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# Sustainable acid mine drainage wastewater remediation: a comprehensive review and bibliometric analysis

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Global wastewater generation is over 500 million gallons per day. Presently, these wastewaters, especially acid mine drainage (AMD), are underutilized resources with the potential for utilizing their chemical fractions and water components in economically and environmentally friendly processes, such as irrigation and energy generation. The common traditional method of AMD management is its environmentally unfriendly disposal, which often results in environmental pollution. Therefore, in this review, a quantitative, qualitative, bibliometric and systematic approach to examining experimental and peer-reviewed articles on AMD wastewater generation, properties, common pollutants, and treatment methods was conducted. Moreover, this review assesses the present remediation techniques implemented in AMD management. Finally, the most current challenges and future outlook on AMD treatment are highlighted, and valuable insights as well as recommendations are provided for actions that need to be taken to improve present AMD wastewater management, treatment, reutilization, and resource sustainability efforts.

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### Environmental significance

Wastewaters, especially acid mine drainage (AMD), are underutilized resources with the potential for their chemical fractions and water components to be reused. AMD reclaiming and remediation are desirable from economic and environmental viewpoints. Treatment strategies, especially integrated or hybrid strategies, have the potential to satisfactorily treat AMD for repurposing (such as irrigation, pretreatment and energy generation), reduce negative environmental impact, lower disposal and management cost as well as the recovery of valuable metals. The potentials of these strategies align with global sustainable goals on waste management and environmental sustainability.

## 1. Introduction

Mining industries have been a major driver of global economic development since ancient times, contributing immensely to the gross domestic product (GDP), employment, government revenue and exports in many countries.<sup>1</sup> The most valuable mineral resources and major drivers of economic growth include coal, gold, iron and copper.<sup>2,3</sup> Unfortunately, the socioeconomic benefits of the mining sector often come at the detriment of the environment since mining activities are notorious for the generation of lethal by-products and waste materials, such as overburden and tailings.<sup>4</sup> Overburden occurs during the stripping of the mining area, comprising soil and rocks with no current economic interest, while the tailings

result from mineral processing and are deposited in wet dams close to the mining area.<sup>5</sup>

These environmental liabilities (overburden and tailings), along with the exposed mined areas, the tunnels and shafts of abandoned and/or active mines, can react with water to form neutral, basic, or, predominantly, acidic leachates.<sup>5</sup> Acid leachate, mainly known as acid mine drainage (AMD) (acid and metalliferous drainage), causes the most detrimental environmental concerns and effects.<sup>3,6,7</sup> Acid mine drainage negatively impacts the receiving environment (Fig. 1) mainly by increasing the solubility of various chemical species and altering the ambient pH of the affected areas.<sup>8</sup> Sometimes, the minerals contained in AMD precipitate at the bottom of receiving waterbodies, altering their chemical composition.<sup>9</sup> Such alternations could stimulate changes in the nutrient loading rates of biostimulatory nutrients, oxygen consuming materials, and toxins, thus rendering the health quality and productivity of aquatic ecosystems unfit for human consumption and agricultural activities.<sup>3,10</sup>

Of all the AMD components, heavy metals such as arsenic (As), chromium (Cr), iron (Fe), aluminium (Al), copper (Cu), zinc

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(Zn), lead (Pb), molybdenum (Mo), and nickel (Ni) are of major concern.<sup>3</sup> AMD provides niches for extreme-pH and toxic-metal tolerant microbial diversity, such as *Acidithiobacillus ferrooxidans*, *Leptospirillum ferrooxidans*, and *Acidithiobacillus thiooxidans*.<sup>3,11,12</sup> These groups of microorganisms play vital roles in accelerated oxidation of iron and sulphur.<sup>12</sup> These pollutants can trigger ecotoxicological, carcinogenic, mutagenic, and teratogenic effects upon exposure.<sup>13</sup> Specifically, heavy metal contamination might have a devastating impact on the ecological balance of aquatic ecosystems. One of the major problems associated with the persistence of heavy metals in aquatic environments is the potential for bioaccumulation and biomagnification of heavy metals in aquatic organisms like pisces (fishes).<sup>14,15</sup> Fishes can accumulate large amounts of metals from water, leading to adverse effects on aquatic systems.<sup>15,16</sup> Fishes are at the top of the aquatic food chain and can be used to evaluate the health of aquatic ecosystems.<sup>14,15,17</sup> The aspect of human health associated with the consumption of heavy metal-contaminated fish is also a public health concern.

The implementation of adequate technologies for long-term AMD remediation by degrading acidity and removing toxic metal ions is, therefore, very important. Conventional technologies, such as membrane separation and ion exchange, have been extensively used for AMD treatment.<sup>18,19</sup> However, these technologies are expensive, have variable efficacies in contaminant removal and generate by-products, like sludge and brines, which can cause secondary contamination if poorly managed.<sup>20</sup>

Hence, there is a global drive to seek energy efficient, eco-friendly and sustainable treatment methods, such as nanotechnology and biological strategies. Nanotechnology has attracted significant research interest as a potential alternative to traditional treatment methods.<sup>21</sup> Nanomaterials present several advantages, including high surface area, easy recovery, and enhanced physical and chemical properties, due to their nano-scale features.<sup>21–24</sup> Biological strategies involving the application of microorganisms, plant biomass, or plant-microbe interactions have increasingly gained attention as green and low-cost technologies for AMD treatment.<sup>25,26</sup> The tendency of plants and microbes to solubilize, adsorb and



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Fig. 1 A mining site in Mpumalanga (A), drainage basins (B–D), and soil around drainage basins (E).

precipitate contaminants in AMD makes them suitable agents for bioremediation.<sup>27,28</sup> Merits such as less energy demands, effectiveness, eco-friendliness and sulfate recovery capabilities make the biological approach a promising technology.<sup>29–31</sup> Moreover, integrated technologies have been explored for AMD

treatment. Such approaches involve sequential or stepwise treatment for AMD beneficiation and valorisation.<sup>3</sup> Integrated systems have the potential to maintain cleaner wastewater production, effectively prevent environmental pollution and provide sustainable AMD management. However, these systems are costly to install and operate.<sup>3</sup> Thus, this review provides insights into global AMD generation and its diverse consequences, with an emphasis on South African AMD generation and management. It also discusses the applicability of various AMD management strategies, merits, demerits, and outlooks.

Table 1 Predominant metal sulphides for acid mine drainage production.<sup>30</sup>

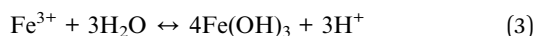
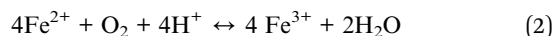
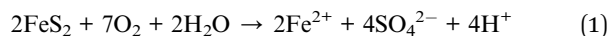
Metal sulphide	Formula
Pyrite	FeS <sub>2</sub>
Millerite	NiS
Arsenopyrite	FeAsS
Covellite	CuS
Chalcopyrite	Cu <sub>2</sub> S
Galena	PbS
Sphalerite	ZnS
Molybdenite	MoS <sub>2</sub>
Marcasite	FeS <sub>2</sub>

## 2. Formation and chemical composition of AMD

Generally, AMD originates from the accelerated oxidation of sulphide-bound minerals (metal sulphides) from both active and abandoned mines (primarily coal and gold) worldwide.<sup>32,33</sup> Various metal sulphides are present in AMD (Table 1), with the



predominant ones being pyrite and marcasite [32. The chemical processes underlying the oxidation of pyrite and formation of AMD are well documented, consisting of the steps illustrated in eqn (1)–(4).<sup>30,34,35</sup> It begins with the hydro-geochemical weathering of sulphide bearing minerals, such as marcasite, arsenopyrite and pyrite.<sup>30,35</sup> The iron sulphate salts generated can dissolve in groundwater, thus increasing the amount of dissolved iron salts. Consequently, the pH may drop below 2, and if there are iron-stone beds present in the strata, the iron level may reach up to 2000 mg L<sup>-1</sup>.<sup>30</sup> The oxidation of pyrite occurs through a series of pathways that require surface interactions of dissolved oxygen and Fe<sup>3+</sup> ions, which form dissolved iron, sulphate and hydrogen ions (eqn (1)). However, the rate of natural oxidation of pyrite and acid production is dependent on various factors, including mineral variability and oxygen and water availability.<sup>30,35,36</sup> Indeed, the process is not exclusively chemical but also biochemical and biological (involving certain bacteria, such as *Acidithiobacillus ferrooxidans*, *Acidithiobacillus thiooxidans*, and *Leptospirillum ferrooxidans*), synergistically playing crucial roles. These bacteria obtain energy from the oxidation of iron and sulphur compounds.<sup>35</sup> Under certain oxidation conditions (pH > 3.5, and the presence of certain microbes), the iron(II) ion (ferrous iron) released in eqn (1) can be further oxidised to iron(III) ions (ferric iron) (eqn (2)). Under oxygen-deprived conditions, eqn (2) does not proceed until the pH increases to 8.5. This shows that eqn (2) is often the rate limiting step in pyrite oxidation (since the conversion of ferrous to ferric is too slow at pH ≤ 5). Moreover, at pH 2.3 to 3.5, ferric iron formed in eqn (2) may precipitate as Fe(OH)<sub>3</sub>, lowering the pH and the amount of Fe<sup>3+</sup> in the process.<sup>30</sup> Furthermore, under extremely acidic conditions (pH < 2), Fe(OH)<sub>3</sub> from eqn (3) is thermodynamically unstable. The remaining Fe<sup>3+</sup> is eventually utilised for additional pyrite oxidation (eqn (4), plunging the pH of AMD into a highly acidic state.<sup>37</sup>



Due to the high acidity created by excess H<sup>+</sup> and sulfuric acid in AMD, a large number of metals and mineral deposits are dissolved by AMD, resulting in an acidic and metalliferous toxic effluent.<sup>38</sup> Evidently, AMD is usually characterized by high acidity (pH 2–4), sulphate (1000–30 000 mg L<sup>-1</sup>) and about 100–2000 mg L<sup>-1</sup> iron ions.<sup>39</sup> Other heavy metals, such as arsenic (As), chromium (Cr), iron (Fe), aluminium (Al), copper (Cu), zinc (Zn), lead (Pb), molybdenum (Mo), and nickel (Ni), make up the chemical composition of AMD.<sup>7</sup> However, the chemical composition of AMD varies from mine to mine and across the world (Table 2).

### 3. Global acid mine drainage (AMD) generation

AMD is discharged from about 20 000–50 000 mine sites worldwide, with major mining operations from China, South Africa, Canada, Australia, and the United States.<sup>48,49</sup>

The United States (U.S.) is a major global coal producer, having recoverable coal reserves of 252733 Mt and total production of 756.2 Mt coal in 2018.<sup>50</sup> Annually, the U.S. records an estimated 17–27 billion gallons of AMD generated primarily from coal mines.<sup>50</sup>

Australia is among the world's leading exporters of minerals (majorly coal).<sup>51</sup> For instance, Australia recorded estimated mineral exports of AUD141 billion, approximately 9% of its gross domestic product in 2015.<sup>39</sup> Having the third-largest coal reserves globally, Australia is the fifth-largest producer and third-largest exporter of coal, generating about half of its electricity from coal. These huge mining activities result in AMD drainage discharge and other environmental threats.<sup>51</sup>

In Canada, mining remains an important economic sector, contributing 5% to the total gross domestic product in 2021.<sup>52</sup> Despite this economic contribution, the mining of valuable minerals causes significant damage to the environment *via* toxic acid mine drainage.<sup>53</sup> For instance, a Canadian survey of metal mines and industrial mineral tailings indicated that there are 750 billion tonnes of waste stone and 1.9 billion tonnes of tailings with the potential to generate AMD.<sup>54</sup> Similarly, mountaintop coal mines in the Elk Valley of British Columbia, Canada, have been reported as the primary sources of selenium in the Elk River and its tributaries.<sup>55,56</sup>

Table 2 Chemical components of acid mine drainage from typical mines across the world<sup>a</sup>

Country	Typical mine	pH	Fe	Zn	Cu	Mn	Al	SO <sub>4</sub> <sup>2-</sup>	References
Australia	Mount Morgan gold	2.70	66	55	65	245	2317	29 547	40
Brazil	Coal	2.33	611.38	62.65	—	37.98	269.37	7410.40	41
Canada	Gold	2.39	788	0.25	3.42	19.40	310	4520	42
Chile	Copper	2.50	627.50	—	2298	224.50	1139	14 337	38
China	Polymetallic	2.50	2490	500	2670	6590	—	24 530	43
Korea	Taejeong coal	3.28	186	—	—	13	40	1950	44
SA	Mpumalanga coal	2.00	8000	—	—	75	300	30 000	45
Spain	Rio Tinto	2.60–2.80	1824	557	184	329	2830	24 700	46
USA	Elizabeth copper	3.30	123	—	—	2.60	13	1200	47

<sup>a</sup> — = Below the detection limits, metal concentration in mg L<sup>-1</sup> and SA = South Africa.



Moreover, in China, AMD accounts for more than 50% of the total mine drainage produced annually.<sup>54</sup> Despite the reduction in active coal mines,<sup>57</sup> China still generates over 3.5 billion tons of AMD annually.<sup>54</sup> Additionally, closed coal mines in China have been reported to cause a series of water environmental problems, such as the deterioration of the water quality of environmental water bodies.<sup>54</sup>

## 4. African acid mine drainage (AMD) generation

Africa is endowed with rich mineral sites with about 30% of the global mineral reserves. However, as most African countries lack friendly terrains and sophisticated infrastructure, the continent's rich minerals remain largely unexplored.<sup>58</sup> Presently, Africa's production contributes only about 8% of global mineral production despite its rich mineral deposits. Therefore, Africa represents mineral-fertile soil for mining investors. In recent times, the African rich mineral reserves have been attracting a significant foreign investment mainly from Russia, China, Canada, Australia and Brazil, resulting in a surge in industrial mining, especially coal and gold.

