief introduction of each mechanism, along with its main strengths and weaknesses, in terms of treatment efficiency and minerals recovery/synthesis, is given below.

4.1. Precipitation

This mechanism relies on the use of alkaline materials, to increase the pH (neutralisation), and/or oxidant agents (oxidative precipitation), to convert metals to insoluble state (e.g., metal hydroxides), hence leading to metals/minerals precipitation, with the prevalently used oxidants and alkaline chemicals being oxides, hydroxides, sulfides, and carbonates (Blais et al., 2008; Lewis, 2010). In oxidant precipitation, metals (e.g., Mn) react with an oxidant agent (e.g., air, oxygen, ozone, chlorine, or sulfur dioxide) to generate a colloidal precipitate (Freitas et al., 2013). On the other hand, the addition of alkaline materials (Table 2) can lead to the precipitation of different chemical species contained in AMD as hydroxides, sulfides, and carbonates (Lewis, 2010), with the underlying mechanism being nucleation, followed by crystallization, and settling (Blais et al., 2008). This also depends on the saturation state of the solution, which can be supersaturated, saturated, or unsaturated, and this will also influence the heterogeneity of the substances that will be formed (Blais et al., 2008; Fu and Wang, 2011). The removal of metals as hydroxides is described in Eq. (5) (Masindi et al., 2018a):



where M is the metal under study and n its oxidation state.

Table 2

Examples of different alkaline materials used for the treatment of AMD.

Mineral	Name	Formula	Reference
Mg-based	Amorphous magnesite	MgCO ₃	(Masindi et al., 2014)
	Periclase	MgO	(Magagane et al., 2019)
	Brucite	Mg(OH) ₂	(Bologo et al., 2012)
	Magnesium bicarbonate	Mg (HCO ₃) ₂	(Akinwekomi et al., 2016)
Ca-based	Limestone	CaCO ₃	(Maree and Du Plessis, 1994)
	lime	CaO	(Geldenhuis et al., 2003)
	Hydrated lime	Ca(OH) ₂	(Mulopo, 2016)
Na-based	Soda ash	Na ₂ CO ₃	(Akinwekomi et al., 2017)
	Sodium hydrosulfide	NaHS	(Wang et al., 2013)
	Caustic soda	NaOH	(Mirbagheri and Hosseini, 2005)
Ammonia	Anhydrous ammonia	NH ₃	(Viadero et al., 2006)
	Ammonium hydroxide	NH ₄ OH	(Maila et al., 2014)
Tailings	Ca, Mg, Na bearing tailings	Ca, Mg, Na	(Kastyuchik et al., 2016)

A wide array of alkaline materials has been examined for AMD treatment and these are summarised in Table 2.

As shown in Table 2, Ca-, Mg-, and Na-based minerals are typically used for AMD neutralisation. Tailings from the mining and processing of ores containing these minerals also exhibit some neutralization capacity, hence, in theory, they can be employed for AMD neutralisation. It should be noted that AMD's chemical composition influences the type of reagents (alkaline materials) that need to be used for the precipitation of chemical species contained in AMD, while the reagents themselves will also influence the composition of the recovered chemical species and their use thereafter (Blais et al., 2008). For example, Silva et al. (2019) reported that Fe³⁺ can be recovered from AMD, which after processing can be used for pigments production. Akinwekomi et al. (2017) suggested the use of Na₂CO₃ and NaOH for the recovery of Fe²⁺, Fe³⁺, and Al³⁺ from AMD, at varying pH gradients, and towards the synthesis of goethite, hematite, and magnetite. Thermally activated (calcined) cryptocrystalline magnesite has been also explored for the selective recovery minerals, using varying pH gradients, from AMD (Masindi et al., 2018b). For context, the optimal pH values for the recovery of different metals/minerals contained in AMD are shown in Table 3, since sequential or stepwise precipitation can be very effective for metals/minerals recovery. However, since there is no distinct line to segregate metal precipitation, the purity of the recovered materials can be affected by co-contamination (Masindi et al., 2018b).

4.2. Adsorption

Adsorption relates to a surface phenomenon in colloidal science and chemistry. It highlights the mechanism at which contaminants migrate in aqueous solutions and are adsorbed onto solid surfaces. As such, adsorption is a solid-water interface process governed by the migration of contaminants contained in a solution (AMD) to the surface of the solid material (adsorbent). Typically, a positively or negatively charged

Table 3

Precipitation of oxy-hydroxides at varying pH gradients using calcined magnesite. Data adapted from (Masindi et al., 2018b).

Element	Optimal pH value
Al	4.5
Fe ³⁺	3.5
Fe ²⁺	8.5
Cu	6.5
Mn	9.5
Ni	8
Pb	6.5
Zn	8

surface is used to remove negatively or positively charged substances, respectively, from aqueous solutions. Apart from physical adsorption, other mechanisms include isomorphous substitution, ion exchange, and complexation (Fig. 2). Adsorption plays a critical role in the attenuation of contaminants from aqueous solutions, including AMD, while the removal mechanism typically depend on the oxidation state since the surface charge of chemical species depend on the pH of the aqueous system (Langmuir, 1997; Shen et al., 2018; Sparks, 1995; Zhao et al., 2019). Adsorption has been widely explored for the removal of chemical species from various aqueous solutions, with its main strengths and weaknesses summarised below (Kalita and Baruah, 2020; Sen Gupta and Bhattacharyya, 2012; Yadav et al., 2019; Zhou et al., 2019):

Adsorption's main advantages include: i) abundant and low-cost adsorbents (e.g., activated carbon) are available; ii) adsorbents can typically be reused for numerous cycles; and iii) the process is simple and does not require a high degree of expertise to be applied. However, the main drawbacks include: i) rapid saturation, which leads to poor performance in highly concentrated solutions such as AMD; ii) high selectivity and affinity hampers its utilization in the decontamination of multi-charged wastewaters such as AMD; iii) this process has been proved to be effective in less concentrated solutions, compared to AMD, and it is mainly used as a polishing technique; and iv) regenerates are usually highly mineralized and heterogeneous, hence making it difficult to recover pure and high quality minerals, while they also require proper handling and disposal, which incur additional costs.

Typically anionic and cationic contaminants are removed through adsorption, making use of materials such as activated carbon (Tran et al., 2020; Tran et al., 2017b), zeolites (Adam et al., 2019; Westholm et al., 2014), clay minerals (Ngulube et al., 2017), and ion exchange resins (Zhu et al., 2017). Different mathematical models and techniques, including adsorption isotherms, kinetics, and thermodynamics, have been employed to identify and describe the underlying mechanisms that govern contaminants removal from aqueous solutions (Tran et al., 2020). The point of zero charge or zeta potential is used to highlight the relationship between the types of pollutants and removal efficacy (Tran et al., 2017a).

For AMD treatment, Zhang (2011) reported the adsorption of Pb^{2+} , Cu^{2+} , and Zn^{2+} from simulated AMD using dairy manure compost. Motsi et al. (2009) explored the adsorption of heavy metals from AMD using natural zeolite, with limited adsorption capacity and regeneration requirements being the main drawbacks. Overall, the key challenge in adsorption is poor selectivity to species of homogenous charge (co-existing ions). Therefore, it appears that adsorption could be used in an AMD treatment train, rather than as a standalone treatment technique, whereas, most likely, adsorption could find little use in circular economy concepts for AMD treatment.

4.3. Filtration

Filtration is a process in which chemical species, solids, and other contaminants (e.g., microbial contaminants) are separated from aqueous solutions through a filter medium, typically a membrane. The filtrate (fluid) pass through the membrane, whereas contaminants are collected by the membrane. Membrane filtration utilises pressure, particle size, concentration gradients, and the aqueous solution flux for contaminants removal. Filtration has gained increased attention in seawater desalination (Kim et al., 2015; Zhou et al., 2015) and for drinking/clean water reclamation and recovery from wastewater (Karakatsanis and Cogho, 2010; Mavhungu et al., 2020). Membranes with different perforations/aperture sizes, efficiencies, and performances have been employed for AMD treatment, including nanofiltration (NF), ultrafiltration (UF), microfiltration (MF), reverse osmosis (RO), and membrane distillation (MD) (Agboola et al., 2014; Fu and Wang, 2011; Shahrin et al., 2019). These have been widely employed for the removal of bacteria, microorganisms, particulates, and natural organic material, which can impart colour, taste, and odor to water and react with disinfectants to form harmful disinfection by-products (Mariah et al., 2006). Pre-treatment is often required to protect the membrane and prolong its lifespan.

Several researchers have explored the use of membranes for AMD treatment, with the role of membranes in AMD treatment being highlighted elsewhere (Agboola, 2019). In this regard, Lopez et al. (2018) evaluated the use of NF for the removal of heavy metals and sulfates from AMD, with the main challenges being membrane fouling, pre-treatment requirements, high-energy demand, brine generation, high cost, and low efficiency in the attenuation of monovalent and bivalent ions. Furthermore, RO has been used, as a polishing step, for the reclamation of drinking water from pre-treated AMD (Masindi, 2017b). However, in general, the main challenge with RO is brine generation (Ji et al., 2010). Nonetheless, the brine itself could act as new source for minerals recovery (Agboola, 2019). Finally, membrane distillation has been employed for salts recovery from brines (Janson et al., 2013) suggesting that it can also be used for the recovery of minerals from AMD or from filtration brines. Overall, due to the concentrated and heterogeneous nature of AMD, filtration as a stand-alone treatment method appears to hold little promise and most likely it could be applied in a treatment train for the polishing of the (pre)treated AMD and particularly for water reclamation.