Although increasing industrial mining activity can improve health and livelihood through local industrial development, it could also have detrimental impacts on human health and the environment.<sup>58</sup> Mining activities remain a significant source of environmental pollution on the continent (particularly in countries and cities where mining activities are currently explored) through the generation and discharge of toxic AMD pollutants, affecting aquatic ecosystems, agriculture and human health.<sup>59</sup> For instance, the expansion of large-scale gold mining in Ghana (1997–2005) was responsible for the decrease in agricultural activities and productivity in the gold mining surrounding areas.<sup>60</sup> Similarly, an environmental analysis of industrial mining activities in Sub-Saharan Africa revealed degradation of the sewerage system and pipe water supply in the medium and long run due to mining discharge.<sup>61</sup> Human exposure to toxic pollutants, such as heavy metals, through the consumption of contaminated water is another major concern in Africa and Sub-Saharan Africa (where only 24% of the population has access to safe drinking water).

Overall, the countries with major mining activities in Africa with potential for huge volumes of AMD discharge include Nigeria, Ghana, Guinea, Burkina Faso, Sierra Leone, Democratic Republic of the Congo (DRC), Liberia, South Africa, Ivory Coast, Niger, Zimbabwe, Tanzania and Zambia. Judging by the large mineral reservoir, capital, technology and technical know-how, South Africa holds significant mineral wealth and stands out as a global mining powerhouse on the African continent.

### 4.1. South African acid mine drainage (AMD) generation

South Africa is one of the most resource-rich nations on the African continent, possessing an abundance of minerals, including coal, gold, platinum and diamonds.<sup>62</sup> For instance, the Witwatersrand Basin in Gauteng Province represents the world's largest gold deposit, yielding more than one-third of the

gold ever produced.<sup>63</sup> Similarly, South Africa is the 7th biggest coal producer globally.<sup>64</sup> In 2022, South Africa's coal mine industry contributed 231 Mt of the 8.6 Gt generated worldwide.<sup>65</sup>

These minerals have positioned the mining industry as a key player in South Africa's economic trajectory, supporting its industrial growth, socio-economic development and trade relationships.<sup>66</sup> The South African mining sector contributes about eight percent (8%) of the nation's gross domestic product.<sup>66</sup> Although the mining industry has formed the foundation and remains the backbone of the South African economy, the operation of mining sites severely threatens water resources and human health. Mining activities (mainly gold and coal mining) generate lethal by-products and waste materials, with AMD being the most notorious.<sup>4,63</sup> AMD is prominent in many uncontrolled mine sites, pyrite-containing tailings, and overburdens from gold and coal mines.<sup>67,68</sup> The complexity of the threats posed by this environmental menace in various localities in South Africa is well documented.<sup>3,4,8,63,64,66,67,69</sup> For instance, in the Witwatersrand Basin alone, over 270 tailings storage facilities (TSFs) store seepage from mine waste dumps, with the majority posing serious threats to both groundwater and surface water quality.<sup>63</sup> Additionally, toxic pollutants in the Klip River (the principal drainage of the southern portion of Johannesburg) resulting from Run-off from the Central Rand Goldfield discharges have been reported.<sup>63</sup> The Klip River wetland drains into the prolific Vaal River, which supplies about 23% of the South African population with potable water.<sup>70</sup> Moreover, the tailings dams in this gold mining region possess major physiochemical characteristics of AMD and have very high levels of heavy metals and oxides, with very low pH ranging from 2.4 to 5.8. These dams discharge about 202 million litres of AMD per day.<sup>71</sup>

Another prominent province in South Africa with huge mining activities is the Mpumalanga Province. Most active mines in this province discharge AMD effluent into the Olifants and Vaal River basins, while a considerable number of mines in the Witbank, Ermelo, and Highveld coalfield regions have been abandoned or collapsed (Fig. 1).<sup>8,64</sup>

It is estimated that about 62 ML per day of mine water is generated from coal mines in the Highveld Coalfield, and approximately 44 ML per day is discharged into the upper Olifants River catchment.<sup>72</sup> The discharged effluents are characterised by very high levels of sulphate ( $\text{SO}_4^{2-}$ ) and iron, along with other heavy metal ions.<sup>21</sup> These lethal effluents are known for the notorious pollution of the Vaal and Olifants River basins, with the Olifants catchment having some of the poorest water quality in South Africa.<sup>73</sup> Further indications of this problem are the pollution of the local rivers, rising salinity and acidity of the water in the Middelburg and Witbank dams.<sup>73</sup> Consequently, irrigation with water from these aquatic environments has affected agricultural yields, resulting in low productivity.<sup>73</sup>

Unfortunately, the future potential for AMD generation and its consequences in the country are expected to be far more severe. This is because mining sites continuously generate AMD even centuries after commercial mining exploration.<sup>6</sup> The



implementation of adequate technologies for long-term AMD remediation is therefore very important.

#### 4.2. Potential adverse effects of AMD and the toxic effects of heavy metals

In mining areas, the seepage/release of AMD effluent into the surrounding aquatic ecosystem is a common occurrence. Consequently, the concentration of heavy metals in such water environments exceeds safety standards, resulting in lethal water pollution, especially in local ecology.<sup>74</sup> For instance, the U.S. records an estimated 17–27 billion gallons of AMD generated primarily from coal mines.<sup>50</sup> The discharged AMD has been reported to contaminate over 20 000 kilometres of freshwater resources, costing about 3.8–\$72 billion for reclamation of these waterways.<sup>50</sup> Moreover, AMD from the eastern United States coal mines was reported to have contaminated more than 6000 km of rivers, while in the western United States, more than 8000–16 000 km of rivers were severely affected.<sup>75</sup>

In addition, acid sulphate soil contamination (possibly caused by AMD) in Australia occupies about 2 15 000 km<sup>2</sup> of the total land area, with 58 000 km<sup>2</sup> and 1 57 000 km<sup>2</sup> identified along the coastline and inland, respectively, as reported.<sup>39</sup> In Australia, the cost of treating AMD from abandoned mines is high, with an estimated value of about \$650 million per year.<sup>75</sup> Hence, the concerns about AMD discharge and other environmental threats have become a critical issue for Australia's mining community. Moreover, in China, AMD accounts for more than 50% of the total mine drainage produced annually.<sup>54</sup> Despite the reduction of coal mines from over 80 000 to around 5800 by the end of 2018,<sup>57</sup> China still generates over 3.5 billion tons of AMD annually.<sup>54</sup> Even closed coal mines in China have caused a series of water problems, such as the deterioration of water quality.<sup>54</sup>

The release of wastewaters, like AMD, to the larger environment or local ecology could disrupt the entire functioning of the ecological system with the potential of a harsh devastating impact on the environment.

Besides, there is an additional cost associated with potable water production. The deteriorating water quality increases water treatment costs as well as social, economic and health repercussions.<sup>73</sup> For example, treated mine water was estimated to be about ten percent (10%) of the daily potable water supplied by Rand water to the municipal authorities for urban distribution in Gauteng Province at R3000 per megalitre;<sup>21</sup> with South Africa being a water-scarce country, this is of great concern.<sup>76</sup>

Furthermore, a recent report showed that contaminated waters, such as AMD, have led to yearly occurrences (approximately a billion cases) of different sicknesses and diseases, especially in less (low-income) countries.<sup>77</sup> These cases occur because contaminated waters have the potential to support waterborne pathogens (like *Escherichia coli*, *Schistosoma* spp., *Salmonella* spp., and *Vibrio cholerae*), causing diseases such as schistosomiasis, diarrhoea, cholera, typhoid, fever, giardia, dysentery, and hepatitis A.

The potential adverse and toxic effects of some heavy metals in AMD have been reported in the literature.<sup>77,78</sup> For example, zinc is prominently involved in cell death in the brain, and cytotoxicity, although short-term exposure to the toxic form of barium, could lead to clinical effects, like brain swelling, muscle weakness, and damage to the heart, liver and spleen. Cadmium has a long half-life in the cell and can easily accumulate in amounts that cause symptoms of poisoning, resulting in acute and chronic toxicity. Common symptoms of severe cadmium exposure include muscular pain, lung cancer, kidney damage, pulmonary emphysema, osteomalacia and osteoporosis.

Exposure to high levels of lead could cause critical clinical effects, such as abdominal pain, constipation, fatigue, sleeplessness, vomiting, diarrhea, convulsions, coma or even death. Moreover, in young children, it has the potential to damage nervous connections, blood damage and brain disorders.<sup>79</sup> Mercury exposure has the potential to cause brain and liver damage in living organisms.<sup>80</sup> Moreover, elemental manganese in drinking water is the biggest concern since manganese in food is not easily absorbed. Manganese toxicity symptoms include neurological problems, muscle cramps, respiratory effects, reproductive and developmental effects, fatigue and aggressiveness. Generally, manganese is released into the environment through the erosion of rocks and soils, mining activities, industrial waste and anthropogenic activities.

## 5. Bibliometric analysis of acid mine drainage

The schematic representation of the bibliometric workflow for retrieving information about the research topic is shown in Fig. 2. The bibliographic data were obtained using the Dimensions database on June 9, 2025. The data are focused on acid mine drainage (AMD) wastewater remediation. The search approach and data collection were conducted in three stages: (1) literature search and data selection; (2) screening and extraction of data; and (3) bibliometric analysis.

A total of 630 documents published from 1994 to 2025 were obtained from the Dimensions database after the screening, selection, and extraction of documents relevant to the research topic. The exported bibliographic data were analyzed and visualized using VOSviewer software version 1.6.20 and Microsoft Excel 2019, Office 365 version 16.0.

### 5.1 Document types of acid mine drainage

The classification of publication types in relation to AMD wastewater remediation is presented in Fig. 3. A total of 630 retrieved publications were categorized into six publication types: article (542 records), chapter (69 records), preprint (6 records), proceeding (6 records), edited book (4 records), and monograph (3 records) (Fig. 3). Article accounted for 86.03%, followed by chapter with 10.95%, preprint with 0.95%, proceeding with 0.95%, edited book with 0.95%, and monograph with 0.48%. The remarkable increase in the number of articles indicates a growing interest in research activity, particularly in the field of mining.



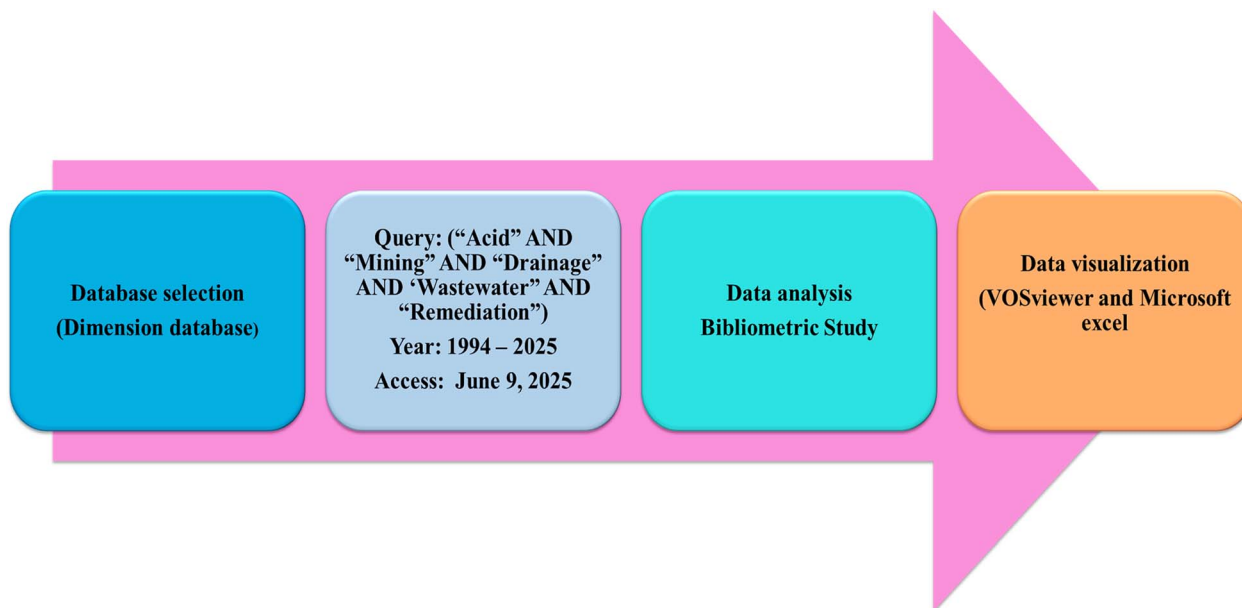


Fig. 2 The process of data collection in the dimension database.

Moreover, this trend reflects the scientific community's recognition of the topic's relevance and pressing societal and environmental challenges that require urgent attention. Furthermore, the increase in articles suggests increased funding opportunities, interdisciplinary collaboration, and advancements in research methodologies. Additionally, the increase in scientific article publications could enhance the visibility of the field, attract more researchers, and contribute to

the development of more refined novel technologies, and practical solution applications.