4.4. Bioremediation

The concept of bioremediation relies on the use of plants (phytoremediation) and biological (microbial remediation) organisms, typically microorganisms such as bacteria and fungi, for the removal of

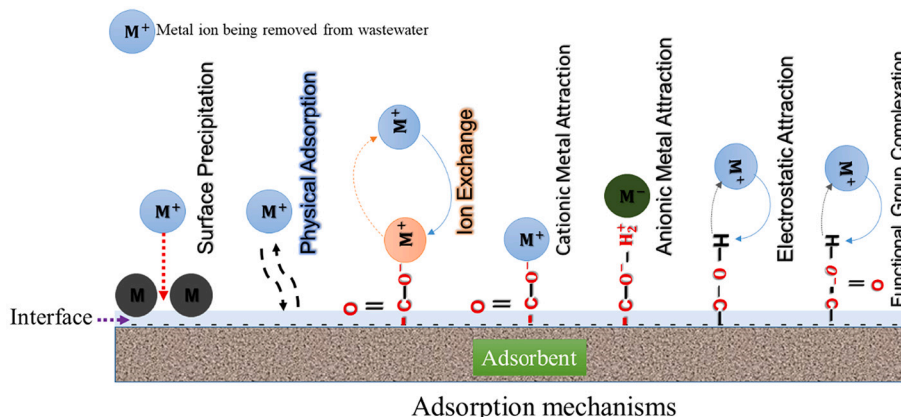


Fig. 2. Illustration depicting the different adsorption mechanisms (adapted from (Tran et al., 2017a)).

contaminants from water and soil or to completely mineralize water and soil-borne pollutants into relatively nontoxic constituents. In this technique, contaminants are removed via absorption/phytoextraction, phytoexcretion/phytovolatilization, phytostabilization, and aerobic and anaerobic bacterial and fungal degradation (Tripathi et al., 2020). In this sense, contaminants are attenuated/removed directly by microorganisms, inside the plant (absorption), and/or outside the plant's body (evaporation or stabilization). Moreover, biochemical and physiological mechanisms such as absorption, accumulation, sequestration, transportation, and degradation facilitate this process (Tripathi et al., 2020). Bioremediation has been proven effective in treating wastewater, including AMD, with phytoremediation being particularly promising for land rehabilitation. Bwapwa et al. (2017) assessed the phytoremediation of AMD (i.e., plant uptake of (semi)metals from AMD) using algae strains and highlighted the feasibility of recovering adsorbed metals due to the ability of algae to hyperaccumulate (semi)metals. Kiiskila et al. (2020) reported the metabolic response of vetiver grass (*Chrysopogon zizanioides*) after contacting AMD and Kiiskila et al. (2019) its efficiency, based on a multiscale long-term study, in treating AMD from the Tab-Simco mine site in southern Illinois, USA. In detail, vetiver rafts were suspended in 100-gallon containers and the following removal rates were observed: Fe 81%, Pb 81%, Ni 38%, Zn 35%, SO_4^{2-} 28%, Mn 27%, Cr 21%, Al 11%, and Cu 8.0%. Furthermore, it was observed that metals were mainly localized on the root surface as Fe plaques, whereas Mn and Zn showed greater translocation from root to shoot. Contrary, short-term and small-scale experiments showed removal efficacies of SO_4^{2-} (91%) and metals (90–100%) with the exception of Pb (15%) and Cu (0.0%). Overall, it was suggested that a floating treatment wetland system using vetiver grass could be cost effective and sustainable for AMD treatment (Kiiskila et al., 2020; Kiiskila et al., 2019).

Even though bioremediation appears promising, it is sensitive to numerous environmental externalities, such as plant and biological organisms' tolerance to varying conditions, including chemical species concentration, pH, and temperature. Furthermore, other drawbacks include (Ali et al., 2013; Asad et al., 2019; Gu, 2018; Syranidou et al., 2017; Wang et al., 2017):

- Poor performance in concentrated solutions such as AMD.
- Long residence time and slow hydraulic retention time is required for the effective removal of contaminants.
- Frequent monitoring of plants and microorganisms, along with the monitoring of their physicochemical properties, is needed.
- Large land areas are required, while the environment should be closely monitored and controlled to ensure the plants and microorganisms health.
- Mineral recovery in phytoremediation is impractical due to very low concentrations throughout the plants' body, while biomining is a not fully-fledged technology yet.
- Disposal of plants can pose secondary pollution and toxicity, hence proper management is required.

4.5. Crystallization

Desalination of aqueous solutions using thermal or freeze crystallization has gained increasing attention (Lewis et al., 2010). This is primarily traced back to the quest to attain zero-liquid-discharge (ZLD) processes and recover valuable minerals/salts. Specifically, thermal crystallization is achieved by means of thermal reactors or evaporators, which evaporate water and crystallize the salts that are dissolved in the aqueous solution. On the other hand, freeze crystallization involves a freezing-melting process, whereby water is crystallized to ice and separated from the concentrated solution (a eutectic point exist where ice and salt solutions simultaneously exists) (El Kadi and Janajreh, 2017). In AMD treatment, water can be recovered either by subjecting AMD into temperatures that are suitable for water to freeze but leave concentrated solution (contaminants) in fluid form, whilst in thermal

crystallization AMD is boiled (which is more energy intensive than freezing) to evaporate the water leaving contaminants in the container. Thermal crystallization is often used to treat effluents with high Fe concentrations (e.g., rejects for hydrometallurgical processes) and recover Fe as hematite, goethite, and magnetite, however, this process is uneconomical for drainage that contains low Fe concentrations (Yang et al., 2021). Freeze crystallization is an emerging technology for mine water and brine effluents (Randall et al., 2011) and it has been found promising for sulphuric acid recovery from AMD (Nleya et al., 2016). Overall, thermal and freeze crystallization have been widely used for desalination but not for AMD treatment due to AMD's concentrated and heterogeneous nature and crystallization's high energy demand, among others.

5. Treatment methods

If abatement techniques are not in place or fail to operate, the generated AMD should be treated, using the aforementioned mechanisms for contaminants removal (section 4), to safeguard human health and the environment. Traditionally, treatment was achieved by active or passive systems; however, more recently, hybrid and integrated systems, where active and/or passive treatment are combined in a step-wise or sequential fashion, have also emerged (Kefeni et al., 2017b; Nleya et al., 2016; Park et al., 2019). It should be noted that integrated and hybrid treatment is encountered interchangeably in the literature, however these treatment methods typically have different goals, aims, and objectives and therefore a distinction is made here. Specifically, in hybrid systems active and passive systems are combined, aiming at providing a high treatment efficiency. On the other hand, integrated treatment refers to the sequential or stepwise treatment of AMD, typically only by active techniques and aiming at removing and recovering metals and minerals at different treatment steps, while water reclamation might also be pursued. Therefore, in the context of circular economy integrated systems can be used to introduce reuse, recycle, and resource recovery paradigms in wastewater treatment. Below, the main strengths and weaknesses of each treatment system are briefly discussed.

5.1. Active treatment

Active systems typically employ large inputs of energy, chemicals, and other materials to drive the treatment process. For this reason, this method is also known as chemical treatment, since typically alkaline chemicals, such as $\text{Ca}(\text{OH})_2$, CaO , NaOH , Na_2CO_3 , NH_3 , MgO , and $\text{Mg}(\text{OH})_2$, are used to increase the pH and precipitate the metals that are contained in AMD. Masindi et al. (2017a) compared the use of various alkaline materials for the active treatment and the flowing neutralisation capacities, from higher to lower score, were identified: $\text{NaOH} \geq \text{Ca}(\text{OH})_2 \geq \text{CaO} \geq \text{MgCO}_3$ (cryptocrystalline magnesite) $\geq \text{MgO} \geq \text{Na}_2\text{CO}_3 \geq \text{Mg}(\text{OH})_2 \geq \text{CaCO}_3$. Similar results had been reported by Potgieter-Vermaak et al. (2006). Furthermore, Kefeni et al. (2018) explored the use of Fe-based minerals for AMD treatment, which were found promising for metals and sulfate attenuation, through adsorption and other mechanisms. However, the production of these alkaline materials requires resources, energy, and infrastructure (Kaur et al., 2018). As such, active treatment tends to be expensive, while its environmental footprint is considered to be higher compared to passive treatment (Skousen et al., 2019).

Apart from the use of alkaline agents, filtration, bio-barriers, and sorption/ion exchange technologies have been employed in active systems. Adsorption techniques for AMD treatment typically entail the use of different natural and synthetic materials, such as activated carbon, clay minerals, and other synthetic compounds or adsorbents (Hong et al., 2014; Masindi et al., 2015b; Xingyu et al., 2013). For AMD filtration, different membranes have been examined, with NF attracting the largest share of attention (Aguar et al., 2018; Al-Zoubi et al., 2010; Lopez et al., 2018). Specifically, membrane treatment has been found

promising in terms chemical species removal, however fouling is a limiting factor, while the need for pre-treating the AMD and managing the generated retentate (brine) further impose on the viability of the process (Agboola, 2019; Kefeni et al., 2017b; Masindi, 2017b; Rambabu et al., 2020). To address these concerns, research has recently focus on MD, which is an emerging technology to treat AMD, retentate, and softened water (Amaya-Vías et al., 2019; Foureaux et al., 2019; Ryu et al., 2019).

Overall, due to their high cost and complex nature, active systems are typically employed in operating mines, where capital is available and personnel is already in place, rather than in abandoned ones. They also provide a higher degree of treatment, as compared to passive systems which are less efficient in contaminants removal but also less complex and costly. A typical AMD active treatment system, where alkaline materials are used for contaminants removal, is shown in Fig. 3.

5.2. Integrated treatment

In integrated or multi-staged or sequential or stepwise AMD treatment, the main focus is the recovery of metals and minerals and/or water reclamation. As such, active techniques are mainly used and therefore integrated treatment can be also considered as a branch of the active treatment. Specifically, active systems can comprise various subprocesses within an overall treatment system (EPA, 1983). However, during the past few years the combination of different active processes in a step-wise fashion, and towards the recovery of different metals/minerals from each step, has gained attention (Simate and Ndlovu, 2014). In detail, in integrated systems, contaminants are removed/recovered in different steps, typically by using different pH gradients (Table 2) and/or different mechanisms. Integrated systems are mainly based on precipitation, adsorption, and filtration (Kefeni et al., 2017b; Nleya et al., 2016), with metals/minerals selective precipitation, by controlling the pH of the AMD in a stepwise fashion, having attracted much attention (Park et al., 2013; Passos et al., 2021). An integrated system can comprise two (Igarashi et al., 2020) or more (Masindi et al., 2018b) steps, where different metals/minerals are removed in each step. Therefore, single-step systems that are followed by a pH correction step (e.g., (Kalombe et al., 2020)) are not considered here as integrated treatment. The main strength of integrated systems, apart from their

high treatment efficiency, is that they can be used for the valorisation and beneficiation of mine drainage, i.e., harbour circular economy concepts (Masindi et al., 2019a). The reclaimed/recovered resources could also be used to reduce cost (Singh et al., 2020) and possibly the environmental impacts of the treatment process. A typical integrated AMD treatment process, where AMD is valorised and benefited (clean water and minerals recovered) is shown in Fig. 4.

5.3. Passive treatment

Passive treatment systems are typically used in abandoned mines and areas where the effluent requires less treatment. Wetlands have been traditionally used, which remove contaminants and neutralize pH through bioremediation (Kiiskila et al., 2020). Different mechanisms are at play in bioremediation, since living organisms can be used to mineralize or render contaminants less harmful (microbial remediation) and/or plants to sorb and bioaccumulate contaminants in their tissues (phytoextraction) or excrete them to the atmosphere (phytovolatilization) (section 4). Due to AMD's relatively toxic nature, bioremediation in wetlands is preferred, while depending on the plant species high treatment efficiencies can be achieved (Kiiskila et al., 2019). Wetlands can be divided into aerobic and anaerobic while smaller system such as limestone drains or channels are also employed for the passive treatment of AMD.

5.3.1. Wetlands

Wetlands can be: i) aerobic, i.e., shallow ponds (<30 cm) that primarily slow down AMD to allow metal oxidation, hydrolysis, and particle settling, or ii) anaerobic, i.e., relatively deep (>30 cm) ponds where anoxic conditions, created due to high biochemical oxygen demand (BOD), promote bacterial sulfate reduction to sulfides, which form insoluble metal precipitates (e.g. FeS_2) and produce alkalinity that causes metal precipitation as (oxy)hydroxides (Skousen et al., 2019).

Aerobic wetlands comprise floral systems that are primarily filled with soil or limestone gravel (lined or unlined). Limestone can be pulverised to increase its reactive surface area and therefore increase treatment efficiency. Through these systems, metal oxidation and precipitation from AMD (e.g., Fe, and Mn) is naturally accomplished (Rambabu et al., 2020). Not only this, but plants in aerobic wetlands also

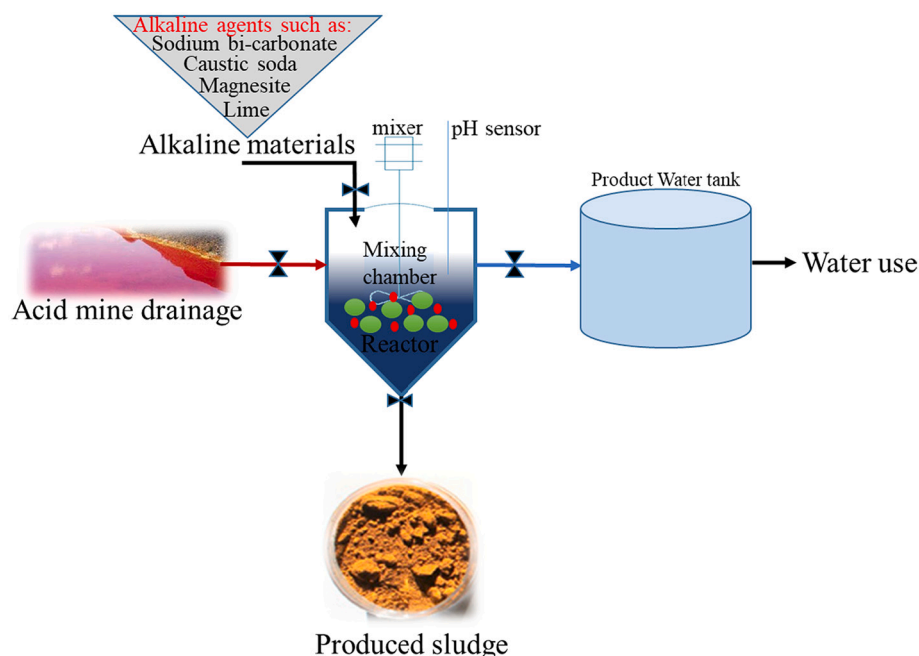


Fig. 3. Schematic illustration of a typical AMD active treatment process (adapted from Masindi et al. (2019a)).

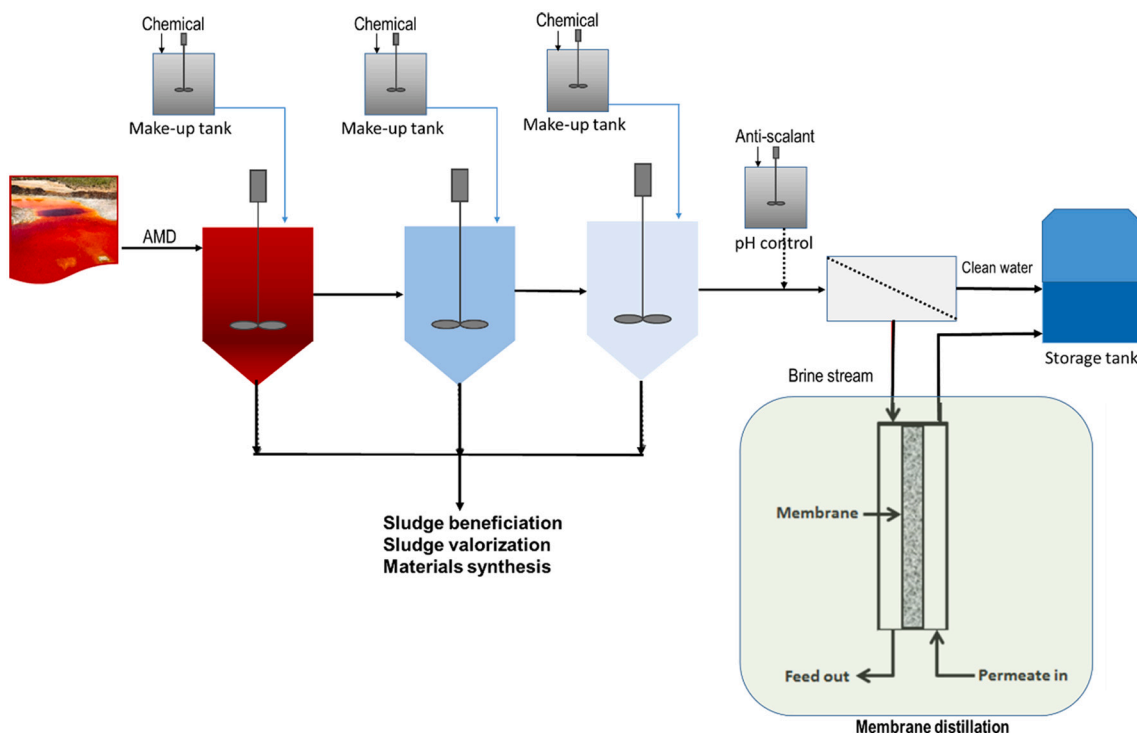


Fig. 4. Schematic of a typical integrated AMD treatment system (adapted from Masindi et al. (2019a)).

absorb nutrients, which they use for their growth (Sheoran and Sheoran, 2006; Skousen et al., 2019). Aerobic wetlands are generally easy to operate and require little to no energy and/or materials/chemicals input after construction (Fig. 5).

Anaerobic wetlands also neutralize acidity and reduce metals, however, in this case, in the absence of oxygen. Specifically, anaerobic reactions consume H^+ , hence the reduction in acidity. Anaerobic wetlands are mainly filled with organic matter, such as compost, and usually underlain by limestone gravel (Skousen et al., 2019). Water percolates through the organic matter to the limestone bed, thus achieving anaerobic conditions. This encourages metals to precipitate as sulfides. Furthermore, the decomposition of the organic matter, which is achieved by different microorganisms already contained in the organic matter, also consumes the dissolved oxygen leading to the production of alkalinity and hydrogen sulfide (H_2S) (Ben Ali et al., 2019; Rambabu et al., 2020; Skousen et al., 2017).

5.3.2. Anoxic limestone channels, limestone ponds, and permeable reactive barriers

Apart from aerobic and anaerobic wetlands, anoxic limestone drains or channels and alkaline leach beds have also been employed for the passive treatment of AMD (Sheoran et al., 2010; Skousen et al., 2017). In

these systems, alkaline materials, typically limestone which is abundant and inexpensive, are used for AMD neutralization and for the precipitation of metals. Anoxic limestone drains and channels consist of buried limestone gravel systems, where AMD flows through and is anaerobically treated (Skousen et al., 2019). However, if O_2 or Al are present within the channel, then Fe and Al hydroxides might form, clogging the system and leading to its failure. Alkalinity producing systems can also be a combination of an anaerobic wetland (anaerobic microorganisms) and an anoxic limestone drain (absence of O_2 and Al in AMD) (Gazea et al., 1996).

Other types of passive systems include various limestone treatment configurations, such as limestone ponds which can be constructed over an AMD upwelling, seep, or underground discharge. In particular, the limestone is placed in the bottom of the pond and AMD flows upward through the limestone to open limestone channels, in which water flows down a steep slope ($\geq 20^\circ$ steepness) to prevent precipitation of metals. In general, these systems oxidize and precipitate metals and add alkalinity to the water (Rambabu et al., 2020; Skousen et al., 2019). Limestone channels and wetlands can also be used together (Fig. 5).

Finally, the use of permeable reactive barriers (PRBs) has also been explored, where AMD passes through a reactive media (e.g., a mixture of Portland cement with fly ash), where contaminants are attenuated. Even

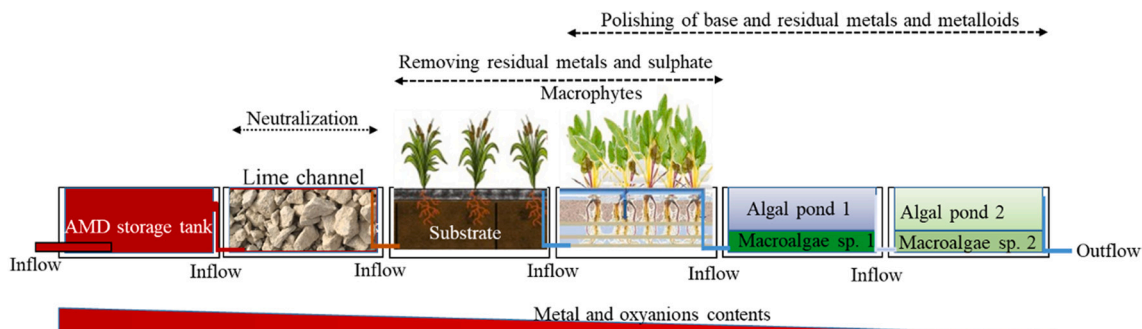


Fig. 5. Schematic illustration of a passive AMD treatment system, i.e., limestone drain with a wetland system.

though PRBs are associated with several limitations (e.g., complexity, clogging, and armouring), they appear to be promising in terms of contaminants removal from AMD (Shabalala et al., 2014). For example, Shabalala et al. (2017) evaluated the use of PRBs for AMD treatment and the removal efficiencies for Al, Fe, Mn, Co and Ni were as high as 87%, 96%, 99%, 98% and 90%, respectively.

Overall, key challenge in passive systems include treatment efficiency and long-term reliability, primarily limited by blockages from precipitates, armouring (covering) of the reactive materials, and preferential flow channelling.

5.4. Hybrid treatment

Hybrid treatment refers to the integration of active with passive processes for the treatment of AMD (Fig. 6). Most often, in hybrid systems neutralization with alkaline media (active) is combined with aerobic or anaerobic wetlands (passive) (Groudev et al., 2008; Moodley et al., 2018; Naidu et al., 2019). Therefore, treatment efficiency can be optimized and a relatively high quality effluent can be obtained. Albeit, the main challenges of hybrid systems include space requirements, sensitivity to certain chemical species, and frequent maintenance, while they are most effective only under certain flow and acidity conditions (Masindi and Tekere, 2020). It should be noted that even though this process appears promising, it has not been studied in detail and more research is required to assess the performance of hybrid systems for large-scale applications (Rambabu et al., 2020; Simate and Ndlovu, 2014).

5.5. Advantages and disadvantages of active, passive, hybrid, and integrated systems

In general, active systems are considered more robust and efficient in AMD treatment than passive ones. However, to achieve high treatment efficiencies they require infrastructure (e.g., tanks, pipes, pumps), specialized O&M services, along with regular chemical and energy inputs (Naidu et al., 2019). However, passive systems also require infrastructure, but this is much simpler and cost-effective (per treated AMD volume) than in active systems. As a result, active systems are perceived as less eco-friendly than passive ones (Skousen et al., 2017). Nonetheless, more research is required towards that end since active systems achieve higher treatment efficiencies than the passive systems, which, among others, is also beneficial for the receiving ecosystems.