## 5.2 Trends in article publications on acid mine drainage

Fig. 4 illustrates the global annual distribution of scientific publications related to AMD remediation from 1994 to 2025. The data show a fluctuating trend in research output over the

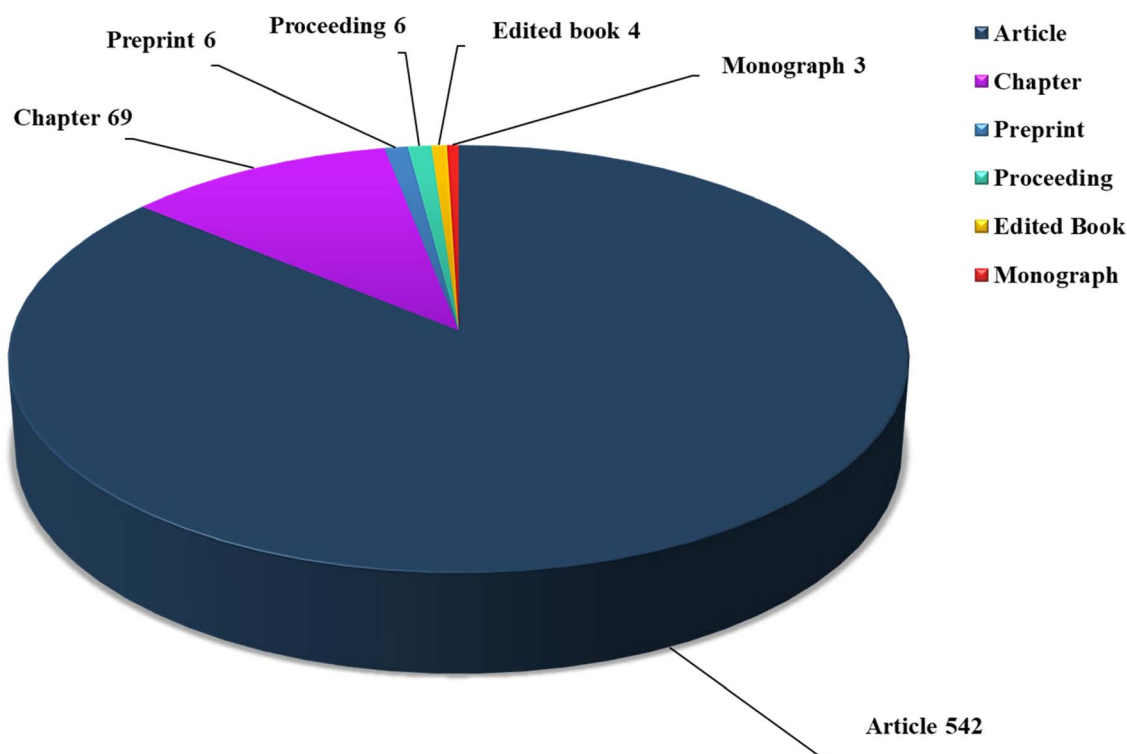


Fig. 3 Types of documents in the dimension database.



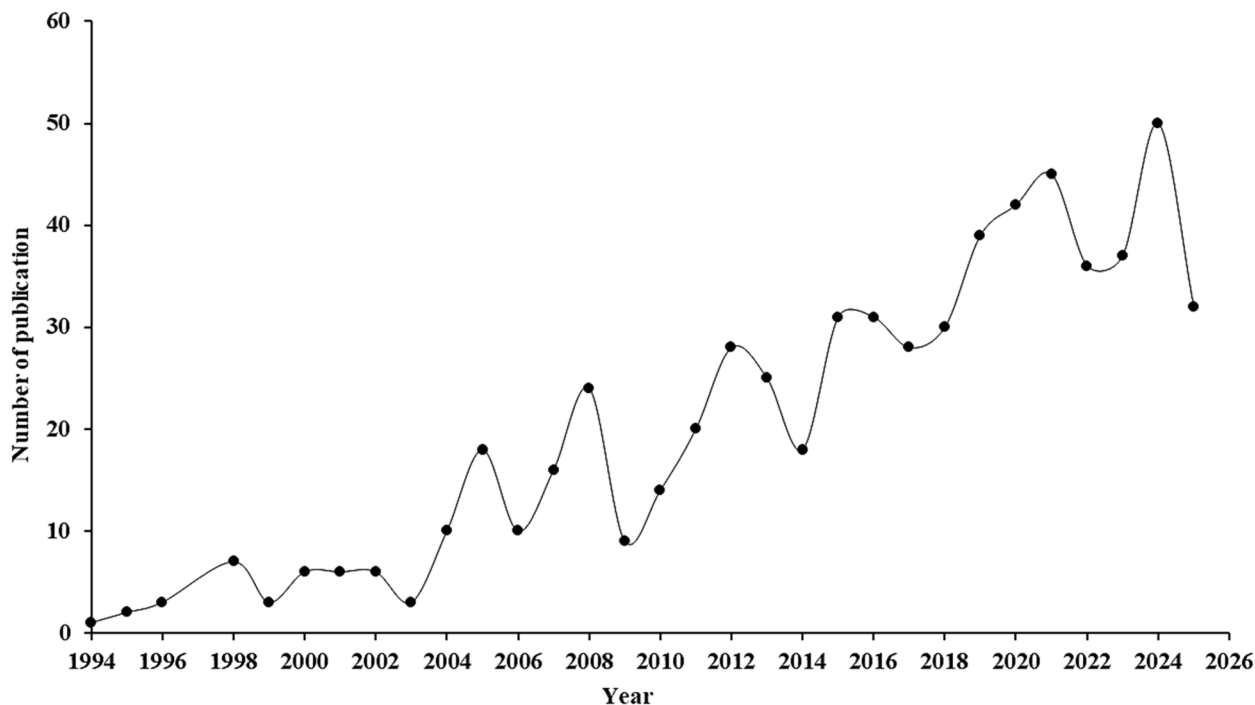


Fig. 4 Scientific publications obtained from 1994 to 2025 related to acid mine drainage wastewater remediation.

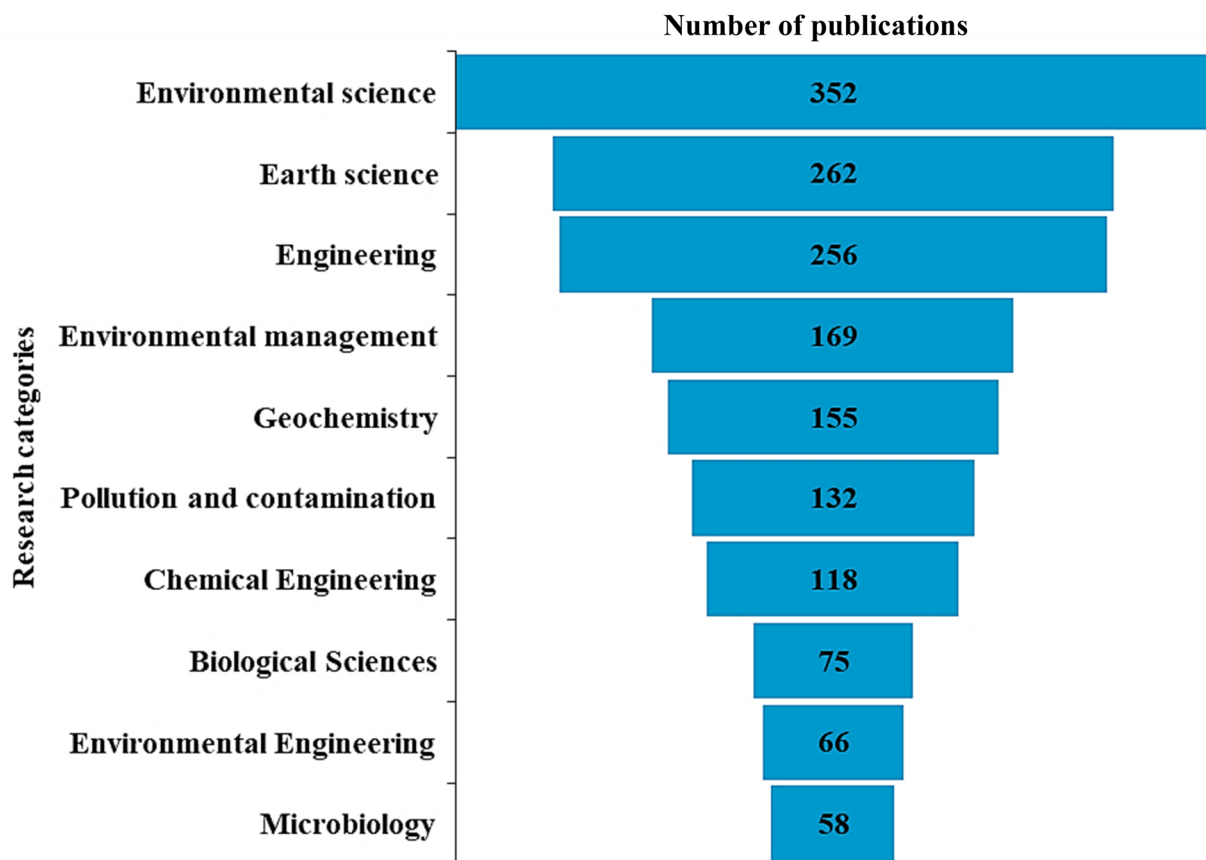


Fig. 5 Top ten research categories from 1994 to 2025 based on scientific publications on acid mine drainage wastewater remediation.



years, reflecting variations in research interest, shifts in scientific focus, funding priorities, and advancements in technology within the field. Notably, the year 2024 recorded the highest number of publications, indicating a surge in research activity and a growing global interest in AMD remediation strategies.

This increase may also be due to visibility of the research field, environmental concerns, stricter regulatory frameworks, development of innovative treatment technologies and practical applications, as outlined above. Additionally, the rising number of publications in recent years suggests that this research field is still expanding rapidly and is expected to continue growing in the future, driven by increasing environmental concerns and the demand for sustainable treatment solutions.

### 5.3 Publications by research categories on acid mine drainage

Fig. 5 illustrates the distribution of the top ten research categories in relation to AMD wastewater remediation. Of the top ten research categories, environmental science had the highest number of research articles (352), followed by Earth science (262) and engineering (256), accounting for 21.42%, 15.95%, and 15.58% of the total articles published, respectively. Other research categories with low publication numbers included biological sciences, environmental engineering, geology, ecology, resource engineering and extractive metallurgy.

This distribution of research categories shows current trends in the present field of research. Additionally, it showed that scientists are exploring different research categories using interdisciplinary approaches to solve the complex challenges posed by AMD wastewater.

The dominance of environmental science indicates a strong attention to understanding and mitigating the impacts of AMD,

while earth science and engineering underscore the efforts to address geochemical processes and develop technological solutions for remediation. The lower publication numbers in biological sciences and related fields suggest that while these areas are emerging, they remain underexplored, presenting opportunities for future research.

Similar findings reported by Ma *et al.*<sup>81</sup> showed the dominance of environmental science and geosciences in AMD remediation, showing the growing environmental concerns and geochemical implications of AMD. Likewise, Zhang *et al.*<sup>82</sup> reported that interdisciplinary research, particularly involving environmental engineering, microbiology, and hydrogeology, is increasingly being employed to develop innovative and sustainable AMD treatment technologies. These studies support the idea that the field is evolving through integrated efforts to solve the environmental impact of mining activities.

### 5.4 Publications in countries, collaborations and sources on acid mine drainage

Research articles on AMD wastewater remediation have been published in sixty-one countries. Of these, thirty-nine met the predetermined minimum threshold of three publications per country. Furthermore, from the thirty-nine successful countries, the total link strength of the bibliographic coupling for each country was calculated, and countries with the highest total link strengths were selected.

Fig. 6 shows the global publication trends across various countries and each country's dominance in relation to AMD wastewater remediation. The results reveal that Africa, North America, South America, Asia, Europe, and Oceania contributed to the global research output on AMD. Among these regions, North America and Asia produced the highest number of

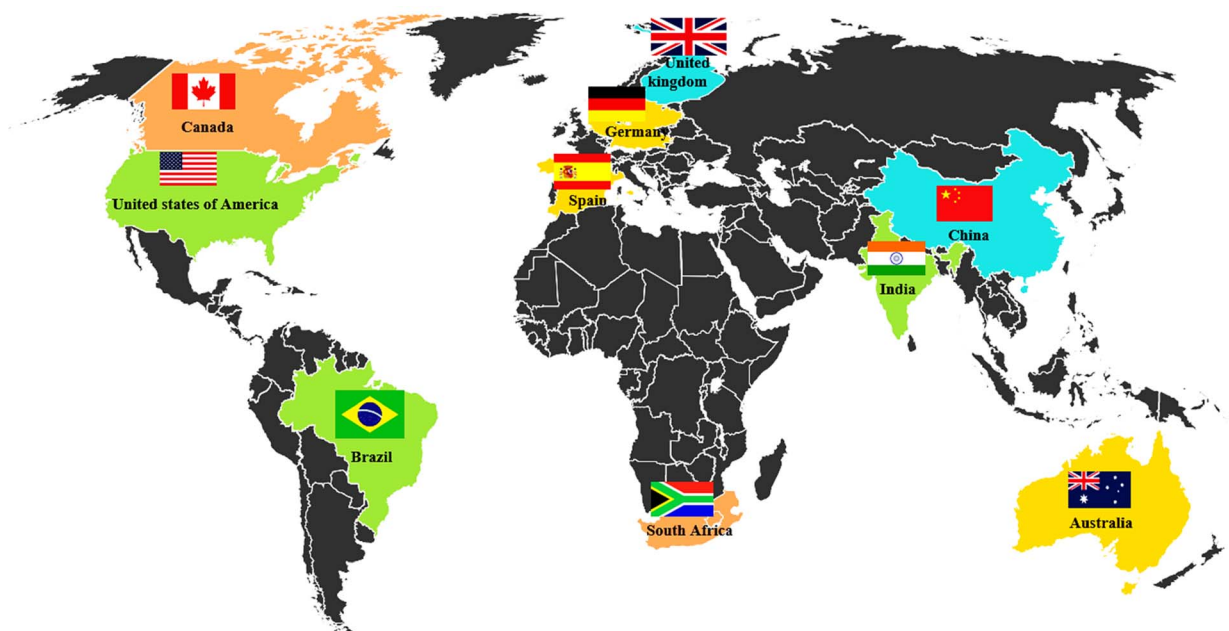


Fig. 6 The 10 most productive countries publishing articles related to acid mine drainage wastewater remediation.



publications. The United States (US), China, Spain, South Africa, Australia, the United Kingdom, India, Canada, Germany, and Brazil emerged as the top ten most productive countries in this field.