On the other hand, passive systems do not require frequent O&M, energy, and chemical inputs and therefore are deemed more suitable for remote locations and abandoned mines (Naidu et al., 2019). Furthermore, their maintenance is relatively simple, while they do not grossly impose on the aesthetic quality of the area that they are installed on, since they can appear natural and support plants and local wildlife. They are also far less expensive than active systems of the same capacity

(Skousen et al., 2019). Nonetheless, passive systems are also associated with many drawbacks, since: i) they might not treat the AMD effluent to a high standard, due to their passive nature, while their efficiency can be also affected by external factors such as weather (e.g., low temperatures can affect the operation of wetlands) (Skousen et al., 2017); ii) blockages can be frequent, greatly constraining their operation (Ben Ali et al., 2019); iii) they cannot be employed in circular economy concepts, since resource recovery is complex and difficult for such systems; and iv) highly mineralised sludge is produced, which is difficult to treat and makes sludge use or beneficiation practically unfeasible (Masindi and Tekere, 2020).

As discussed above, to overcome the limitations of active and passive systems, hybrid and integrated treatment systems have been recently proposed and explored. In hybrid treatment, active and passive systems are combined towards the effective and sustainable treatment of AMD, albeit resource recovery and water reclamation is also difficult to achieve. Integrated treatment typically comprises active systems in a step-wise fashion, with the main aim being resource recovery (metals/minerals) and possibly water reclamation. Overall, both hybrid and integrated systems are versatile and can provide high contaminants removal efficiencies. Furthermore, integrated systems can be also used to introduce reuse, recycle, and resource recovery paradigms in AMD treatment.

6. Life cycle assessment in AMD prevention and treatment

The environmental sustainability of different AMD treatment systems has been examined using the LCA methodology (Table 4). Most works have focused on active and/or passive systems, while the body of knowledge on hybrid and integrated system is limited. Few LCA studies have also focus on the environmental sustainability of AMD prevention systems.

6.1. Environmental sustainability of prevention technologies

Regarding the environmental sustainability of AMD prevention technologies, Sarkkinen et al. (2019) assessed the environmental performance of five different tailing pond cover systems (Table 4), with the advanced hardpan cover liner being the most environmentally friendly solution, however, this can be site and case specific. Broadhurst et al. (2015) examined the environmental benefits and impacts of incorporating desulfurisation flotation for the removal (and possibly recovery) of sulfide minerals from sulfide-bearing mine waste, as a means to prevent AMD formation. The removal of sulfide minerals from the tailings had a positive impact on the majority of the examined impact categories, while valuable resources, such as water, residual metals, and sulfur could also be recovered and further reduce environmental impacts, but this was outside of the system boundary of the LCA study. Finally, Reid et al. (2009) examined different scenarios regarding the

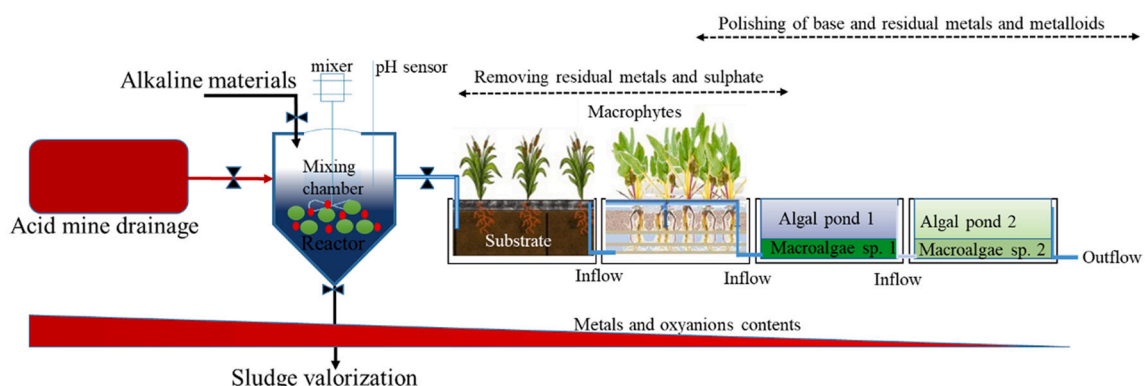


Fig. 6. A typical hybrid treatment process, where alkaline neutralization (active) is combined with an aerobic wetland (passive).

Table 4
LCA studies on the environmental sustainability of AMD prevention and treatment systems.

No	Treatment method	Functional unit	Geographic location	LCA method	System boundary	Main findings	Reference
1	Active (seawater neutralised red mud and quicklime)	1000 m ³ untreated acidic pitwater	Mount Morgan, Queensland, Australia	Total CO ₂ eq and net energy use	Cradle to grave (raw material extraction to disposal)	It is possible to reuse seawater neutralised red mud from alumina refineries to treat AMD	(Tuazon and Corder, 2008)
2	Passive (disposal or disposal and backfilling)	Management of 3,328,065 t of solids and 15,130,720 m ³ copper and zinc tailings	Abitibi, Quebec, Canada	IMPACT 2002+	Cradle to grave (From mine development to closure)	Local characteristics (e.g., mineral ore grade, soil, and topography) could significantly influence the total environmental impacts	(Reid et al., 2009)
3	Active (two) and passive (five)	1 kg of acidity neutralized per day	Stockton Coal Mine, South Island, New Zealand	ReCiPe 2008	Construction, operation, and maintenance of AMD treatment technologies	Passive treatment has lower environmental impacts than active, however active treatment is more efficient in treating AMD	(Hengen et al., 2014)
4	Active (desulfurisation flotation, using a xanthate collector, and/or dewatering and then discharge to tailings impoundment)	100 t of dry tailings per day from a sulfide-bearing mine	South Africa	USEtox and ReCiPe 2008	The pre-disposal treatment of base metal sulfide tailings is included in the system boundary but not the treatment of the generated sulfide-rich stream	Existing LCA models cannot reliably and comprehensively assess environmental impacts associated with solid mineral wastes	(Broadhurst et al., 2015)
5	Integrated (magnesite, lime, soda ash and CO ₂ bubbling).	1 m ³ of treated AMD	South Africa	ReCiPe 2008	Cradle to gate, i.e., not including the recovered water but the recovered mineral resources are included in the analysis	Renewable electricity can minimize the process environmental footprint, while resource recovery appears to be promising	(Masindi et al., 2018c)
6	Passive (dispersed alkaline substrate technology)	1m ³ of treated AMD	Spain	ReCiPe 2008	Cradle to gate, i.e., end-of-life aspects of the treatment plant and the disposal or valorisation of the generated sludge are external to the system boundary	Even though the treatment plant has an initial step environmental impact, this becomes negligible within a few years (4.5 yr)	(Martínez et al., 2019)
7	Five different cover structure options, i.e., moraine-based, biofuel fly ash/steel slag, stabilization with ordinary Portland cement or with composite binder, and AHCL.	Covering of a 150-ha pond (comparative analysis between different options)	Ostrobothnia region, Finland	IPCC 2013, Cumulative Energy Demand, ReCiPe 2016	LCA based on the materials used in the cover alternatives accounted for the production of the materials and transport to the site	AHCL was the most environmentally friendly cover option, however results can be affected by case and site-specific parameters	(Sarkkinen et al., 2019)
8	Passive (biological treatment) and active (adsorption system)	Treatment of 20 m ³ per day	Southwest China	CML 2001, ILCD, and Eco-indicator 99 (I, I)	Cradle to grave, i.e., construction, operation, and disposal of both active and passive systems	Passive treatment is more environmentally friendly than active since its operational phase has a minimal impact compared to active treatment which is energy and material intensive to operate	(Wang et al., 2020)
9	(1) immobilized microalgal system, (2) conventional treatment process of AMD using lime, and (3) hybrid system of calcined eggshells and microalgae	The treat 1.0 m ³ of AMD	Newcastle, NSW, Australia	Does not specify	Cradle to grave	The immobilized microalgal system exhibited the lowest environmental impacts across the examined impact categories	(Abinandan et al., 2020)
10	CEReS process using coal mine AMD	Processing of 1 t of PCBs	Does not specify	Does not specify	PCBs transportation, shredding and three core processes of the CEReS method: pyrolysis, bioleaching and char leaching	CEReS could reduce environmental impacts compared to PCBs incineration and also aligns with the circular economy concept, securing the sustainable supply of critical raw materials	(Kouloumpis and Yan, 2019)
11	CEReS process using AMD from sulfidic coal wastes	Does not specify	Poland	Does not specify	Does not specify	Co-processing AMD with electronic waste streams appears to be a low-cost waste management treatment option with demonstrable environmental benefits	(Bryan et al., 2020)
12	CEReS process using coal mine AMD	Treatment 1 t of PCBs	Poland	ReCiPe 2016 v1.1 Midpoint (H)	PCBs transportation, sorting, and shredding and CEReS four main stages, i.e., pyrolysis, bioleaching, char-leaching and solvent extraction/electrowinning	14 kt of PCBs can be co-processed with 79 kt of coal mine waste and compared to landfilling and incineration lower environmental impacts in	(Kouloumpis and Yan, 2022)

(continued on next page)

Table 4 (continued)

No	Treatment method	Functional unit	Geographic location	LCIA method	System boundary	Main findings	Reference
					(SX/EW). Valuable by-products were also included in the analysis	toxicity but higher in climate change related categories were observed. By-product valorisation (brine and especially the recovery of copper) reduced the scores of the toxicity, eutrophication and metal Depletion categories Decarbonized electricity and reductions in the water and energy inputs were proposed.	

post-closure management of the tailings from a copper and zinc underground mine and highlighted the importance of including land-use along with the long-term impacts of tailings management in the system boundaries of LCA studies.

6.2. Environmental sustainability of active and passive treatment systems

One of the first LCA studies that focused on the environmental sustainability of AMD treatment was published in 2008 and dealt with the use of seawater neutralized red mud (bauxite residue from the alumina refining process) instead of the conventional quicklime treatment (Tuazon and Corder, 2008). In terms of total CO₂eq emissions, the seawater neutralized red mud had an 80% lower score and consumed 66% less electricity, albeit red mud's neutralization potential was 12 times lower than that of quicklime, imposing on the total fuel consumption and transportation among others (Tuazon and Corder, 2008). In another LCA study by Martínez et al. (2019), the environmental sustainability of dispersed alkaline substrate (DAS) treatment was examined. DAS is a passive treatment process that is based on the use of fine-grained alkaline materials (limestone, magnesite, and magnesium oxide) for AMD neutralization, mixed with an inert high porosity material (in this case wood chips) to accommodate the flow of AMD. It was identified that the construction of the DAS treatment plant, along with the energy use, had a negligible impact compared to the use of the alkaline materials, which were identified as the main environmental hotspot of DAS treatment (Martínez et al., 2019).

Wang et al. (2020) compared a passive (onsite field-scale bioreactor) with an active (adsorption using activated alumina) system for the treatment of antimony (Sb) rich mine drainage and the passive treatment, which had a lower energy consumption, was more environmentally friendly. The main contributor to the environmental impacts was the construction phase whereas the operation phase had a minimal contribution. This was not the case for the active system, where energy and the activated alumina (which after use needs to be disposed of) consumed during the operation phase were the main contributors on the total environmental impacts (Wang et al., 2020).