As illustrated in Fig. 6, the US ranked first with 128 publications, 4870 citations, and a total link strength of 22 294. This was followed by China with 87 publications and Spain with 53 publications, represented in light green, teal blue and light gold shades, respectively, on the world map. The high volume of publications from these countries shows their constant commitment to addressing AMD-related environmental challenges *via* research and innovation. Similar findings were reported by Zhang *et al.*,<sup>83</sup> who observed that the US and China consistently led in AMD research output, particularly in the development of passive and active treatment systems. Likewise, a bibliometric analysis by Chen and Ma<sup>84</sup> confirmed that the most influential research on AMD remediation originated from countries with significant mining activities and advanced environmental research infrastructure. These findings align with the results presented in this study and underscore the global recognition of AMD as a crucial environmental concern.

The collaboration networks among countries contributing to scientific publications in the field of AMD wastewater remediation are shown in Fig. 7. A total of six clusters, 696 link and a total link strength of 79 890 were observed among the countries. The size of the circles corresponds to the volume of scientific publications produced by the respective countries, while the lines represent the cooperation networks among the

various countries. The shorter distance between the two circles indicates strong collaboration between countries.

As shown in Fig. 7, the medium circles indicate low publications in the various countries. Very low publications, as evidenced by the small circles, were observed with Iran, Nigeria, Russia, Czechia, Nederland, Colombia, Morocco, Mexico, Ecuador, Switzerland, Denmark, Ireland, Turkey, Greece, Pakistan, and Romania. The larger circle observed with the United States, China, Spain, the United Kingdom, South Africa, Australia, India, Canada, Germany, and Brazil indicates that article publication is high and that these countries play crucial roles in contributing to global research networks in this field of AMD.

The US, China, and Spain collaborated with thirty-eight countries. Australia and Germany also partnered with thirty-eight countries, and both initiated a new collaboration with Greece. South Africa and Nigeria also established collaboration networks with 37 countries. Other countries, such as the United Kingdom, India, Canada, Brazil, Iran, Romania, South Korea, France, Chile, Mexico, Indonesia, Malaysia, Peru, Italy, Russia and Portugal, also established strong collaborations with each other and other countries. Moreover, the strong collaborative link strengths observed among these countries indicate strong international partnerships, which further enhanced the global research network, contributing to knowledge dissemination and advancing sustainable remediation technologies.<sup>85,86</sup>

Research articles on AMD wastewater remediation have been published in 259 sources. Of the 259 sources, forty-seven met the predetermined minimum threshold of three publications

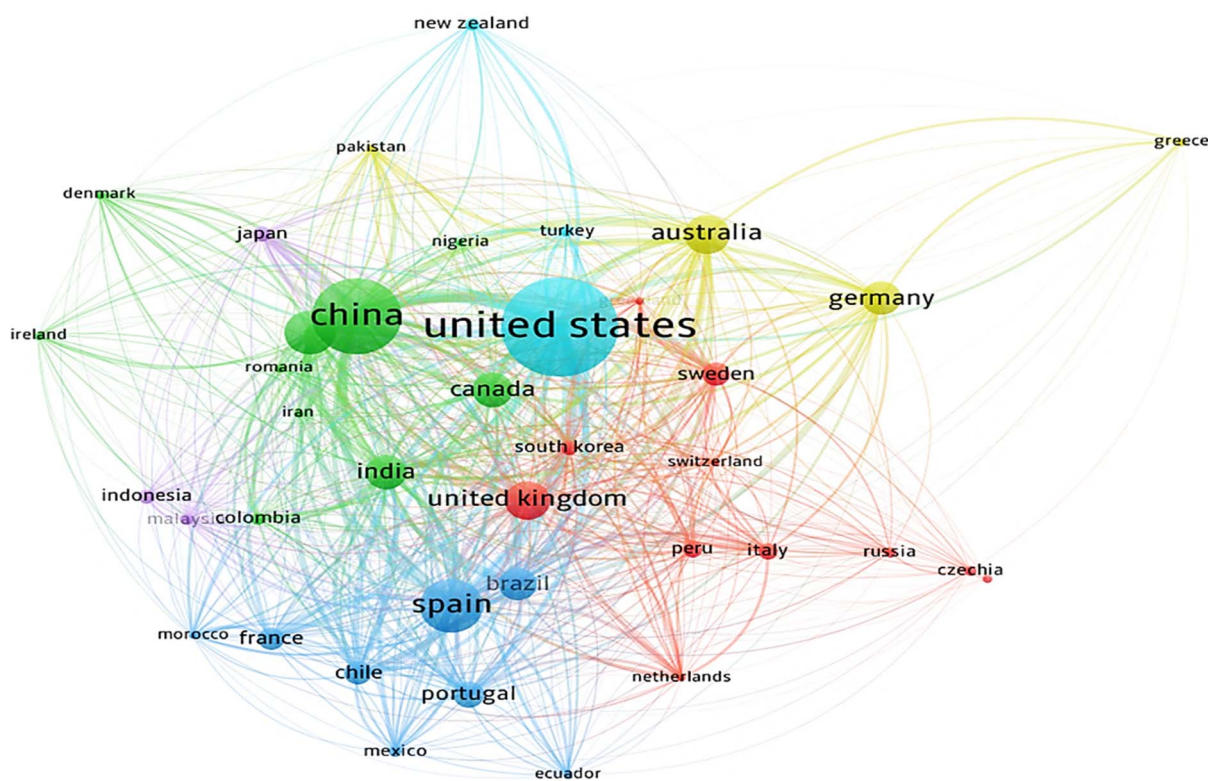


Fig. 7 Collaboration networks among countries from 1994 to 2025 that published articles related to acid mine drainage wastewater remediation.



Table 3 The top 10 sources that published articles between 1994 to 2025 on acid mine drainage wastewater remediation

Journal source	Publications	Citations	Total link strength	Publisher
Environmental Science and Pollution Research	40	886	4780	Springer
Chemosphere	26	1585	3548	Elsevier
The Science of the Total Environment	25	2831	3572	Elsevier
Journal of Hazardous Materials	18	1575	2393	Elsevier
Journal of Environmental Management	17	542	2325	Elsevier
Environmental Monitoring and Assessment	16	250	853	Springer
Water Research	14	681	1283	Elsevier
Environmental Pollution	13	713	1203	Elsevier
Mine Water And The Environment	12	176	575	Springer
Water, Air, & Soil Pollution	12	331	1976	Springer

per source. The top ten most productive sources publishing articles relating to the research topic are presented in Table 3.

*Environmental Science and Pollution Research*, including *Chemosphere* and *The Science of the Total Environment*, were the most productive in publishing articles related to AMD wastewater remediation. More interestingly, it is worth noting that *Environmental Science and Pollution Research* was the most prolific publishing source with forty publications, 886 citations and a total strength of 4780, underscoring its impact in the field. Similarly, Zhang *et al.*<sup>87</sup> and Wang *et al.*<sup>88</sup> identified *Environmental Science and Pollution Research* and *The Science of the Total Environment* as the main publishing sources of AMD environmental remediation research after bibliometric analyses.<sup>87,88</sup> Furthermore, most articles were published by Elsevier, showing the publisher's strong reputation and influence in environmental science.<sup>85</sup>

### 5.5 Analysis of authors, coauthors and affiliated institutions on acid mine drainage

Table 4 presents the authorship analysis based on the number of publications, citations, and institutional affiliations retrieved from the Dimensions database. Of the 2130 authors, 127 meet the predetermined threshold of 3 publications. For each of the 127 authors, the total link strength was calculated

appropriately; then, the authors with the highest total link strength were selected.

The bibliometric analysis identified Ayora C., from the Institute of Environmental Assessment and Water Research, and Nieto J.M., from the University of Huelva, both in Spain, as the most prolific authors in the field of AMD wastewater remediation, each contributing 16 publications. Their works had 1105 and 1071 citations, respectively, indicating a high impact within the scientific community. Other notable contributors from the University of Huelva, Spain, include Macías F., Caraballo M.A., and Cánovas C.R., who have also demonstrated significant productivity and influence in this research field.

In addition to Spain, China has emerged as a prominent contributor, with active researchers, such as Dang Z., Li Y., and Liu Y., affiliated with the China University of Mining and Technology. Similarly, Li X. and Lu G., associated with the South China University of Technology, have also played crucial roles in AMD remediation research. This, therefore, suggests that although the United States has produced a large volume of publications in AMD wastewater remediation (Fig. 6 and 7), researchers from Spain and China are among the most actively publishing and highly cited. This shows their pivotal role in advancing the field and the commitment of other researchers and countries in this field.

Table 4 The top 10 authors that published articles from 1994 to 2025 on acid mine drainage wastewater remediation

Author	Publications	Citations	Total link strength	Affiliated Country	Affiliated Institution
Ayora, C	16	1105	8068	Spain	Institute of Environmental Assessment and Water Research
Nieto, Jm	16	1071	9198	Spain	University of Huelva
Macias, F	13	565	7846	Spain	University of Huelva
Caraballo, Ma	10	621	6900	Spain	University of Huelva
Dang, Z	10	414	4984	China	China University of Mining and Technology
Li, Y	10	272	3440	China	China University of Mining and Technology
Li, X	9	723	3642	China	Central South University Institute of Soil Pollution Control and Remediation
Cánovas, Cr	8	225	4331	Spain	University of Huelva
Liu, Y	8	266	3674	China	China University of Mining and Technology
Lu, G	8	361	4441	China	South China University of Technology



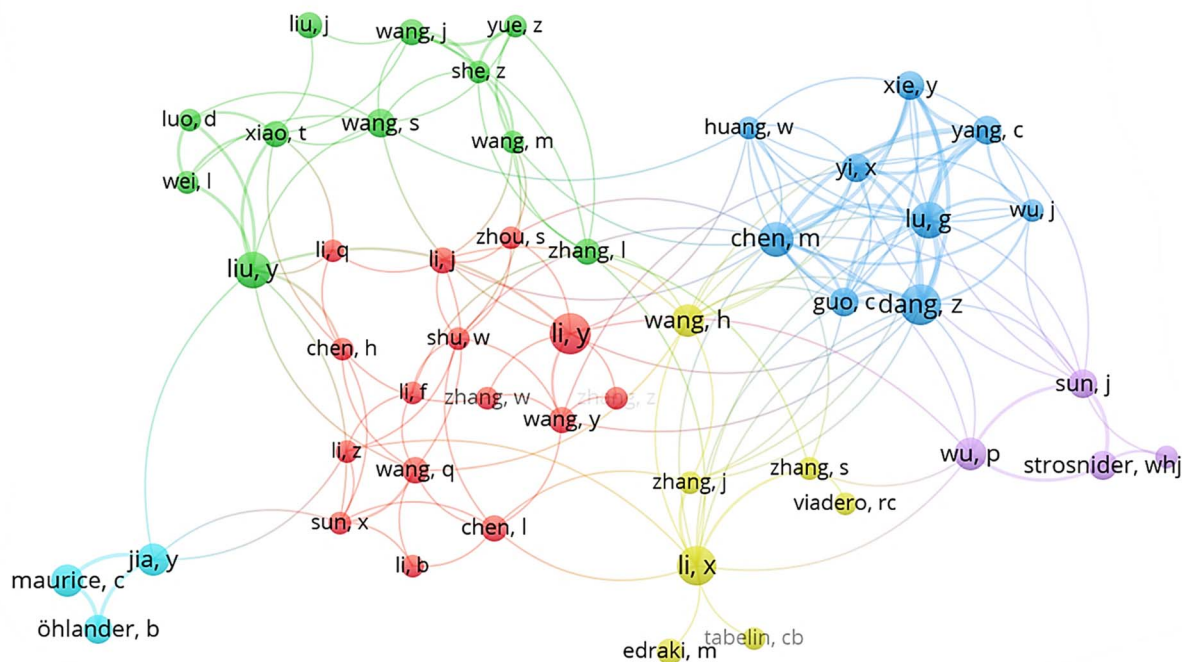


Fig. 8 Collaboration networks among authors/coauthors from 1994 to 2025 that published articles related to acid mine drainage wastewater remediation.

The findings in this study are consistent with previous bibliometric studies that have reported the significant contributions of Spanish and Chinese scholars to AMD treatment technologies.<sup>87,89</sup> For instance, Zhang *et al.*<sup>87</sup> emphasized increasing research outputs from Chinese institutions in AMD treatment using sustainable and advanced materials, while Wei *et al.*<sup>89</sup> reported the contributions of Spanish researchers in geochemical modelling and passive remediation systems.

The co-authorship network between authors is illustrated in Fig. 8. In this visualization, the thickness of the connecting lines between the two authors represents the strength of their collaboration, corresponding to the number of shared publications. Out of a total of 127 coauthors, 49 meet the pre-determined threshold (at least 3 publications). For each of the 49 coauthors, the total link strength was calculated; then, the coauthors with the highest total link strength were selected.

The network revealed six distinct clusters, with a total of 165 links and a total link strength of 282, indicating a high level of collaborative activity among the authors. These clusters indicated by 6 colours show that authors within the same cluster are citing each other and working on related topics compared to the cross-cluster. Among the authors, Chem M. demonstrated the highest collaboration, having co-authored publications with 17 different researchers. This was followed by Dang Z., who collaborated with 15 authors and produced 10 coauthored publications.

Other collaborating authors included Li X., Li Y., Liu Y., Jia Y., and Maurice C., all of whom exhibited strong co-authorship networks and contributed significantly to the field.

Similarly, Zhang *et al.*<sup>87</sup> observed a comparable clustering pattern in the environmental sciences, in which co-authorship

networks revealed strong group collaboration. Likewise, in a bibliometric analysis of wastewater treatment research, Chen and Zhao<sup>90</sup> reported that leading researchers tend to form connected clusters and are often centered around a few prolific authors. This network-based insight not only shows the collaborative dynamics in the field but also helps in identifying researchers and potential research partnerships.

### 5.6. Analysis of the organisation on acid mine drainage publication

The most active organizations publishing articles related to AMD wastewater remediation are depicted in Table 5. Of the 842 organizations, 100 meet the minimum threshold. From 100 organisations, the total link strength of the bibliographic coupling was calculated, and the highest total link strength was selected. From the results obtained, the University of Huelva in Spain was the most active organisation publishing articles in relation to the research topic, with 25 publications and 1249 citations in comparison to other organisations. This was followed by the University of the Witwatersrand in South Africa, with 16 publications and the Institute of Environmental Assessment and Water Research in Spain, with 11 publications. These findings indicate a significant contribution to AMD remediation research by organizations in Europe, Asia, the US, and Africa, reflecting growing global interest and collaboration in addressing this environmental challenge.

In addition, the dominance of these organizations can be attributed to their specialized research centres, long-term projects focused on mine water treatment, and access to AMD-affected regions for experimental studies. Moreover, their



Table 5 Top 10 organisations that published articles from 1994 to 2025 on AMD remediation

Organization	Publications	Citations	Total link strength	Country
University of Huelva	25	1249	9002	Spain
University of the Witwatersrand	16	879	3392	South Africa
Institute of Environmental Assessment and Water Research	11	791	5535	Spain
South China University of Technology	11	414	2546	China
West Virginia University	11	260	557	USA
United States Geological Survey	10	606	1204	USA
Universitat Politècnica De Catalunya	10	677	3544	Spain
Ohio University	9	206	1269	USA
Colorado School of Mines	8	72	954	USA
Guizhou University	8	182	1761	China

high total link strength suggests strong co-authorship networks and active participation in collaborative research, which are vital for advancing technological and scientific solutions for AMD treatment.

### 5.7. Analysis of document citations on acid mine drainage publications

The top 10 cited documents in relation to AMD wastewater remediation are depicted in Table 6. Of the 630 documents, only 417 meet the threshold of a minimum of 3 citations. As listed in Table 6, the most cited was Johnson (2005a) with 1723 citations and a total link strength of 102, followed by Sheoran (2006) with 649 citations, compared to the other documents. This indicates that these articles had a significant impact on this field of research possibly due to their novel contributions, methodologies and substantial findings to the ongoing research topic. Their high citations suggest that these articles have become widely referenced studies, serving as a basis for further research or validation of emerging hypotheses within the field of study.

Other documents, such as Park (2018), Simate (2014) and Nordstrom (2011), also contributed valuable insights to AMD wastewater remediation. Moreover, Cheng (2008), Naidu (2019a), Mendez (2007), and Sánchez-Andrea (2013) have over 300 citations, while Yu (2015) has citations below 300. Collectively, these documents underscore the significance of AMD wastewater remediation as a critical area of environmental research, particularly in relation to mining-impacted water

systems. The high citation rates show a growing commitment to finding sustainable, efficient, and cost-effective solutions to mitigate the adverse environmental effects of AMD. Their continued citation also reveals the persistent global challenge posed by AMD and the need for ongoing interdisciplinary research to address its environmental, ecological, and human health impacts.

### 5.8. Insight from bibliometric findings on AMD remediation research in future directions

The present bibliometric analysis on AMD (with a major focus on sustainable AMD remediation) and its treatment provides a concise summary of scholarly published research on AMD, identifying current trends and pinpointing research or knowledge gaps in AMD-based research. This detailed research mapping facilitates a better comprehension of sustainable acid mine drainage wastewater remediation strategies and advancements.

The high AMD-related research efforts from environmental science, earth science and engineering to address the AMD environmental menace are commendable, while lower publication numbers in biological sciences and related fields suggest that these approaches are evolving and remain underexplored, presenting opportunities for future research. Application of biotechnological tools could provide a better understanding of the bio-based approach in AMD treatment towards improved AMD treatment and global sustainability goals.

Moreover, research articles on AMD wastewater remediation have been published in sixty-one countries. Of these, thirty-nine met the predetermined minimum threshold of three publications per country. This implies that AMD-based remediation research has not attracted attention or priority in many countries of the world. Hence, there is a need to promote AMD-based remediation research in such countries to ensure proper awareness of governmental support, scholastic activities and environmental sustainability. Furthermore, some countries had low AMD-based research activities, as indicated by low publications in these countries (such as Iran, Nigeria, Russia, Colombia, Morocco, Mexico, Ecuador, Switzerland, Denmark, Ireland, Turkey, Greece, Pakistan, and Romania). Research activities in this field should be promoted, especially because of the existence of active mining sites in these countries.

Table 6 Documents and citations related to acid mine drainage wastewater remediation

Document	Citations	Total link strength
Johnson (2005a)	1723	102
Sheoran (2006)	649	230
Park (2018)	569	637
Simate (2014)	542	428
Nordstrom (2011)	401	166
Cheng (2008)	396	276
Naidu (2019a)	380	585
Mendez (2007)	361	68
Sánchez-Andrea (2013)	334	773
Yu (2015)	289	17



Additionally, initiating necessary collaborations with other countries is another important research effort required for environmental sustainability, especially with countries that have the skill, technology know-how and funding. This will enhance the global research network, common goal, novel knowledge dissemination and promote sustainable remediation technologies.<sup>85,86</sup>

Moreover, tackling environmental challenges, like AMD improper disposal, requires interdisciplinary approaches; the biological and related efforts towards AMD remediation are still a research niche that needs further exploration. This identified research gap agrees with the report by Zhang *et al.*,<sup>82</sup> who indicated that interdisciplinary research efforts are increasingly necessary to develop innovative and sustainable AMD treatment technologies. Presently, hybrid or integrated systems combining different technologies, such as chemical precipitation with biological treatment or chemical precipitation with membrane filtration, are still underexplored. Additional research in this regard is needed to leverage these synergistic strengths.

Furthermore, the bibliometric analysis outputs showed that AMD treatment research is still ongoing, and it is imperative to identify novel cost-effective, sustainable methods of AMD treatments and meet unmet needs in ongoing treatment efforts.

The use of innovative strategies, including artificial intelligence, robotics, and optimization modelling for enhanced treatment, is presently attractive and desirable. With the increasing global need for rare earth minerals to sustain the artificial intelligence (AI) revolution, mining activities are bound to increase in many countries with huge mineral deposits, hence the importance of innovative strategies for AMD treatment to combat the large volume of AMD to be disposed of.<sup>85,86</sup>

## 6. Treatment of acid mine drainage (AMD)

### 6.1. Conventional strategies for the treatment of acid mine drainage (AMD)

The repercussions of discharged AMD on soil, waters and human health may persist for hundreds of years if not managed properly. Hence, the implementation of adequate remediation technologies is vital over the operational lifetime of mine and abandoned mine sites.<sup>20</sup> Various conventional techniques, nanotechnology, and green and integrated approaches have been implemented to alleviate the negative impacts of AMD and safeguard human health and the environment.<sup>3,26,52,91-93</sup> The choice of a treatment technique is dependent on factors such as cost, treatment efficiency, AMD composition and quantity.<sup>26</sup> An ideal AMD treatment solution should be cost-effective and have a high sulphate and heavy metal removal efficiency, high water quality generation, minimal waste generation, and easy process development and management.<sup>94</sup>

Conventional techniques usually employ large inputs of chemicals, energy and other materials to drive the treatment process.<sup>3</sup> Chemical precipitation forms the most common

conventional AMD treatment technique.<sup>94</sup> It primarily uses alkaline chemicals, such as  $\text{Ca}(\text{OH})_2$ ,  $(\text{OH})_2$ ,  $\text{CaO}$ ,  $\text{NaOH}$ ,  $\text{Na}_2\text{CO}_3$ ,  $\text{NH}_3$ ,  $\text{MgO}$ , and  $\text{Mg}$ , to increase the pH and precipitate the metals contained in AMD.<sup>3</sup> This neutralisation technique proves to be effective in the precipitation of heavy metals to metal hydroxide precipitates and sulphate to sludge.<sup>20</sup> For instance, Masindi *et al.*<sup>95</sup> reported that alkaline chemicals, caustic soda, hydrated lime, periclase and magnesite have a suitable capacity for the removal (after pH alteration) of chemical contaminants in AMD at a low dosage. The study observed that caustic soda, hydrated lime, periclase and magnesite increased the pH of an AMD sample from 1.8 to 12.9, 10.9, 9.7, and 9.1, respectively, at a dosage of  $10 \text{ g L}^{-1}$ .<sup>95</sup>

Effective chemical precipitation can also be achieved using oxidative precipitation. This involves the addition of oxidising agents, such as hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) to AMD. It facilitates the conversion of heavy metal ions from their soluble reduced state to a less soluble oxidised form.<sup>94</sup> Although chemical precipitation is effective in recovering valuable metals in the form of precipitates and can be optimised according to the specific pollutants present, certain drawbacks are associated with this technique. For example, it generates additional waste products, requires pH adjustment to achieve optimal precipitation and can be influenced by competing ions present in the water.<sup>94</sup> Besides chemical precipitation, other conventional methods, such as adsorption, filtration and ion exchange technology, have been implemented for AMD remediation.<sup>3,20</sup>

Adsorption uses adsorbents such as activated carbon, zeolites and clay minerals for the removal of AMD.<sup>3,71,94</sup> Treatment of AMD using the adsorption technique presents several advantages, including high regeneration capacity, a wide pH range and a high metal binding capacity.<sup>96</sup> Despite these benefits of adsorption for AMD treatment, it is faced with some significant drawbacks. For instance, adsorption requires high selectivity, and the affinity of adsorption can affect its efficiency in decontaminating multi-charged wastewater, like AMD. Additionally, it has a rapid saturation tendency, which can lead to poor performance in highly concentrated solutions, thus limiting its potential for AMD treatment.<sup>96</sup> Most times, the regenerates from the adsorption process are highly mineralised and heterogeneous, thus posing a huge challenge to pure and high-quality mineral recovery.<sup>94</sup> Moreover, the disposal of regenerates requires proper handling, thus increasing the overall cost.<sup>3</sup>

Filtration technique employs physical barriers, such as sand, activated carbon, cellulose and anthracite coal, for the removal of contaminants and solid particles from AMD.<sup>3,53</sup> Depending on the specific application, filtration can also use various types of membranes, including microfiltration, ultrafiltration, nanofiltration, and reverse osmosis membranes.<sup>97</sup> The filtration process can be carried out using either depth or surface filtration pathways.<sup>94</sup> Depth filtration generally offers a higher contaminant-holding capacity compared to surface filtration, thus forming a critical component of water purification technology.<sup>98</sup>

Moreover, filtration plays a critical role in AMD treatment and is often combined with other processes like chemical



precipitation, ion exchange and biological treatment to achieve efficient remediation.<sup>94</sup> The major advantages associated with this technique are the efficient removal of particulate matter and reduced suspended pollutants and solids, resulting in improved water clarity. Moreover, it requires a relatively simple implementation and maintenance process.<sup>94</sup> The efficient implementation of the filtration system is confronted by setbacks; for example, it requires regular maintenance and replacement of the filter.<sup>99</sup> Additionally, it may not be effective in removing dissolved pollutants, while high flow rates could lead to reduced filtration efficiency.<sup>94</sup>

Furthermore, certain pre-treatment steps, including pH adjustment, coagulation, flocculation, and sedimentation, might be required prior to filtration for optimal AMD treatment.<sup>20</sup> The pretreatment process could also be aimed at preventing the clogging of the filtration media.<sup>100,101</sup> Of course, these pre-treatment steps may require additional costs for electricity input and management of the generated wastes, thus increasing the overall cost of the remediation process.

Ion exchange involves the reversible interchange of ions between a solid medium and an aqueous solution. An ion exchange system comprises a chemically inert polymer matrix with anionic or cationic functional groups.<sup>102</sup> The mechanism of operation entails the passage of AMD through a bed of ion exchange material (ion exchange resin), which is charged with monovalent cations, such as sodium.<sup>102</sup> This technology has proved highly efficient in AMD treatment, with over 95% metal and water recovery efficiency. However, it requires some pre-treatment that increases the process overall cost.<sup>103–105</sup> For instance, Feng *et al.*<sup>103</sup> recorded 100% heavy metal removal and 98% water recovery efficiency from AMD effluent treated using an ion exchange treatment plant with a capacity of 2.5 ML per day. However, the process requires 2.5 kg m<sup>-3</sup> acid for the regeneration of resin, 2.5 kg m<sup>-3</sup> of lime and an estimated operational cost of US\$0.40 m<sup>-3</sup>.<sup>103</sup>

## 6.2 Nanotechnology-based strategies in AMD remediation

With the increasing trend in mining operations (especially in gold and coal mining), heavy metals pose a great threat to the environment due to their discharge in AMD above permissible limits. Heavy metals have toxic impacts on humans and the environment. Interestingly, emerging nanotechnology has attracted considerable attention as a promising treatment technique for AMD pollution and its remediation.<sup>106,107</sup> Nanotechnology involves the modification of matter at the nanoscale to create new and unique materials/products.<sup>108</sup> Emerging nanotechnology (nanoremediation) provides nanomaterials for the decontamination of AMD.<sup>20,21,26,94</sup> Nanoremediation is also effective in the removal of pathogenic microorganisms, such as *Vibrio cholerae* (Cholerae) and *Salmonella typhimurium*.<sup>106</sup> It employs nanomaterials, including iron oxide nanoparticles, zinc oxide nanoparticles, and charcoal ash, for AMD remediation.<sup>49</sup> The nanomaterials, especially metallic nanoparticles, have a characteristic high surface adsorption tendency that allows their surface functional groups to adhere, bind, and remove AMD pollutants.<sup>26</sup> Nanoremediation is cost effective

and compatible with other treatment technologies.<sup>106</sup> The potential of nanomaterials in AMD remediation has been reported by several researchers.<sup>49,109–111</sup> For instance, Dlamini *et al.*<sup>109</sup> evaluated the flocculation efficiency of FeCu bi-metallic nanoparticles (FeCuBNPs) for coal mine water remediation. Their findings showed that FeCuBNPs flocculated at a low dosage (0.2 mg mg<sup>-1</sup> L<sup>-1</sup>), with a high flocculation efficiency of 99% achieved at pH 7 and a lower flocculation of 95% achieved at acidic pH 3.<sup>109</sup> Furthermore, Ji *et al.*<sup>110</sup> reported that a nanomaterial, polyethyleneimine-diatomaceous earth nanoparticle (PEI-DE-NP), was very effective in the removal of Cu from freshwater polluted by AMD. The results also revealed that the concentrations of other metals were considerably reduced after PEI-DE-NPs were in contact with the polluted fresh water.<sup>110</sup>