Moreover, Abinandan et al. (2020) performed a comparative analysis, from the environmental perspective, between immobilized acid-adapted microalgal, calcined eggshell–microalgal, and limestone for the removal of Fe from AMD and the first achieved a better environmental performance. The immobilized acid-adapted microalgal AMD treatment technology had also the potential of Fe recovery from AMD, while after treatment the microalgae could be used for biodiesel production, however, these were outside of the LCA system boundary. Finally, Bryan et al. (2020) examined the possibility of co-processing AMD with electronic waste (low-grade printed circuit boards - PCBs) to produce metals and other valuable products and at the same time minimize environmental impacts. This was achieved by using AMD to generate biolixiviant, a highly corrosive solution that was then used to leach metals from processed PCB waste. This process is entitled CEReS (Kouloumpis and Yan, 2019) and compared to landfilling and

incineration CEReS minimizes environmental impacts related to toxicity but, unless decarbonized electricity is used, greatly (tenfold) increases the climate change impact (Kouloumpis and Yan, 2022).

6.3. LCA studies on hybrid and integrated systems

Hengen et al. (2014) examined the environmental sustainability of both active and passive AMD treatment technologies and identified that passive systems had lower environmental impacts but active were more promising for the effective treatment of AMD from large-scale mining operations. In their results, they noted that combining active and passive technologies, i.e., hybrid treatment, can balance operational treatment requirements with environmental impacts (Hengen et al., 2014). Furthermore, Masindi et al. (2018c) examined the environmental sustainability of an integrated AMD treatment system, at semi-industrial scale, where coal mine AMD was treated in a stepwise fashion to simultaneously remove contaminants and recover valuable resources, i.e., water, iron, gypsum, and brucite. The authors highlighted that the introduction of renewable energy to drive the treatment process can greatly reduce the total environmental footprint of the process and that even though resource recovery appears promising, more research is required towards this end (Masindi et al., 2018c).

6.4. State of the art and future research direction in LCA

The existing body of knowledge on the environmental sustainability of AMD prevention/abatement and treatment is limited (Table 3), suggesting that there is scope for further research and development (R&D). The absence of LCA studies and the need for their introduction to improve the environmental sustainability of mining activities has also been highlighted elsewhere (Asif and Chen, 2016). From the reviewed LCA studies it can be inferred that different functional units, system boundaries, and life cycle impact assessment (LCIA) methods have been used, suggesting that comparison of their results cannot be direct. Different treatment methods have also been examined, mainly focusing on passive and active systems, and in different geographical locations spanning from Australia (Tuazon and Corder, 2008) to South Africa (Masindi et al., 2018c) to Canada (Tuazon and Corder, 2008) and China (Wang et al., 2020)(Wang et al., 2021), among others.

Overall, due to its lower material and energy inputs during operation passive treatment appears to be more environmentally friendly than active, however active treatment achieves better contaminants removal efficiencies. This suggests that their combination, through hybrid systems might be the optimal solution for improved contaminants removal and reduced environmental impacts. However, this assertion needs to be supported by LCA. Integrated systems appear to have high environmental footprints; however, valorisation (reclaimed water) and beneficiation (recovered metals/minerals) pathways can credit them system with environmental benefits, traced back to the avoided impacts from the production processes of the reclaimed/recovered materials. However, similar to the hybrid systems more research is required on the

environmental sustainability of integrated treatment systems.

7. AMD valorisation and beneficiation

AMD contains numerous valuable metals, which could be used towards the synthesis and recovery of different minerals. Water can also be reclaimed, suggesting that AMD could be perceived as a resource rather than as a waste (Masindi et al., 2019b; Silva et al., 2019). The recovery and reclamation opportunities may also reduce the overall cost and

environmental footprint of the treatment process (Masindi et al., 2021). Specifically, AMD valorisation could be achieved through water reclamation (e.g., drinking water (Masindi et al., 2018a)), while its beneficiation could be achieved through the recovery of metals, such as Fe (Akinwekomi et al., 2017) and other metals/minerals (Masindi et al., 2019b; Silva et al., 2012). However, the main challenge for AMD beneficiation lies in the minerals co-contamination during recovery. This is attributed to the presence of chemical species that easily precipitate, co-precipitate, and co-adsorb, hence co-contaminating the

Table 5
Different piloted integrated technologies, along with their main advantages and disadvantages.

Technology	Location	Advantages	Disadvantages	TRL	References
ABC	South Africa	Treats water to the required discharge limit Recovery of sulfate	Hazardous, heterogeneous and highly mineralised sludge generation Requires precise barium dose to balance the stoichiometry and residual barium	4-6	(Mulopo, 2015)
Adapted SAVMIN process	South Africa	Treat water to the required discharge limit Feasibility of recovering valuable minerals The SAVMIN process is mature (TRL 9) therefore scaling up will be straightforward	Hazardous sludge generation Generation of toxic sludge due to mixture of heavy metals Complex process	2-3	(Abdrakhmanova et al., 2019)
ALSX	USA	Producing a high-grade and dense (0.1% to 5% on a dry weight basis) rare earth concentrate Commercially valuable rare earth oxides can be produced	Process at pilot scale Water is not reclaimed (is only fit for discharge) Sludge is produced that requires handling and treatment	2-3	(Ziemkiewicz et al., 2021)
BaCO ₃ solid-state reaction process	Japan	Effective for the beneficiation of AMD sludge Able to synthesize M-type hexaferrite, which has many industrial applications	Requires barium carbonate and hematite to operate An AMD treatment process is also required, preferably one that recovers water	2-3	(Liu et al., 2019).
BOF SRO	South Africa	Reclaim drinking water Recover gypsum	Voluminous sludge rich in Fe Require high dosage of basic oxygen furnace slag. Generation of brine which is difficult to dispose	2-3	(Masindi et al., 2017b)
MASRO(E)	South Africa	Treat water to the required drinking limit Recovery of Fe-based minerals Recovery of gypsum and lime	Cost intensive Brine generation Heterogeneous Fe-sludge with unviable recovery feasibility	4-6	(Masindi, 2017b) Masindi et al., 2019a) (Masindi et al., 2018a)
DESALX	Australia	High (> 90%) water reclamation potential Water quality is fit for aquifer re-injection	As- and Sb-rich sludge is produced Gypsum-based sludge is produced	9	(Clean TeQ Water, 2022a; Clean TeQ water, 2022b)
EARTH	South Africa	Uses ion exchange to recover uranium and sulfate The sulfate can be used to produce ammonium sulfate Water quality for discharge or re-use (wash water)	Cost intensive Generation of toxicants from ion exchange regenerants Require acid for operation.	6-7	(Howard et al., 2009)(Simate and Ndlovu, 2014)
FGR - DAF	Brazil	Able to greatly remove SO ₄ ²⁻ removal from coal AMD Able to reclaim water for irrigation, industrial uses, or as wash water	No insight about cost and energy profile The generated sludge will require treatment	4-5	(Rubio et al., 2007) (da Silveira et al., 2009)
HiPRO	South Africa	Drinking water is reclaimed Gypsum is synthesized and recovered Brine free and zero-liquid-discharge process	Cost intensive Hazardous sludge from the ozonation process	9	(Karakatsanis and Cogho, 2010)
HydroFlex	USA	High reduction in sulfate (<100 mg L ⁻¹) and metals (<1 mg L ⁻¹) levels Reclamation (>95%) of non-potable water that can be used in hydraulic fracturing	Metals are not recovered Policy and regulations challenges Various engineering challenges	6-7	(Peterson et al., 2014) (Lane, 2016).
LoSO ₄	Denmark and USA	Greatly reduce sulfate level (≤50 mg L ⁻¹ when high levels of AL are available) and recovers >95% of the aluminum-based salt High quality treated effluent for reuse or discharge Sulfate levels are reduced to 200–1000 mg L ⁻¹ Metal impurities such as Ni, Cd, Cu, and Zn are removed as metal hydroxides Low sulfate concentration effluent fit for discharge or re-use	High consumption of chemicals during treatment Preferably AMD with high sulphate content should be used Sulfate levels below 200 mg L ⁻¹ require enhanced precipitation chemistry. Water reclamation and particularly minerals/metals recovery appears to hold little potential	9	(Sandru et al., 2017; Veolia, 2016)
Metso Outotec process	Finland	Low sulfate concentration effluent fit for discharge or re-use	Sulfate levels below 200 mg L ⁻¹ require enhanced precipitation chemistry. Water reclamation and particularly minerals/metals recovery appears to hold little potential	9	(Metso Outotec, 2022)
ROC technology	South Africa	Drinking water reclamation through filtration Minerals recovery from brine using freeze technique Recover Fe-based minerals for the synthesis of goethite, hematite and magnetite	Cost intensive Still at pilot scale Complex process that require intense training Challenges with the synthesis of Fe-based minerals.	4-5	(Zvinowanda et al., 2014)
TUT MBA	South Africa	Recovery of valuable minerals such as barium sulfate. Reclamation of product water that can meet the discharge limits and minimize ecological contamination Recover barium using GYP-SLIM	Generation of hazardous, heterogeneous and highly mineralised sludge in magnesium stage. Require precise barium dose to balance the stoichiometry and residual barium.	4-5	(Bologo et al., 2012)

resultant product (Madzivire et al., 2011). To enhance the precision of the minerals recovery process, and enable AMD beneficiation and valorisation, geochemical software tools such as PHREEQC can be used to predict mineral synthesis when using different materials such as limestone, lime, periclase, brucite, soda ash, caustic soda for AMD treatment (Parkhurst and Appelo, 1999).

7.1. Water reclamation

The dissolved solids in AMD, apart from being contaminants themselves, also increase electrical conductivity (EC) and turbidity. Therefore, by removing and recovering dissolved solids, i.e., AMD beneficiation, a high quality effluent can also be produced. This raises opportunities for water reclamation, which can be used for a variety of purposes, spanning from direct discharge to the environment (Naidu et al., 2019), to irrigation (Hentati et al., 2014), to industrial reuse (Choi et al., 2019), or even to drinking water (Masindi et al., 2019a).

7.1.1. Drinking water reclamation

To reclaim drinking water, the polishing of the pre-treated AMD is typically achieved through filtration techniques, which eliminate the majority of the remaining dissolved metals. For example, Bruce et al. (2009) evaluated the use of neutralization, UF, and RO (HiPRO process) for the reclamation of drinking water from AMD. The plant was commissioned and operated in September 2007 in South Africa, and its capacity was upgraded to 20 ML day⁻¹ in June 2008, achieving water recovery >99% (Bruce et al., 2009). The BOF SRO (basic oxygen furnace soda ash and reverse osmosis) (Masindi et al., 2017b), the MASRO(E) (magnesite softening, reverse osmosis, (eutectic freeze crystallization)) (Masindi, 2017b), and the ROC (Zvinowanda et al., 2014) systems have been also proposed for drinking water reclamation from AMD in the South African setting, where the large volumes of AMD produced year-round along with water scarcity concerns have pushed innovation. These and other water recovery systems from AMD, along with their technology readiness levels (TRLs), are summarised in Table 5. Finally, in Australia where water availability is also a concern, the DESALX process has been proposed, where water that is fit for aquifer re-injection can be produced (Clean TeQ Water, 2022a; Clean TeQ water, 2022b) while, Ryu et al. (2019) reported that RO could be used, as a polishing step, to reclaim drinking water from AMD. To reduce membrane fouling, Fe and Al were removed using an integrated submerged direct contact membrane distillation (DCMD) – natural Australian zeolite sorption system, where around 50% of the water was recovered in 30 hours, while sulfuric acid (H₂SO₄) and valuable metals (Cu, Zn and Ni) could also be recovered (Ryu et al., 2019).