Despite its promising attributes in AMD treatment, the application of nanoremediation has some severe environmental and health demerits. Nanoparticles can easily spread and disperse in nature and thus might contaminate the environment.<sup>106</sup> For example, zero valent iron (nZVI) NP has a great capacity for sequestering a variety of contaminants found in AMD water. However, a high dose of nZVI might form a cluster, thus losing its nanoparticle features and becoming toxic to the environment.<sup>106</sup>

Furthermore, Gómez-Sagasti *et al.*<sup>112</sup> studied the impacts of nZVI-NPs on bacterial communities and revealed that the cytotoxicity of nZVI-NPs was dose and species-dependent and influenced by the process conditions. The authors added that oxidation of high concentrations of nZVI supports the release of reactive oxygen species (ROS), leading to oxidative stress, cell membrane disruption and death.<sup>112</sup> Exposure to NPs in humans can also cause genotoxicity, inflammation, lipid peroxidation and pulmonary disease.<sup>113</sup>

Additionally, high concentrations of nZVI affect plant metabolism by reducing the transfer of nutrients from roots to shoots, thus disrupting growth and eventually causing the death of the plant.<sup>113–115</sup> Due to their small particle size and high persistence in the environment, NPs can easily spread and disperse in nature, increasing the risk of bioaccumulation in living organisms to toxic levels.<sup>97</sup> Overall, the application of chemically synthesized NPs might result in problem-shifting, challenging the sustainability of nanotechnology in AMD treatment. Hence, research interest is shifting towards more sustainable approaches, such as green technologies, for AMD remediation.

## 6.3 Green strategies for the treatment of acid mine drainage (AMD)

The higher cost and energy requirement of chemically based conventional methods, and non-sustainable synthetic nanotechnology for AMD treatment have given rise to increasing demand for efficient and cost-effective strategies, such as green remediation with a lower environmental footprint. The technique of green remediation focuses on the use of microorganisms, such as bacteria, fungi, and plants, for the treatment of AMD.<sup>3,94,116–119</sup>

Microbes and plants represent ideal biological agents for the remediation process due to their ability to adsorb, solubilize,



Table 7 Some benefits and drawbacks of various green technologies used for AMD treatment

Green technology	Benefits	Drawbacks	References
Phycoremediation	Rapid microalgal growth, economy; eco-friendliness; sustainability	Inhibition of microalgal growth due to heavy metal toxicity	26 and 131
Nanobioremediation	Large surface area, simplicity; stability; biocompatibility; low toxicity; sustainability	Poor selectivity; slow process; difficulty in recovering nano-wastes; possible leaching of new contaminants from nanomaterials to the waste stream	109, 111 and 178
Mycoremediation	Versatile; cost-effective; non-toxic; high metal tolerance	Slow; specific to contaminants and environmental conditions	26, 137 and 141
Phytoremediation	Cost-effectiveness; sustainability; no substrate requirement	Low biomass production; risk of contaminant transfer; limited plant species; slow; inability to remove all pollutants	26 and 128
Biosorption	Effective for heavy metal removal, cost-effective and readily available; can be modified to enhance adsorption capacity; versatile	The efficiency depends on heavy metal concentration, contact time, and biomass characteristics	94 and 179

and precipitate heavy metals and other pollutants in AMD.<sup>26</sup> Primarily, the green remediation strategy employs biochemical and physiological mechanisms, such as bioaccumulation, bioadsorption, phytoextraction, sequestration and microbial degradation, to facilitate contaminant removal.<sup>120,121</sup>

Some green remediation technologies that have gained significant insight into the treatment of AMD effluent include phytoremediation, phycoremediation, mycoremediation, nanobioremediation, and biosorption.<sup>26</sup> The benefits and drawbacks of some of these emerging technologies are summarised in Table 7. The efficiency of these strategies is influenced by the physiological conditions of the organisms, effluent composition, and bioprocess parameters, including temperature, pH, moisture and dissolved oxygen.<sup>26,94</sup> Hence, there is a need for the optimization of process parameters during the remediation process.

**6.3.1. AMD treatment using phytoremediation.** Phytoremediation technology usually entails the application of selected plant species (metallophytes) for the treatment of AMD-contaminated soil and water using the mechanisms of phytoextraction and phytostabilization.<sup>26,106,118</sup> During phytoextraction, the metallophyte takes up heavy metals from

contaminated sites and sequesters them into its tissues, such as vacuoles, cell walls, and cell membranes. Similarly, phytostabilization involves the confinement of AMD-heavy metal ions within the metallophyte rhizosphere, thereby causing a reduction in metal availability to ecosystems.<sup>30</sup>

Notably, the metallophyte might act as a heavy metal accumulator, hyperaccumulator, excluder, or indicator.<sup>122</sup> Heavy metal accumulators take up metals into aboveground biomass, while hyper-accumulators absorb metals at over 1% dry weight. Moreover, excluder takes up metals in the rhizosphere by precipitation, while indicator takes up more metals so that they attain balance with the external environment.<sup>28,122</sup>

Various plant species from families, such as Poaceae, Brassicaceae, and Asteraceae, have been reported to possess great potential for remediation of metal-contaminated mine sites.<sup>26,118,123</sup> For example, species such as *Salvinia molesta*, *Chrysopogon zizanioides*, *Hyparrhenia hirta*, *Setaria sphacelata*, *Azolla filiculoides* and *Eichhornia crassipes* have been reported to have over 70% removal efficiencies for heavy metals, with *Lemna minor* showing up to 100% removal efficiency (Table 8).

The selection of plant species as ideal candidates for the effective decontamination of heavy metal-contaminated AMD

Table 8 Removal efficiencies of heavy metals from AMD by some plant and microalgae species

Plant species	Heavy metal removal efficiency	References
<b>Plant species</b>		
<i>Hyparrhenia hirta</i>	99% Fe	180
<i>Chrysopogon zizanioides</i>	81% Fe; 81% Pb	92
<i>Azolla filiculoides</i>	100% Cu	181
<i>Lemna minor</i>	74% Cu	181
<i>Eichhornia crassipes</i>	99.5% Cr(vi)	182
<i>Salvinia molesta</i>	80.99% Cd; 96.96% Pb; 92.85% Cr; 97.01% Ni; 94.12% Fe; 96.77% Cu; 96.22% Mn; 96.38% Zn	183
<b>Microalgal species</b>		
<i>Chlorella vulgaris</i>	Mo <sup>2+</sup> 99.9%; Cu <sup>2+</sup> 64.7%	132
<i>Nannochloropsis oculata</i>	Cu <sup>2+</sup> 89.29%	133
<i>Spirulina</i> sp	Fe <sup>2+</sup> 99%; Pb <sup>2+</sup> 95%; Zn <sup>2+</sup> 93%; Cu <sup>2+</sup> 94%	134
<i>Spirulina platensis</i>	Ni <sup>2+</sup> 95%; Al <sup>3+</sup> 87%; Cu <sup>2+</sup> 62%	135
<i>Chlorella vulgaris</i>	Ni <sup>2+</sup> 87%; Al <sup>3+</sup> 79.1%; Cu <sup>2+</sup> 80%	135



relies on factors such as high biomass production, bioaccumulation capability, and metal and acid tolerance potential. For instance, *Chrysopogon zizanioides* (vetiver grass) possess high metal, acid and drought-tolerant physiological characteristics and thus can be employed for the remediation of metal-contaminated sites.<sup>124</sup> This plant species can tolerate Zn concentration of about 302–531 mg kg<sup>-1</sup>, Fe concentration of 63–920 mg kg<sup>-1</sup>, Mn concentration of 415–648 mg kg<sup>-1</sup>, and Cu concentration of 13–66 mg kg<sup>-1</sup> in its root and shoot.<sup>125</sup> Similarly, *Atriplex halimus* accumulates up to 440 and 830 mg kg<sup>-1</sup> of Zn and Cd in its biomass, respectively, when cultivated on a mine tailing site<sup>126</sup>). Another species, *Thlaspi caerulescens*, accumulates 13 000–19 000 mg kg<sup>-1</sup> Zn and 50–250 mg kg<sup>-1</sup> Cd while growing on AMD-contaminated sites.<sup>127</sup>

Despite the cost-effective and sustainable attributes of phytoremediation technology, its efficacy is challenged by drawbacks, including low biomass production in phytoremediators, risk of contaminant transfer, limited plant species, a very slow process and not applicable in multi-pollutant removal.<sup>128</sup>

**6.3.2. AMD treatment using phycoremediation.** The phycoremediation strategy employs microalgae (single or consortium) as the bioremediation agent for the removal of hazardous pollutants (e.g., heavy metals) in AMD.<sup>129</sup> Microalgae have the natural ability to utilise wastewater pollutants, such as heavy metals, nitrogen and phosphorus, as nutrients for growth, with concomitant wastewater treatment and cost-effective production of microalgal biomass.<sup>129</sup> The process of phycoremediation is facilitated by functional groups in the microalgal cell walls, which act as binding sites for the uptake of heavy metals.<sup>130</sup> Additionally, microalgae secrete phytochelatin (an oligomer of glutathione), which acts as a chelator for heavy metal detoxification. Phycoremediation can be carried out by extracellular sorption and intracellular accumulation mechanisms.<sup>131</sup>

Extracellular sorption is faster and occurs just after contact of microalgae with heavy metals in AMD. It involves electrostatic interaction of the heavy metal ions with anionic algal cell ligands, micro-precipitation, surface complexing, and covalent bonding of polymeric molecules, such as proteins and lipids.<sup>130</sup> Conversely, intracellular accumulation is a slow and species-specific approach involving the binding of metal ions to intracellular compounds following the passage of the heavy metals into the microalgal cytoplasm.<sup>26</sup> The utilisation of microalgae for heavy metal removal is considered an ideal technology for AMD bioremediation due to its attributes, including low cost, ease of cultivation, rapid growth, high biomass production, high efficiency, sustainability, and eco-friendliness.<sup>42</sup>

Microalgal genera such as *Chlorella*, *Spirulina*, *Scenedesmus*, *Ulothrix*, *Chlamydomonas*, and *Nannochloropsis* have been identified as suitable candidates for phycoremediation of AMD, with over 80% removal efficiency for various heavy metals being reported (Table 8). For instance, Urrutia *et al.*<sup>132</sup> assessed the potential of *Chlorella vulgaris* for the removal of heavy metals from mine tailings water. The authors reported 99.9% and 64.7% removal efficiencies of Mo and Cu, respectively, using *Chlorella vulgaris*.<sup>132</sup> Similarly, Martinez-Macias *et al.*<sup>133</sup> investigated the ability of *Nannochloropsis oculata* to remediate heavy metals in AMD. Their findings indicated the remarkable

potential of microalgae species to eliminate heavy metals, with 89.29% removal efficiency of Cu recorded.<sup>133</sup>

In another related study, Van Hille *et al.*<sup>134</sup> reported optimum Pb (95%), Cu (94%), Fe (99%), and Zn (93%) removal efficiencies during the biotreatment of AMD using *Spirulina* sp. by alkalinity generation and precipitation. Similarly, Almomani and Bhosale<sup>135</sup> tested the ability of *Spirulina platensis* and *Chlorella vulgaris* to recover heavy metals in AMD. Maximum recovery of Ni (95%), Al (87%), and Cu (62%) by *Spirulina platensis* was obtained, while Ni (87%), Al (79.1%), and Cu (80%) were recorded using *Chlorella vulgaris*. Furthermore, Martínez *et al.*<sup>136</sup> obtained 92.8% Fe<sup>2+</sup> removal by *Muriellopsis* sp. from AMD after 12 h of cultivation.

Notably, the efficiency of microalgae to detoxify AMD is dependent on factors such as the physiology of the organisms, microalgal species, heavy metal ion concentration, temperature and pH, with the most critical factor being the AMD nutrient profile.<sup>131</sup> A high nutrient AMD effluent consisting of a huge organic load (N, P, and BOD) usually results in the excessive growth of microalgae (algal bloom). Therefore, achieving an ideal nutrient balance for microalgae optimum growth may require adequate pretreatment, which might increase the overall cost of AMD treatment.