7.1.2. Irrigation with untreated and treated AMD

In water scarce regions AMD-contaminated streams, which are primarily enriched with trace metals, have been directly used for irrigation (Garrido et al., 2009b; Grewar, 2019). However, trace metals from untreated AMD will eventually leach to the soil and translocated onto agricultural produce, degrading the ecosystem and impacting human health. For example, in a case study in Potosí, Bolivia the secondary risk of ecotoxicological contamination was highlighted when AMD-influenced irrigation water was used in potato cultivation, affecting both the agricultural land (high Cu, Pb and Zn concentrations) and the commercially grown potatoes, implying that agricultural products from this region may represent a potential health risk (Garrido et al., 2009a). In addition, Lin et al. (2005) examined agricultural soils directly irrigated with AMD in Guangdong, China, and heavy metal concentrations along with pH values as low as 3.9 were identified in soil, while heavy metals, particularly cadmium (Cd) which was well above China's prescribed limit, had also leached to agricultural produce. Therefore, it appears that irrigation with AMD and AMD-influenced waters should be avoided.

A more promising strategy is to use treated AMD for irrigation. To

this end, a simple and cost-effective method is to treat the AMD with limestone and its derivative lime. Specifically, Jovanovic et al. (2002) examined the use of lime-treated AMD for the irrigation of agronomic and pasture crop species in Mpumalanga, South Africa and increased yields, compared to rain-fed crops, were reported. Albeit, shallow rooting depths were also observed, possibly due to high soil acidity, compaction, and phosphorous (P) deficiency in deeper layers. However, after the third year of irrigation, soil pH increased, which suggests that proper irrigation and fertilisation practices should be in place when using lime-treated AMD (Jovanovic et al., 2002). A few years later, the same group reported that treated AMD can provide irrigation water during the dry winter season in South Africa, provided that salt runoff and salt leaching could be intercepted, thereby minimizing the impact to groundwater (Jovanovic et al., 2004). Annandale et al. (2006) and Hentati et al. (2014) reported similar findings. Finally, Robinson et al. (1998) reported the feasibility of using neutralization and ion exchange for treating AMD and reclaiming water for agricultural purposes. The probable synthesis of ammonium nitrate and high purity calcium sulfate from the effluent regenerants in ion exchange, which could be used as fertilisers, was also reported (Robinson et al., 1998).

7.1.3. Industrial reuse

Various processes have been used to reclaim industrial water from AMD, with filtration being the most utilised (Agboola, 2019; Qu et al., 2009). Specifically, Aguiar et al. (2018) used NF to treat AMD and reclaim industrial reuse water, focusing on membrane fouling, chemical cleaning, and membrane ageing. The same group also evaluated the use of UF-NF for the treatment of gold mining effluents and industrial water reclamation, however the two-phase treatment process that was used to prevent fouling was costly (Aguiar et al., 2016). Furthermore, Ricci et al. (2015) used a sequence of MF, NF, and RO to recover H₂SO₄, separate noble metals, and reclaim high quality industrial reuse water from AMD. Finally, MD has also been employed for water reclamation from brine originating from AMD (Choi et al., 2019).

Other examined processes include adsorption and precipitation. For example, Nleya et al. (2016) used the Dowex MSA-1 ion exchange resin for H₂SO₄ recovery, while water of re-usable quality was also obtained in the acid upgrade process. However, adsorption has an overall low treatment efficiency, due to quick saturation, and waste adsorbents can cause secondary pollution, while even though precipitation manages to reclaim water suitable for discharge, the main challenge pertains to the generated sludge that requires further treatment or handling (Fosso-Kankeu et al., 2020). Solvent extract (e.g., the HydroFlex process) has been also used to treat AMD and reclaim non-potable water for use in hydraulic fracturing activities (Peterson et al., 2014; Winner Water Services, 2022).

Overall, filtration appears promising for industrial water reclamation from AMD. However, due to the high content of dissolved metals and hardness contained in the AMD, membrane filtration also faces many challenges, including scaling (Jiang et al., 2017), fouling (Aguiar et al., 2016; Pino et al., 2018) and damage due to different chemicals and pH (Pandey et al., 2012; Pangarkar et al., 2013; Warsinger et al., 2015) making the process complex and costly to apply.

7.2. Demonstration and full-scale commercial AMD beneficiation and valorisation systems

Even though mature AMD treatment plants, i.e., TRL 9, are already in operation for more than two decades, such as the Wheal Jane active treatment plant in Cornwall, UK (Coal Authority, 2022) and Veolia's 0,91 ML hourly capacity coal mine wastewater treatment plant in Pennsylvania, USA (Veolia Water Technologies, 2020), these have mainly focused on AMD decontamination rather than valorisation and/or beneficiation. More recently, Clean TeQ's 2 ML daily capacity gold mine wastewater treatment plant (TRL 9) in Victoria, Australia, which is also able to reclaim water (aquifer re-injection quality), commence

operation (Clean TeQ water, 2022b). However, even though significant strides have been made in AMD beneficiation (metals/minerals recovery), existing technologies remain mainly at bench (TRL 2-3) or pilot (TRL 4-6) scale (Table 5). Below a brief discussion on AMD treatment systems for water reclamation and metals/minerals recovery is given.

7.2.1. High Recovery Precipitating Reverse Osmosis (HiPRO)

In 2007 the eMalahleni drinking water reclamation plant (daily capacity 20 ML) was built (a demo unit had already been successfully commissioned in 2005) to treat AMD originating from several mines from the greater area of eMalahleni (Witbank), South Africa using the HiPRO process (Hutton et al., 2009). A few years later the plant's capacity was extended to a maximum of 50 ML, with the treated effluent being used as a potable water source for eMalahleni municipality (Anglo Operations, 2021). The HiPRO process is based on lime addition, ozonation, sand filtration, UF, and RO and apart from reclaiming water, gypsum and mineral rich sludge can also be recovered (Karakatsanis and Cogho, 2010). Therefore, HiPRO is a commercial technology (TRL 9) that has already operated at large scale, mainly focusing on water reclamation.

7.2.2. DESALX

Another industrial scale (TRL 9) water reclamation (aquifer re-injection) plant is Clean TeQ's gold mine drainage plant, in Victoria, Australia. This technology was first piloted on-site in 2016 and then an industrial scale (daily capacity 2 ML) treatment plant was built and commenced operation in 2019 (Clean TeQ water, 2022b). The treatment is provided by precipitation followed by a membrane free desalination process that is entitled DESALX® (Clean TeQ Water, 2022a). Specifically, first precipitation is used to reduce Sb and As concentrations (Clean TeQ water, 2022b) and then the DESALX® process, which comprise two continuous ionic filtration (CIF®) modules (i.e., ion exchange) that remove multivalent cations (e.g., Ca and Mg) and anions (e.g., SO_4^{2-}), while to avoid brine generation monovalent ions (sodium (Na) and chloride (Cl)) pass through the system (Clean TeQ Water, 2022a). Aquifer re-injection quality water is recovered (> 90%), while a small amount of As- and Sb-rich sludge is produced along with gypsum-based sludge (Clean TeQ water, 2022b). The gypsum-based sludge can be valorised (e.g., in a lime precipitation system) to produce gypsum solids (Clean TeQ Water, 2022a), suggesting that beneficiation opportunities might be also available.

7.2.3. FGR - DAF

The flocs generator reactor (FGR) - dissolved air flotation (DAF) technology has been proposed and examined for the treatment of AMD in Brazil. Specifically, a treatment train comprising heavy metal ions precipitation, flocculation, and solids (flocs)/liquid separation was examined at pilot scale ($\sim 1 \text{ m}^3 \text{ h}^{-1}$) and it was found promising for AMD treatment (da Silveira et al., 2009). Initial results from a $2 \text{ m}^3 \text{ h}^{-1}$ pilot unit installed at Carbonifera Metropolitan, South Brazil had suggested that FGR - DAF is more promising than conventional DAF (Rubio et al., 2007). In this regard, SO_4^{2-} removal from coal AMD by a DAF-precipitation-flocculation system was examined, and it was identified that the DAF of sulfate-loaded flocs has potential for coal AMD treatment and other voluminous sulfate-bearing effluents (Amaral Filho et al., 2016). Overall, the main strength of FGR - DAF technology is that is able to reclaim water that is fit for irrigation, industrial uses, or as wash water (da Silveira et al., 2009). Finally, even though that DAF is a proven technology (Amaral Filho et al., 2016) and has operated at industrial scale (Rubio et al., 2007), the FGR - DAF technology for AMD treatment and water reclamation appears to have been tested only on pilot-scale (TRL 4-6). Finally, the produced sludge, after drying, was proposed to be used as filler by the local ceramic industry (da Silveira et al., 2009).

7.2.4. HydroFlex

HydroFlex is a water purification technology that has been optimised

to treat AMD and produce non-potable water for use in hydraulic fracturing (Peterson et al., 2014). HydroFlex is based on solvent extraction (floatation liquid-liquid extraction) and it has been reported that water recovery is >95%, while sulfate levels are reduced to <100 mg L⁻¹ and the levels of common metals contained in AMD, such as Al and Fe, are also greatly reduced (<1 mg L⁻¹) (Peterson et al., 2014). Demonstration projects (TRL 6-7) of HydroFlex in the USA showed that sulfate removal was in the range 81% to 98%, resulting to a treated effluent that met the limits for environmental release in most cases, while its use, instead of freshwater, by fracking companies for hydraulic fracturing could reduce their environmental footprint (Lane, 2016). Finally, a by-product of the process is sodium sulfate, which could be recycled in the process to effectively remove barium (Ba) and strontium (Sr) from the AMD and further improve its quality and possibly open up new reuse opportunities (Peterson et al., 2014). In addition, this selective extraction process also produces a concentrated stream of sodium sulfate (6-10%) (Lane, 2016), which raises possibilities for valorisation. Finally, He et al. (2013) also examined the possibility of recovery water from AMD for hydraulic fracturing and noted that regulatory issues and engineering challenges that include compatibility with fracturing chemicals, scaling and biological growth in the well, and solid waste management might hamper the scaling up of this technology.

7.2.5. BaCO₃ solid-state reaction process

The aforementioned pilot scale (FGR - DAF (da Silveira et al., 2009)) and industrial scale (DESALX® (Clean TeQ Water, 2022a) and HiPRO (Anglo Operations, 2021)) AMD treatment processes mainly focus on water reclamation. However, they also produce a metal/mineral-rich sludge, which raise the possibility of sludge beneficiation. Therefore, here also a process where the AMD sludge is beneficiated is discussed as an example. Specifically, in a beach scale (TRL 2-3) study, the Ca- and Fe-rich dewatered AMD sludge, collected from abandoned Japanese mines, was used to synthesize M-type calcium substituted barium hexaferrite. Specifically, dry AMD sludge was wet ball-milled with barium carbonate (BaCO₃) (and hematite depending on the AMD sludge composition), then dried, crushed, homogenized, and calcined, which led to the synthesis of M-type hexaferrite (Liu et al., 2019). Among others, the synthesized M-type hexaferrite, which was prepared through the aforementioned BaCO₃ solid-state reaction process, can be used as a microwave absorber or magnetic material (Liu et al., 2019). This process also highlights the great potential of sludge that has been generated through AMD treatment processes for metals/minerals synthesis and recovery.