**6.3.3. AMD treatment using mycoremediation technology.** Another emerging concept of green remediation of AMD involves the use of fungi as biological agents in the removal of toxic pollutants present in AMD.<sup>26,117–119,137</sup> Fungi are highly acid tolerant and proliferate in diverse acidic habitats, such as AMD, volcanic springs, and acidic industrial wastewater. For example, some filamentous fungi, such as *Aconitum* and *Cephalosporium*, can grow at pH 0. Other fungi capable of growing in acidic habitats include *Aspergillus* sp., *Geotrichum* sp.,<sup>138</sup> *Acidiella bohemica*,<sup>139</sup> *Penicillium* sp., *Scytalidium acidophilum*<sup>140</sup> and some yeasts.<sup>137</sup>

Heavy metal removal from AMD by fungi is predominantly achieved by the mechanisms of extracellular and intracellular sequestration,<sup>26</sup> with the fungal cell surface acting as ligands to bind heavy metal ions from the AMD stream.<sup>137</sup>

Extracellular sequestration is initiated by the secretion of fungal metabolites, including organic acids (e.g., gluconic acid and acetic acid) and siderophores, to facilitate the precipitation, immobilization, and transformation of heavy metal ions.<sup>26</sup> In the intracellular sequestration technique, heavy metals are transferred into the cell *via* specific transporters, including ATP-binding cassettes. This mechanism involves the complexation of metals by peptide ligands and the compartmentalization of the obtained metal complexes in vacuoles.<sup>26,137</sup>

Besides being acid tolerant, fungi possess other significant characteristics that qualify them as ideal candidates for the bioremediation of AMD. First, fungi secrete stable organic colloids for the uptake and removal of pollutants, like iron, from AMD-contaminated wastewater.<sup>137</sup> Indeed, fungi play a crucial role in ferric ion or sulphur reduction, a critical electron transport process in AMD that contributes to the biological neutralisation of acidic effluent.<sup>137</sup>

Various studies have supported the use of fungi as a promising candidate for the bioremediation of AMD. For instance,



Palanivel *et al.*<sup>141</sup> studied the efficacy of *Aspergillus hiratsukae* LF1 and *Aspergillus terreus* LF2 in the remediation of abandoned mine sites. Their findings revealed enhanced heavy metal tolerance and accumulation of the fungi, with maximum Cu removal efficiencies of 21–57% and 24–69% recorded for *Aspergillus hiratsukae* LF1 and *Aspergillus terreus* LF2, respectively. Similarly, a six-month investigation of the heavy metal detoxification potential of *Gloeophyllum sepiarium* reported 94% recovery of Cr(vi) from chromium-polluted sites.<sup>142</sup> Additionally, *Yarrowia* spp. demonstrated a significant heavy metal removal efficacy, removing 97% (Hg) from a medium ( $16 \mu\text{g mL}^{-1} \text{Hg}^{2+}$ ) during a bioremediation experiment.<sup>143</sup> Moreover, Merten *et al.*<sup>144</sup> reported 9.8–65% removal efficiencies of Ni, Cu, Cd, Al, U, Sr, Mg, Na, Ca, Mn, Co, Fe, and rare earth elements from mine drainage water in Eastern Thuringia, Germany, using *Schizophyllum commune*.

Although mycoremediation is a promising and sustainable green technology for the detoxification of hazardous pollutants in AMD, it is challenged by limited fungal species suitable for the removal of pollutants from AMD.<sup>26</sup> Therefore, bioprospecting novel fungal strains capable of bioaccumulation and removal of heavy metals from AMD is still imperative to increase the community of fungi species with heavy metal bioremediation potential.

**6.3.4. AMD treatment using nanobioremediation technology.** This is an emerging green technology that focuses on the detoxification of organic and inorganic pollutants in AMD using biologically produced nanoparticles.<sup>49</sup> This technique serves as an alternative to less sustainable and more chemically dependent nanotechnology. The green synthesized nanoparticles are mostly preferred because of their biocompatibility, eco-friendliness, lower-toxicity, higher stability, catalytic reactivity, lower cost, larger surface area and sustainability.<sup>145</sup>

Due to their ease of cultivation, high growth rate, and ease of genetic manipulation, the biological synthesis of nanoparticles from microbes has gained increased research interest in the remediation of waste effluent contaminated with sulphate and heavy metals.<sup>110,146</sup> However, their efficient implementation is influenced by factors such as microbial source, pH, incubation time, pressure, temperature, and metal salt concentration.<sup>146</sup>

Microorganisms, including bacteria, fungi, actinomycetes, algae, or their metabolites, are being employed as potential bio-factories for the synthesis of metal-based nanoparticles.<sup>147</sup> These bio-nanoparticles could be suggested as an ideal alternative to the predominantly used inorganic nanoparticles. The microbial production of NPs occurs intracellularly or extracellularly and is catalysed by microbial enzymes, such as NADH-dependent reductase.<sup>148</sup> Heavy metal sequestration and removal from AMD are achieved through adsorption, oxidation–reduction, surface complexation, or precipitation.<sup>145</sup>

The remarkable bioactivity of microbially synthesized nanoparticles for the remediation of high-strength heavy metal-laden wastewater has been considerably reported.<sup>149–151</sup> Chen *et al.*<sup>151</sup> generated biogenic ferrous sulphide NPs using *Geobacter sulfurreducens* for the bioremediation of AMD at pH 5.0 and 150 rpm. The authors recorded efficient removal of various heavy metals, including 92.6%  $\text{Pb}^{2+}$ , 62.2%  $\text{Mn}^{2+}$ , 78.7% Cd,

88.5%  $\text{Zn}^{2+}$ , 76%  $\text{Cu}^{2+}$  and 62.5%  $\text{Ni}^{2+}$ , from the effluent. Similarly, Mahanty *et al.*<sup>150</sup> synthesized biogenic iron oxide nanoparticles using *Aspergillus tubingensis* for the detoxification of heavy metal-polluted wastewater. Their findings showed that the maximum removal efficiencies of 98% (Pb), 96.45% (Ni), 92.19% (Cu), and 93.99% (Zn) were obtained under optimum conditions of pH 6.0, 308.25 K, and  $1 \text{ g L}^{-1}$  iron oxide nanoparticles.<sup>150</sup> Moreover, *Citrobacter freundii* Y9-based selenium nanoparticles reduced 45.8–57.1% and 39.1–48.6% of Hg in a contaminated site to insoluble mercuric selenide under anaerobic and aerobic conditions, respectively.<sup>149</sup>

Despite being more sustainable than chemical nanotechnology, green synthesis of nanoparticles for AMD bioremediation is still limited by the aggregation of nanomaterials, slow process operation and less selectivity.<sup>26</sup>

**6.3.5. AMD treatment using biosorption.** Biosorption is a green technology that involves the utilization of living or non-growing biomass to detoxify polluted water. It is employed to remove heavy metals from AMD based on the ability of certain biomass to bind and accumulate metals onto their surfaces.<sup>31,152</sup> These biomass materials contain functional groups, like carboxyl, amino, and hydroxyl groups, that selectively bind onto metal ions, forming complex bonds.<sup>94</sup> Then, the metals are removed from the solution and immobilised onto the biomass effectively, thus reducing their concentration in the effluent.<sup>31,153</sup>

The selected biomass is usually processed through drying, grinding, and, sometimes, chemical pretreatment to enhance its metal-binding capacity.<sup>94</sup> After the biosorption process, the metal-loaded biomass can be separated from the water through physical means, such as filtration or centrifugation.<sup>154</sup> The regenerated biomass might be subjected to a regeneration process, such as acid or alkali treatment, to restore its metal-binding capacity for future use.<sup>154</sup> Thus, biosorption could be suggested as an attractive option for AMD treatment due to its cost-effectiveness and potential use of low-cost biomass materials.

Evidently, the bioremediation of wastewaters using some common biosorbents, such as banana biomass, *Citrus grandis* (pomelo) leaves, coconut tree sawdust, sugarcane bagasse, cedar leaf ash, rose biomass and *Moringa oleifera* seeds, is well documented in the literature.<sup>155–161</sup>

The study by Aman *et al.*<sup>157</sup> investigated the potential of rose flower biomass as a biosorbent for the removal of heavy metals from industrial effluents. The authors reported a maximum zinc biosorption capacity of  $46.08 \text{ mg g}^{-1}$ .<sup>157</sup> This report may provide a platform for the future development of an efficient, cost-effective and sustainable biosorbent bioremediation system to treat zinc-contaminated AMD effluent. Similarly, a study conducted by Dev *et al.* (2020) revealed that citrus peel (bare and immobilized) biomass possesses a high biosorption efficacy for heavy metal removal from contaminated water, with a reported  $116.2 \text{ mg g}^{-1} \text{Se(IV)}$  biosorption capacity.

Moreover, Amin *et al.*<sup>158</sup> reported  $227.2 \text{ mg g}^{-1}$  biosorption capacity of a biochar prepared from banana peel biomass for the removal of lead from heavy metal-contaminated effluent. Furthermore, Lim *et al.*<sup>159</sup> investigated the removal of Pb(II)



from simulated wastewater contaminated with Pb(II) using *Citrus grandis* (pomelo) leaves as a biosorbent. The authors recorded a high removal of 207.2 mg g<sup>-1</sup> of Pb(II) from wastewater. Their findings, therefore, suggest the remarkable potential of pomelo leaf biomass as a biosorbent in the bioremediation of heavy metal-polluted wastewater, such as AMD effluents.

*Moringa oleifera* has emerged as an efficient and cost-effective biosorbent for the treatment of AMD. *M. oleifera* contains cationic polyelectrolytes, amino, carboxylic and sulfonic functional groups on its surface<sup>162-165</sup> that improve its surface binding potential.<sup>164</sup> For instance, during AMD biosorption, the cationic polyelectrolytes attach themselves to the contaminants and create binding between them, condensing the contaminants as flocs.<sup>165</sup> Similarly, amine groups have good binding affinity that enables them to adsorb compounds with cationic or anionic charges at different pH values.<sup>164</sup>

The use of *M. oleifera* for heavy metal removal has several benefits, including lower cost, biodegradable sludge production and lower sludge volume. Moreover, it does not affect the pH of the effluent.<sup>165</sup> The significant binding affinity of *M. oleifera* biomass towards heavy metals presents it as a good biosorbent for AMD remediation.<sup>163</sup> Indeed, its potential for heavy metal removal has been well reported in the literature. For example, a study by Sajidu *et al.*<sup>166</sup> reported 92% lead removal using unmodified *M. oleifera* seeds. Similarly, Tavares *et al.*<sup>167</sup> investigated the biosorbent efficacy of *M. oleifera* husks, seeds, and pods for lead removal from wastewater and reported efficiencies of 98.3%, 99.4%, and 96.6%, respectively. Furthermore, Bhatti *et al.*<sup>168</sup> reported a 90% removal of zinc using *M. oleifera* biomass.

Most studies on the potential of the Moringa plant for heavy metal removal were carried out using its seed, leaves or pods.<sup>166,167</sup> However, each part of *M. oleifera* is either associated with food or medicinal use.<sup>163,169</sup> Therefore, the direct use of *M. oleifera* for AMD remediation may pose a threat to sustainability. Based on the availability and increasing demand for different parts of *M. oleifera* for food and medicinal purposes, its residues can be used as green biosorbents for eco-friendly bioremediation of heavy metals from AMD effluents.

Overall, the efficiency of the biosorption process can be influenced by the interaction of various parameters, including pH, heavy metal concentration, contact time, biosorbent dosage, agitation speed and biosorbent characteristics.<sup>94,165</sup>

Despite being a promising, efficient, and eco-friendly strategy, bioremediation technologies are challenged by some drawbacks that hinder their applicability for large-scale implementation.

These drawbacks are as follows. (1) AMD high metal concentration and low pH inhibit the growth of living organisms, (2) the disposal of plant species after AMD bioremediation can lead to secondary pollution and toxic effects, (3) the process could be slow limiting its commercial application for the removal of pollutants in AMD, (4) microbial or plant remediation results in the partial removal or breakdown of toxic pollutants in AMD, and (5) high costs for the operation and maintenance of the bioreactors.

Unfortunately, a single technique for the absolute treatment of AMD could be less satisfactory due to the complexity of AMD composition. Therefore, the implementation of an integrated or hybrid approach to AMD remediation is attractive.

**6.3.6. Conventional vs. nanotechnology vs. green strategies for AMD treatment.** Different conventional, nanotechnology, green and integrated strategies have been implemented for the remediation of AMD.<sup>26,52</sup> The choice of a treatment or remediation technique is dependent on cost, treatment efficiency, AMD composition and quantity.

Conventional strategies are the most common method employed in AMD treatment, including chemical precipitation, adsorption, filtration, and ion exchange; they employ large inputs of chemicals, energy and other materials to drive the treatment process.<sup>3,20,71</sup>

Chemical precipitation is the most common conventional AMD treatment technique. It primarily uses alkaline chemicals to increase the pH and precipitate the metals contained in AMD. Although chemical precipitation is effective in recovering valuable metals in the form of precipitates, it generates additional waste products. Although adsorption (having high binding and regeneration capacity) could be preferred to the chemical precipitation approach due to the generation of additional waste, the adsorption technique is limited by high selectivity, rapid saturation, affinity dependence, competing ions and high cost.<sup>3,94</sup> Other commonly implemented conventional strategies are filtration and ion exchange. The efficient implementation of both techniques is limited due to the need for regular maintenance, ineffectiveness in removing dissolved pollutants and additional costs incurred due to necessary pre-steps, including pH adjustment, coagulation, flocculation, and sedimentation.<sup>20,94,99</sup>

However, emerging nanoremediation has attracted considerable attention as a promising AMD treatment technique compared to conventional strategies.<sup>106,107</sup> Nanoremediation is effective in removing pathogens (such as *Vibrio cholerae* and *Salmonella typhimurium*) in AMD,<sup>106</sup> while antimicrobial potential is limited in other techniques. Nanomaterials are characteristically high in surface area-to-volume ratio, which improves their surface functionality and efficiency.<sup>26</sup>

However, nanoremediation is cost effective and compatible with other treatment technologies; its application has some environmental and health demerits. A major concern is that nanoparticles can easily spread and disperse in nature; thus, they might contaminate various environmental compartments with cytotoxicity and human poisoning potential, resulting in problem-shifting and limiting nanoremediation in AMD treatment sustainability.<sup>106,112</sup> This limitation can be resolved by applying a more sustainable approach: biological approach and green-based nanotechnologies.