7.2.6. MASRO(E)

The Magnesite Softening and Reverse Osmosis (MASRO) process has been tested, at pilot scale (20,000 liters per day (LPD), TRL 4-6), for the treatment of AMD from gold and coal mines in South Africa (Masindi et al., 2019a). Specifically, this system was built at the Council for Scientific and Industrial Research (CSIR) premises, in Pretoria, South Africa (Fig. 7), and follows a multi-stage approach (integrated treatment), that includes: i) neutralization with calcined cryptocrystalline magnesite to increase pH and selectively recover the metals at varying pH gradients, ii) lime softening to remove residual sulfate and magnesium, iii) addition of soda ash to remove residual calcium and further soften the water, iv) pH balance and antiscalant, and v) RO for drinking water reclamation (Masindi et al., 2018a; Masindi et al., 2017b). Eutectic freeze crystallization has been proposed to be added to the treatment train (MASROE) to reclaim minerals from the RO retentate (Masindi, 2017b). Through this system, valuable minerals can be synthesized and recovered, which include: i) Fe-based minerals, such as Fe-hydroxide, goethite, hematite, and magnetite, b) Mg-rich gypsum from the residual sulfate, and c) limestone from the soda as softening stage, while drinking water that meets South Africa's prescribed limits can be reclaimed through RO (Masindi, 2017b; Masindi et al., 2019a). However, MASRO(E)'s main drawback is the generated sludge, which



Fig. 7. The MASRO(E) pilot plant at the CSIR, South Africa along with the recovered minerals (photos courtesy of Vhangwele Masindi, Muhammad S Osman, and Rhulani Shingwenyana).

management can be complex and costly (Masindi, 2017b; Masindi et al., 2018a; Masindi et al., 2017b; Masindi et al., 2019a).

7.2.7. TUT MBA

The Tshwane University of Technology Magnesium Barium Alkali (TUT MBA) process employs magnesium for AMD neutralization and metals removal, while sulfate is removed using barium, i.e., barium sulfate is formed. Similarly to MASRO, the produced sludge in the magnesium reactor, which is highly mineralised and heterogeneous, is costly and difficult to manage (Bologo et al., 2012). The TUT MBA system was also tested by Masindi (2017a) for the integrated treatment of AMD when using cryptocrystalline magnesite and barium chloride, and similar results with the ones obtained with the use of magnesium and barium (Bologo et al., 2012) were reported. The process has been tested on pilot scale (TRL 4-5) and therefore to further R&D is required to access its feasibility and sustainability at larger scales.

7.2.8. ABC

The alkali-based-calcium (ABC) system was developed by CSIR, in South Africa, and it entails the neutralization of AMD with lime and the subsequent treatment of the product water with barium carbonate, to attenuate the residual sulfate. To minimise cost and make the process self-sustainable minerals were recovered from the produced sludge and re-fed to the system (Geldenhuys et al., 2003; Hlabela et al., 2007; Maree et al., 2004b). In this regard, the GypSlim process was proposed to recover sulfur from the product waste (Nengovhela et al., 2007). The process was also examined at pilot scale (TRL 4-5), in the South Africa setting, and it was identified that it is possible to greatly reduce AMD's sulfate levels at a relatively short treatment time (Mulopo, 2015).

7.2.9. Adapted Savmin process

The Savmin process was initially developed by Mintek, Savannah Mining, and the Wren group for the treatment of AMD in South Africa (Bowell, 2004) and was recently adapted, at bench scale (TRL 2-3), by Abdрахmanova et al. (2019) for AMD integrated treatment. The Savmin process comprises five steps, i.e., i) heavy metals and magnesium precipitation, ii) gypsum de-supersaturation, iii) ettringite precipitation, iv) carbonation, and v) recycle of aluminium hydroxide (Bowell, 2004). The adapted Savmin process utilises lime neutralization to remove metals as hydroxides, precipitate gypsum, and form ettringite, with the added benefit of water reclamation and H_2SO_4 and gypsum recovery, while it is also possible to bind a sulfate ion to ettringite (Abdрахmanova et al., 2019). Even though the adapted Savmin process was examined at bench scale, the Savmin process is an industrial process (Sandru et al., 2017) and therefore it can be assumed that the adapted Savmin process can be rapidly scaled up to TRL 9.

7.2.10. LoSO₄ sulfate reduction technology

LoSO₄ employs a two-step precipitation process, using two different technologies developed by Veolia, i.e., Multifl and Turbomix, and is able to reduce sulfate in mine water to $<100 \text{ mg L}^{-1}$ and recover $>95\%$ of the aluminum-based salt (Veolia, 2016). Similarly to Savmin process, it includes ettringite precipitation for sulphate removal from AMD with aluminum recovery. The LoSO₄ was laboratory tested with mine water from Boliden Kevitsa mine (Cu, Ni mine) and LKAB Svappavaara mine (iron mine), both in Denmark. Results suggested that up to 95% of Al recovery is possible from AMD, but to avoid the high consumption of chemical reagents AMD with high sulphate content should be preferably used (Sandru et al., 2017). This system also provides a high quality treated effluent for reuse or discharge and can be applied in industrial scale, therefore its TRL can be assumed to be 9 (Veolia, 2016). Finally, it has also been highlighted that with high excess of Al, sulfate levels can be reduced to $\leq 50 \text{ mg L}^{-1}$ (Sandru et al., 2017).

7.2.11. Other processes

Other examined processes include the EARTH (Environmental And Remedial Technology Holdings) technology, which uses an ion exchange column to recover uranium and sulfate from AMD (Howard et al., 2009; Simate and Ndlovu, 2014); the ROC technology, which recovers valuable minerals and reclaim drinking water from AMD (Zvinowanda et al., 2014); the BOF SRO which can reclaim drinking water and recover gypsum from AMD (Masindi et al., 2017b); the ALSX (acid leaching-solvent extraction) for the recovery of rare earth oxides from AMD (Ziemkiewicz et al., 2021); and industrial treatment technologies such as the Metso Outotec sulfate removal process (Metso Outotec, 2022) which is based on Ettringite precipitation. Other technologies, such as electrochemical and ion-exchange, that could be used to valorise and/or beneficiate AMD are also discussed elsewhere (Fosso-Kankeu et al., 2020). In term of cost efficiency, Arnold et al. (2019) performed a comparative analysis of ion exchange, selective extraction and ultrafiltration, electrocoagulation, and dissolved air flotation technologies and electrocoagulation-dissolved air flotation yielded the lowest comparative life-cycle cost. These aforementioned processes,

along with their main advantages and disadvantages, are listed in Table 5.

7.3. Synthesis and recovery of valuable minerals

AMD typically include metals and sulfate (Table 1), which suggests the possibility of its beneficiation (minerals/metals recovery). However, the co-contamination of the recovered minerals has been an issue of prime concern, since this affects their purity, suggesting the need for specific treatment initiatives. Specifically, Ricci et al. (2015) proposed the use of MF and NF to produce H_2SO_4 and separate and recover noble metals from gold mine drainage. The entire process was governed by pressure-oxidation modality (Agboola, 2019). The recovery of Fe^{2+} and Fe^{3+} species has been also widely explored (Akinwekomi et al., 2017; Chen et al., 2014; Mohan and Chander, 2006; Yan et al., 2015). Apart from Fe^{2+} and Fe^{3+} , Mohan and Chander (2006) also evaluated the use of lignite for sorption of Mn, Zn, and Ca, with the adsorbed minerals being recovered as regenerants, albeit a quick saturation was observed.

Furthermore, (semi)metals have been synthesized and recovered from AMD, including goethite, hematite and magnetite (Akinwekomi et al., 2020; Akinwekomi et al., 2017). Goethite can be used as pigment in the clay, paint, and coating industries, while it has been also used as an adsorbent for (semi)metals (Basu et al., 2015; Mohan and Pittman Jr, 2007). Hematite has been employed in pigments synthesis, removal of (semi)metals and oxyanions, and for radiation shielding, amongst others (Bhateria and Singh, 2019; Duuring et al., 2018; Phuan et al., 2017). Similarly, magnetite can be used in ferro-fluid technology, storage of information, photo-degradation for organic contaminants, photo-anode, catalyst, biomedicine, controlled drug delivery, removal of water contaminants, and magnetic nanoparticles formation (Akinwekomi et al., 2017; Bui et al., 2018; Kefeni et al., 2017a; Wei and Viadero Jr, 2007).

Furthermore, Martí-Calatayud et al. (2014) evaluated the recovery of H_2SO_4 from AMD by means of a 3-compartment electro dialysis unit, where H_2SO_4 , free from Fe^{3+} species, was obtained in the anodic compartment as a result of the co-ion exclusion mechanism in the membranes. The difference in the pH and H_2SO_4 values, between the membrane phase and the external electrolyte, promotes the dissociation of complex species inside the membranes, which impede the transport of Fe^{3+} and sulfates in the form of complex ions toward the anodic and cathodic compartment, respectively. Nleya et al. (2016) examined the recovery of H_2SO_4 , using Dowex MSA-1 ion exchange resins, and their results showed that H_2SO_4 can be recovered by the resins via the acid retardation process and subsequently be upgraded to near market values (up to 70% H_2SO_4) using an evaporator. Finally, AMD sludge can be an important source for the synthesis of Ca and Fe based minerals. For example, Liu et al. (2019) were able to synthesize M-type hexaferrite from AMD sludge, which, among others, finds applications as microwave absorber or magnetic material.

7.4. Co-treatment of AMD with municipal wastewater

The co-treatment of AMD with municipal wastewater (MWW) has long attracted the interest of the scientific community. For example, Keefer and Sack (1983) examined the use of mineral resources, i.e., Fe-recovered from AMD, for MWW treatment, i.e., removal of phosphate and turbidity, and results were promising. Wei et al. (2008) highlighted that the AMD sludge can be very promising for P removal from secondary sewage effluents, while more recently Muedi et al. (2021) suggested that enriched ferric oxyhydroxide, recovered from AMD, can be used for As removal from simulated wastewater.