The use of bio-based nanoparticles serves as an alternative to the less sustainable and more chemically dependent nanotechnology. Bio-nanoparticles exhibit biocompatibility, eco-friendliness, lower toxicity, higher stability, catalytic reactivity, lower cost, larger surface area and sustainability.<sup>145</sup> Crucial limitations, such as aggregation of nanomaterials, reclamation of nanowastes after treatment, slow process operation and less selectivity.<sup>26</sup>



The operational cost and energy demand of chemically based conventional methods, and non-sustainable synthetic nanotechnology for AMD treatment have given rise to efficient and cost-effective, green (biological) remediation with a lower environmental footprint. The eco-friendliness of these techniques usually involves the use of biological materials (bacteria, fungi, and plant biomasses) for the treatment of AMD.<sup>94,117,118</sup> Additionally, their ease of cultivation, rapid growth, high efficiency and sustainability make these strategies preferable.<sup>42</sup>

Additionally, these biological agents represent an ideal agent for AMD remediation due to their ability to adsorb, solubilize, precipitate, accumulate, form ligands and in some cases utilize the pollutants in AMD.<sup>120,121</sup> For instance, plants use the mechanisms of phytoextraction and phytostabilization, taking up heavy metals from the contaminated sites and sequestering them into their tissues, such as vacuoles, cell walls, and cell membranes. Interestingly, plants have been reported to have >70% removal efficiencies for heavy metals.<sup>26,106,118</sup>

Moreover, biosorption is another biological approach that binds and accumulates metals onto their surfaces. Compared to the conventional strategy, the use of biosorption for heavy metal remediation has several benefits, including lower cost, biodegradable sludge production and lower sludge volume.<sup>31,152</sup> The major challenges to bio-based AMD remediation include its highly process condition dependent, low biomass production, risk of contaminant transfer, limited remediating species (limited to suitable species), and the very slow nature of the process.<sup>128</sup> Conclusively, a single technique for the absolute treatment of AMD could be less satisfactory; hence, the application of an integrated approach could be desirable and suitable.

## 7. Integrated approaches to AMD treatment

Integrated technologies for the treatment of AMD involve the implementation of multi-strategies, such as sequential selective precipitation, adsorption, and filtration processes.<sup>3,170</sup> These systems usually combine two or more treatment strategies for the removal of AMD contaminants.<sup>106</sup> Despite AMD being a source of pollutants that threatens both human and environmental health, AMD contains minerals, which can be recovered as valuable resources. Additionally, fit-for-purpose water can be reclaimed.<sup>3</sup>

Beneficiation and valorisation of wastewaters, like AMD, are achieved through water reclamation and mineral recovery, respectively. Beneficiation involves the treatment of AMD effluent to reclaim water while reducing its environmental consequences. For instance, AMD beneficiation partly involves the removal and recovery of dissolved solids. Therefore, the removal of these AMD constituents increases opportunities for water reclamation, which could be fit for direct discharge to the environment and for a variety of purposes, such as irrigation, industrial reuse and anthropogenic uses.<sup>39,45,106</sup> For instance, Masindi<sup>45</sup> evaluated the integration of cryptocrystalline magnesite ( $\text{MgCO}_3$ ) and barium chloride ( $\text{BaCl}_2$ ) for AMD treatment and beneficiation. Cryptocrystalline magnesite was

applied to neutralise AMD and precipitate metals; the reclaimed water was then polished by  $\text{BaCl}_2$  to remove residual  $\text{SO}_4^{2-}$ . Their results also showed that 99% of metals and 99% of  $\text{SO}_4^{2-}$  were removed.

Moreover, various techniques, including electro dialysis, diffusion dialysis, distillation, acid retardation, freezing crystallisation, solvent extraction, and membrane technology, have been implemented for simultaneous AMD treatment and sulphuric acid ( $\text{H}_2\text{SO}_4$ ) recovery.<sup>44,171,172</sup> Similarly, Ricci *et al.*<sup>171</sup> successfully integrated microfiltration and nanofiltration membrane techniques to recover  $\text{H}_2\text{SO}_4$  and valuable metals from AMD. Other findings on the valorisation of AMD for the recovery of heavy metals are summarised in Table 9.

The integrated systems are versatile and effective in AMD treatment, resulting in high contaminant removal efficiency.<sup>94</sup> Integrated techniques have the potential to align with the circular economy and the 4R (reduce, reuse, recycle, and recover) principles. Therefore, the application of such technologies in AMD treatment could reduce the overall cost in AMD management.<sup>3</sup>

Unfortunately, due to their complexity, the commercial application of integrated systems faces challenges associated with high energy inputs, large land requirements, risks of system failure, secondary pollution, and high capital and operational costs.<sup>3,94,106,173</sup> For instance, Miranda *et al.*<sup>175</sup> evaluated the economic and environmental impacts of using industrial by-products to neutralise coal mine water and recover rare earth elements (REEs). The results revealed that industrial by-products had a higher neutralising capacity and were efficient for the recovery of REEs. However, the process raised various environmental and ecological concerns due to the transportation of materials to the site, earth excavation, deforestation, and oil spill.<sup>173</sup> Also, integrated systems require the frequent usage of various chemicals with different toxicity levels. Therefore, the implementation of integrated systems for AMD remediation could escalate the release of a mixture of toxic chemicals with possible ecotoxicological risks.<sup>26</sup>

## 8. Mechanism of AMD wastewater remediation

Studies have shown that contaminants present in AMD are removed through different mechanisms.<sup>78,174</sup> Some of the mechanisms in the remediation/removal of contaminants in AMD include solubilization, adsorption, ligand formation, complexation, speciation, oxidative processes and biological uptake or utilization, which produces secondary products that can further be converted to less toxic by-products.

Contaminant speciation on adsorbents is usually influenced by process conditions, such as pH and temperature. Certain pH conditions favour higher migration of contaminants to the adsorbent or filter material surface.<sup>174</sup> Precise pH modulation could also cause the precipitation of contaminants to a considerable extent. However, non-optimal pH conditions result in the competition of the different contaminant ions in AMD, thus hindering the migration of efficient removal of individual



Table 9 Integrated techniques for selective recovery of heavy metals from AMD valorisation

Technique	Metal ions	Recovery description	References
Selective precipitation	Cu and Zn	A field trial was carried out on AMD produced at an active mine using a selective precipitation pilot plant. Under the AMD condition of 1.4 L min <sup>-1</sup> , Cu and Zn precipitates with a purity of 80% and a precipitation rate of 90%	184
Chemical oxidation technology (H <sub>2</sub> O <sub>2</sub> -NaOH technology)	Fe	The chemical oxidation pilot process was conducted on AMD with high Fe concentration. Under acidic conditions, H <sub>2</sub> O <sub>2</sub> was able to rapidly oxidize Fe <sup>2+</sup> to Fe <sup>3+</sup> . The pH was adjusted to 3.8 using NaOH and Fe precipitates were formed. An average of 26.85% Fe was recovered	185
Membrane distillation and adsorption system	Cu	Cu was selectively recovered from the synthesized AMD solution by membrane distillation and adsorption systems. The pH of AMD was adjusted to 5.0–5.2 using KOH. Selective adsorption of Cu was carried out using direct contact membrane distillation in a multi-modified mesoporous silica SBA-15 material. 24.53 mg g <sup>-1</sup> of Cu was absorbed	186
Metal sulphide precipitation and membrane filtration process	Cu	Metal sulphide precipitation and membrane microfiltration were employed for the recovery of copper from synthetic AMD. The recovery of copper was approximately 100%, and turbidity values in the treated solution were lower than 2 NTU for sulphide stoichiometric dosages of 120%	187
Sequential selective precipitation and fluidized bed homogeneous crystallization (FBHC)	Fe and Al	A combination of sequential selective precipitation and fluidized bed homogeneous crystallization was used to recover Fe <sup>2+</sup> and Fe <sup>3+</sup> from AMD. Process conditions include a pH of about 9.25, [H <sub>2</sub> O <sub>2</sub> ]/[Al(III)] molar ratio of 2.0 and an upward flow rate (U) of 30.5 m h <sup>-1</sup> . Ferric hydroxide (Fe(OH) <sub>3</sub> ) and bayerite ( $\alpha$ -Al(OH) <sub>3</sub> ) pellets were prepared sequentially	188
Nanofiltration (NF) and solvent extraction (SX)	Cu	The feasibility of recovering copper from AMD was studied by pilot-scale tests. Nanofiltration (NF) was applied to nanofiltration (NF) and solvent extraction (SX) Cu concentrate from AMD solutions, followed by solvent extraction of Cu. 97% of Cu was recovered	189
Sequential selective precipitation	Fe, Al, and Mn	The selective precipitation of dissolved iron, aluminum, and manganese in the AMD from the Samma-Taejeong coal mine by adding oxidants and neutralizers was tested. The sequence of metal removal by oxidation, followed by neutralization, was Fe > Al > Mn.	44



Table 9 (Contd.)

Technique	Metal ions	Recovery description	References
Electrochemical reactions	Fe, Al, Cu, Zn, and Ni	Recovery rates of 99.2–99.3%, 70.4–82.2%, and 37.8–87.5% were obtained for dissolved Fe, Al, and Mn, respectively Electrochemical reactions utilised to oxidize Fe(II) to Fe(III) while producing neutralizing agents (containing a high concentration of hydroxide) for the selective recovery of dissolved metals (Fe, Al, Cu, Zn, and Ni) from AMD	170

contaminants to the target site on the adsorbent surface. Similarly, absorption capacity is influenced by available vacant sites on the adsorbent surface until saturation.<sup>175</sup> Moreover, the removal capacity of the adsorbent is higher at high contaminant concentrations due to the high driving force for the migration of contaminants to the adsorbent.<sup>176</sup>

Electrostatic attraction due to the difference in the charges of the contaminants in AMD and the surface charge of the adsorbent or filter material can effectively remove contaminants. In addition, the hydrophilic and hydrophobic features of the contaminants and the removal agents determine the removal effectiveness of the different contaminants. Likewise, removal agents have the potential to interact with contaminants to form ligands or complexes (complexation process) due to their chelating capacities. The attachment of contaminants to the adsorbent and removal from AMD through mechanisms highlighted above are usually achieved by bonds such as hydrogen bonds,  $\pi$ -hydrogen bonding and electrostatic interactions.<sup>78</sup>

## 9. Industrial process used on a large scale

An industrial process commonly used on a large scale for AMD treatment is preliminary treatment and polishing. The preliminary stage is primarily the active chemical treatment (lime neutralization), such as the high-density sludge strategy (HDSS). This method is highly effective and flexible for treating large volumes of highly acidic and metal-contaminated wastewater and is considered the industry standard for large-scale AMD treatment.<sup>119,177</sup>

HDSS involves AMD neutralization (typically choice lime is introduced to the AMD to achieve a pH > 9, causing dissolved heavy metals to become insoluble and precipitate out as metal hydroxides), followed by oxidation and precipitation (OP), flocculation and settling (FS), sludge recycling (SR) and water discharge stages. The OP stage involves the aeration of neutralised AMD to facilitate the oxidation of ferrous iron to ferric iron, which precipitates more effectively. The FS stage aggregates the fine solid particles into bigger flocs received or settle as sludge that can be recycled (at the SR stage). Afterwards, treated AMD is discharged and sometimes further treatments

are implemented to achieve higher quality standards for reuse at the industrial, irrigation, and domestic levels.<sup>119,177</sup>

The secondary industrial processes to achieve higher polishing (further treatment) include membrane filtration, reverse osmosis and nanofiltration with the potential to remove residual soluble solids towards achieving high-quality water suitable for reuse. Biological treatment is another strategy for further treatment with larger-scale potential, thereby requiring higher investment and operational technical know-how. Finally, an integrated system that combines multiple strategies, like chemical precipitation, with biological treatment to leverage the merit of each approach for optimum AMD treatment, could lead to reusable treated AMD and sometimes metal recovery.<sup>119</sup>

## 10. Conclusions, outlook and recommendations on the remediation of AMD

The complexity and toxicity of AMD composition make it a serious threat to human health and the immediate environment of which AMD is disposed of. The remediation of AMD is challenging because of this compositional complexity and has continued to attract global research attention. In the present study, the remarkable increase in the number of articles reporting different remediating strategies indicates a growing global interest in AMD remediation research activities. The United States (country), the University of Huelva, Spain and the University of the Witwatersrand, South Africa, emerged as the leading contributors to AMD-related research with a strong collaborative network. Moreover, AMD-based research activities in countries like Iran, Nigeria, Russia, Colombia, Morocco, Mexico, Ecuador, Switzerland, Denmark, Ireland, Turkey, Greece, Pakistan, and Romania are presently low. Moreover, tackling AMD-based environmental challenges requires interdisciplinary approaches to develop innovative and sustainable AMD treatment technologies.

Therefore, with the global scabbling for rare earth minerals to sustain the artificial intelligence (AI) revolution, mining activities are bound to increase in many countries with huge mineral deposits. It is recommended that such countries promote AMD-based remediation research towards achieving long-term environmental sustainability. Additionally,



international collaborations promoting AMD-based interdisciplinary research should be strengthened with a unified effort involving academia, industry, and policymakers.

Furthermore, the use of innovative technologies, including artificial intelligence, robotics, and machine learning, for enhanced treatment and mineral recovery from AMD, could be vital to achieving environmentally sustainable technologies. Additionally, the application of molecular techniques (presently, the biological approach is underexplored), including proteomics, metabolomics and transcriptomics, could be useful to provide a better understanding of the bio-based approach in the remediation of toxic pollutants in AMD and/or simultaneous recovery of valuable metals from AMD.

## Ethical statements

This article has not been previously published anywhere or submitted for publication consideration elsewhere.

## Author contributions

Emmanuel. C. Ngerem: conceptualization, methodology, data curation, writing – original draft, writing – review & editing. Isaac A. Sanusi: conceptualization, methodology, data curation, writing – review & editing, resources, supervision. Tatenda Dalu: conceptualization, writing – review & editing, supervision. Terence N. Suinyuy: conceptualization, writing – review & editing, supervision.

## Conflicts of interest

The submitted article was approved by all the contributing authors without any conflict of interest.

## Data availability

All data used in this review have been made available, included within the manuscript and cited accordingly (no new data were generated in this work).

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