Nonetheless, a simpler method is to co-treat both wastewater effluents at the same time. Specifically, Johnson and Younger (2006) designed a single-stage passive system (aerobic wetland) in NE England that was able to effectively treat both mine water and secondary sewage effluents at a 3:1 ratio (the authors expected that this ratio would drop to 1:1 during high rainfall). A few years later, Strosnider et al. (2009) used

a four-stage continuous flow reactor system (at bench scale) to co-treat high-strength AMD with MWW at a 2:1 ratio (the system includes anoxic limestone treatment). This passive treatment method was in need of further optimisation but was deemed promising, since it was able to effectively treat Fe-rich AMD using minimal fossil fuel and materials input. The main drawback was that the produced sludge contained high levels of (toxic) elements/contaminants, nonetheless, these could possibly be recovered (e.g., Fe) and therefore provide a revenue source (Strosnider et al., 2009). This system was also able to eliminate fecal indicator bacteria that are typically contained in MWW (Winfrey et al., 2010) and practically remove the BOD content (Strosnider et al., 2011). The regeneration ability, after incubation, and the effect of residence time was also examined and it identified that treatment cells regeneration was possible, while only the zinc removal was observed to increase with increasing resident time (Strosnider et al., 2013). The same group used cubitainer incubations to examine the passive co-treatment (mixing) of AMD with MWW, at a 1:1 ratio and with or without the addition of limestone. It was identified that MWW has a relatively small effect on alkalinity and the addition of limestone is required for alkalinity generation and increased treatment efficiency, however, limestone also released Mn and other contaminants during co-treatment (Strosnider and Nairn, 2010).

Hughes and Gray (2013) evaluated, at bench-scale, the possibility of co-treating AMD with MWW using the activated sludge process (plug-flow and sequencing batch reactors), by examining the following scenarios: i) adding raw AMD to the activated sludge aeration tank, ii) pre-treating the AMD, prior to adding it to the aeration tank, by mixing it with digested sludge, and iii) pre-treating the AMD by mixing it with screened MWW. The optimal scenario, in terms of AMD neutralization and metals removal, was the latter, however, significant MWW alkalinity was consumed, suggesting an alkali supplement may be necessary (Hughes and Gray, 2013). A two-stage treatment, i.e., AMD-MWW batch mixing followed by anaerobic biological treatment, was also effective in phosphate (from 9 to ~100% depending on the mixing ratio) and other contaminants removal (Deng and Lin, 2013). Furthermore, a dual-chamber microbial fuel cell (DC-MFC) was proposed for AMD-MWW co-treatment (ratio 1:1) and it was shown that in a relatively short time (120 h) co-treatment was possible (Vélez-Pérez et al., 2020). Furthermore, Spellman et al. (2020a) highlighted the possibility of post-aeration co-treatment (AMD-MWW mixing, followed by sludge settling), since mild AMD would be treated without grossly imposing on the wastewater treatment plant processes, however, more research is required to scale up the process. Finally, (Masindi et al., 2021), examined the co-treatment of raw coal AMD with raw MWW and the optimal treatment ratio was 1:7 (AMD to MWW), while focus was also placed on the fate and partitioning of the chemical species that precipitated in the produced sludge. For a more thorough discussion on AMD-MWW co-treatment and its potential benefits the reader is referred to Spellman et al. (2020b).

7.5. Factors hampering AMD valorisation and beneficiation

Even though steps have been made in introducing the concept of circular economy in AMD treatment, primarily with advancements being made in water reclamation and to a lesser extent in minerals synthesis and recovery, a number of factors exist that hamper its sustainable application at industrial scale. Currently, the main limitations pertain to the quantity and purity of the recovered materials, along with specific conditions required for minerals recovery. Specifically, AMD is rich in metals (Table 1), with Fe and S concentrations as high as 8,000 and 80,000 mg L^{-1} (ppm), respectively, Al and Mn concentrations as high as 500 and 100 mg L^{-1} , respectively, while trace levels of Zn, Cu, Cd, Co, Pb, Cr, As, and Hg, among others, can also be present (Akinwekomi et al., 2020; Akinwekomi et al., 2017; Masindi, 2017b; Masindi et al., 2019a).

As such, Fe- and S-based minerals can mainly be recovered from

AMD in relatively large quantities. For example, 50 kg of Fe-based minerals were recovered when treating 3,500 L of AMD with 25 kg of calcined cryptocrystalline magnesite (Masindi et al., 2019a). Gypsum can also be recovered from AMD, attributed to AMD's large sulfate concentrations, using processes such as the GYPSLIM (Maree et al., 2013). However, due to its low purity, the recovered gypsum is of little use for industrial applications. The large Fe and sulfate concentrations are traced back to the oxidation of FeS₂, FeAsS, and other Fe-bearing sulfide minerals during AMD formation. Other chemicals species can be considered as impurities, which are incorporated to coal and gold resources during deposition processes.

Regarding the purity of the recovered materials, the highly mineralised and heterogeneous nature of AMD is the main factor that hampers the recovery of high-grade minerals, with the underlying reason being the problem of co-contamination (Masindi and Tekere, 2020). More specifically, it has been reported that the fractional and sequential recovery of mineral resources from AMD encounters massive challenges due to very fine discrepancies, in terms of pH and those overlapping regions of minerals precipitation (e.g., Fe and Al can co-precipitate), which can grossly affect the purity of the recovered minerals (Masindi et al., 2018b). For example, the gypsum piles produced when AMD is treated with lime are contaminated with Fe, (Hlabela et al., 2007). Therefore, the recovery of pure minerals from AMD has yet to be achieved as studies to date have highlighted that impurities persist across the recovered minerals (Kefeni and Mamba, 2020; Kefeni et al., 2017a; Kefeni et al., 2017b).

Finally, for the recovery of metals contained in AMD specific conditions need to be met. Essentially, the acidic pH conditions in AMD can enable the recovery of minerals/metals in a step-wise fashion, with the following pH sequence being suggested: Fe can be recovered at pH $\geq 3 - 3.5$, gypsum at pH $\geq 4 - 10$, Al at pH ≥ 6.5 , Mn at pH ≥ 9.5 , Cu at pH ≥ 7 , Zn at pH ≥ 8 , Pb at pH ≥ 8 , Ni at pH ≥ 9 , Mg at pH > 10 , and Ca at pH > 12 (Masindi et al., 2018b). However, in real-world applications, minerals synthesis is complex, requiring also complex processes, such as co-precipitation. For instance, magnetite production requires the co-precipitation of Fe³⁺ and Fe²⁺, thus cannot be typically pursued at a linear process (Akinwekomi et al., 2020; El Ghandoor et al., 2012). Furthermore, for goethite, magnetite, and hematite production, apart from using a stepwise approach, where gypsum and calcium carbonate can also be synthesized, each recovered material needs to be calcined at different temperatures (Akinwekomi et al., 2020), which from industrial perspective is a costly and complex venture.

Overall, the main challenges for AMD valorisation and beneficiation greatly depend on the technology used and can be summarised as follows:

- Generation of complex, highly mineralised, and heterogeneous sludge on the precipitation technology.
- For the adsorption technology, saturation of the adsorbent and need for frequent regeneration due to quick saturation. Poor selectivity and limited capacity for concentrated solutions such as AMD is also a main challenge. Therefore, adsorption cannot be employed as a standalone method for metals removal and recovery.
- For the filtration technology, high cost and electricity input, along with generation of highly concentrated brine, which is difficult to treat and manage.
- For integrated processes, the complexity of the overall treatment process, high costs, along with the low quantity and purity (co-contamination) of the recovered minerals.

However, the main advantage of AMD valorisation and beneficiation is that revenue from the recovered and synthesized minerals and also from the reclaimed water could offset operational costs and possibly environmental impacts (Singh et al., 2020). Furthermore, the product water could be treated to a chosen standard, e.g., for irrigation or even for drinking water, thus forming closed-loop or ZLD systems in AMD

treatment.

8. Conclusions and future research directions

8.1. Conclusions

Mining activities are notorious for their environmental impact, with acid mine drainage (AMD) being one of the main concern. AMD not only degrades the environment but can also increase water scarcity in areas with strong mining industries, such as South Africa and China. Practical and cost-effective solutions to prevent AMD formation and particularly for its sustainable industrial scale treatment have yet to be introduced. Conventional AMD treatment technologies, i.e., active and passive, cannot effectively address the problem, mainly due to sub-par performances, failures, high cost, and the generation of toxic and hazardous wastes which can lead to secondary pollution. Considering the challenges of conventional treatment, hybrid and integrated technologies have also been proposed. These maintain or combine benefits of conventional approaches while limiting drawbacks. In addition, these approaches are perceived as sustainable and eco-friendly and they might also find application in circular economy. Specifically, due to its high dissolved metals content AMD can be viewed as a valuable resource rather than as a waste, provided cost-effective minerals recovery and water reclamation methods are available.

The existing body of knowledge has highlighted that valuable minerals, such as goethite, hematite and magnetite, can be synthesized and recovered from AMD along with reclaiming clean water. Other minerals such as gypsum can be synthesized from the treatment process as well. This could open new revenue streams and possibly render AMD treatment systems self-sufficient, since revenue from the synthesized and recovered minerals and the reclaimed water could offset running costs and at the same time reduce environmental impacts. Regarding the technologies that can be incorporated in hybrid and integrated treatment systems, the most promising appears to be precipitation, while for (drinking) water reclamation various filtration technologies have been examined and particularly reverse osmosis (RO). Adsorption finds little application in minerals recovery due to quick saturation of the sorbents and regeneration requirement, while it is also non-selective.

Overall, the sequential and fractional recovery of valuable minerals from AMD, following the integrated approach, appears to be the most promising treatment approach under the circular economy context. However, the main challenges for minerals recovery and water reclamation include high costs and complexity, along with the generation of heterogeneous sludge and co-contamination in the recovered materials.

8.2. Future research avenues and directions

To make AMD treatment feasible under the circular economy concept, future research should focus on:

- Upscaling laboratory research findings on introducing circular economy concepts in AMD treatment and bridge the lab-piloting interface. This will minimize failures associated with upscaling emerging technologies and also enhance the recovery of high quality and quantity metals/minerals, along with water reclamation.
- Designing AMD treatment systems with the fit-for-purpose view and paradigm, where AMD could be treated to reclaim water that meets the standard for irrigation, industrial, discharge, and drinking purposes amongst others.
- Enhancing the purity of the recovered minerals using niche technologies that seek to improve the quality of product minerals and avoid the co-contamination problem.
- Demonstrating zero-liquid-discharge (ZLD) processes on pilot scale and upscale the freeze desalination and membrane distillation process.

- Carrying out detailed techno-economic and feasibility studies on emergent technologies for AMD valorisation and beneficiation to identify their economic viability and environmental performance.
- Assessing the environmental sustainability of emerging treatment technologies, using approaches such as life cycle assessment (LCA). By doing so, environmentally sustainable avenues for AMD valorisation and beneficiation will be identified.
- Pursuing an in-depth analysis of the market needs for the minerals that could be produced from AMD and streamline efforts on recovering materials that are in high demand in the market. This will reinforce the economic viability of circular economy in AMD treatment and avoid the production of materials with little to no commercial value.
- Preform well-informed and realistic techno-economic evaluations for passive, active, hybrid, and integrated technologies.

CRedit authorship contribution statement

V. Masindi: Conceptualization, Methodology, Writing – original draft. **S. Foteinis:** Conceptualization, Methodology, Writing – original draft, Writing – review & editing. **P. Renforth:** Validation, Writing – review & editing. **J. Ndiritu:** Validation, Writing – review & editing. **J. P. Maree:** Validation, Writing – review & editing. **M. Tekere:** Validation, Writing – review & editing. **E. Chatzisymeon:** Validation, Writing – review & editing.

Declaration of Competing Interest

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

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