



# **Bibliometric Analysis of Trends and Research Progress in Acid Mine Drainage Remediation from 1990 to 2023**

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Abstract: Acid mine drainage (AMD), arising from mineral resource exploitation, has transformed into a significant global environmental issue for the mining sector, posing considerable risks to water, soil, ecosystems, and human health. In this study, the current status and cutting-edge dynamics of AMD remediation research were evaluated using a bibliometrics approach. Publications on AMD remediation were collected from the Web of Science Core Collection (WOSCC) database, and the relevant literature was analyzed quantitatively using various statistical methods, including keyword co-occurrence and burst analysis. In total, 2743 articles related to AMD remediation published from 1990 to 2023 were obtained. The number of publications tended to increase annually, with a relatively fast rate of increase in recent years. Recent research related to AMD remediation has mainly focused on the ecological risks, the environmental geochemical cycling, the application of sulfate-reducing bacteria and adsorption, and the recovery of heavy metals (HMs) and rare earth elements (REEs). It is anticipated that these topics of AMD remediation research are expected to be at the forefront of future research endeavors. In addition, the current status, advantages, and challenges of AMD remediation technologies are discussed from both active and passive management perspectives, providing a theoretical basis and reference for AMD remediation.

Keywords: acid mine drainage; heavy metal; rare earth elements; bibliometric analysis; research progress

# 1. Introduction

In 2018, the mining industry provided 22.7 billion tons of energy, metals, and important non-metallic minerals for humankind, with a total output value of USD 5.9 trillion, equivalent to 6.9% of global GDP, and the development of mining has also brought a large number of environmental problems and challenges such as acid mine drainage (AMD). AMD is formed by the hydrogeochemical weathering of sulfur-containing minerals (prite, marcasite, and phrrothite) exposed to water and oxygen. This reaction is catalyzed by sulfur-oxidizing microorganisms and Fe<sup>3+</sup>. In addition, natural bacteria can assist in the decomposition of sulfide to accelerate AMD formation [1]. AMD is a long-term source of pollution that is produced in large quantities during the mining of mineral resources; major sources include rock piles, tailings ponds, and rainwater run-off in underground and open-pit mines [2]. AMD contains a large number of toxic and harmful substances, which can cause serious harm to water and soil environments when untreated water flows into



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**Copyright:** © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). rivers, lakes, and groundwater. AMD is highly acidic and contains high concentrations of sulfate and dissolved heavy metal (HM) ions (e.g., Fe, Cu, and Zn) [3]. AMD not only corrodes the rails, pipelines, pumps, and other mining equipment and building structures but also affects project construction. Furthermore, the AMD-induced acidification of water and soil affects the survival of microorganisms, algae, plankton, and fish in water and plants in soil [4]. In cells, trace amounts of HMs are necessary for enzymatic activity and biological processes. However, over a certain threshold concentration, HMs will impact enzymatic and metabolic activities and even lead to cell death [5]. HMs exhibit persistence in water bodies, as they do not degrade; they remain in the environment for extended durations. The adsorption of HMs by aquatic organisms and enrichment in plants can lead to bioaccumulation across numerous organisms, including humans [6].

Owing to the complex composition of AMD, including a large number of toxic and harmful substances, especially HMs, treatment and remediation are very difficult and complex. Therefore, it is important to develop efficient, sustainable, environmentally friendly, and low-cost AMD treatment technologies. Over the past few decades, researchers have explored the formation of AMD and its harmful effects, treatment and remediation technologies, and control countermeasures. The status of research on a variety of AMD remediation technologies (including physical, chemical, and biological methods) has been reviewed, including reviews of the application of constructed wetlands (CWs), focusing on the role of plants and microorganisms in removing HMs from AMD, as well as the effects of retention time, packing and medium type, preprocessing, and other parameters [7,8]. Phytoremediation technology for AMD remediation [9]. Membrane treatment technology has been reviewed for AMD remediation and metal recycling, including its general impact and the factors determining its efficiency [10]. However, no review has described the general research status or research trends related to AMD remediation.

Bibliometry is an interdisciplinary subject that combines mathematics, statistics, and philology for quantitative analyses of the literature [11]. Quantitative analyses of the literature reveal the knowledge structure of disciplinary fields and provide insight into the development of disciplines, temporal patterns, and future trends. This approach has gradually become an important means of predicting the development of various research fields [12]. Bibliometrics has been applied to a wide range of fields, including the treatment of emerging contaminants in wastewater [13], treatment of industrial wastewater [14], remediation of wastewater contaminated by antibiotics [15], and biological treatment of leachate [16]. However, few studies have used bibliometrics to systematically analyze AMD remediation.

In this study, a bibliometric analysis was conducted to evaluate the research history and trends in AMD remediation research. The main contributions of the study include the following:

- (1) The present study provides a succinct synthesis of prevailing AMD remediation technologies (active and passive treatment), evaluates the progress achieved in their application, and presents the advantages and challenges intrinsic to each therapeutic modality.
- (2) Through the application of HistoCite Pro 2.1, VOSviewer 1.6.18, and CiteSpace software 6.1.R3, a publication trajectory of the literature related to AMD remediation is constructed and visualized from an array of perspectives, encompassing annual publications, journals, authors, research institutions, countries, the highly cited literature, and keywords.
- (3) Based on keyword co-occurrence and burst analysis, the present study scrutinizes and proposes a research focus and development status in the field of AMD remediation from 1990 to 2023 while highlighting the current study limitations and future perspectives related to AMD remediation.

# 2. Data Acquisition and Methods

# 2.1. Data Source

Data were collected from the Web of Science Core Collection (WOSCC) database. The WOSCC database is the most frequently used indexed database for this type of analysis [17]. The retrieval strategies were as follows: TS = ("acid\* mine drainage" OR "acid mine wastewater" or "acid rock drainage") AND TS = (treatment\* OR remov\* OR remed\* OR treating), timespan = 1990–2023. To improve the accuracy of the analysis, conference papers and book chapters were filtered, and papers that were not relevant to the topic were removed based on the search results. Ultimately, 2783 relevant documents were retained. Some keywords were redundant, and these were unified (e.g., "acid mine drainage", "acid mine drainage", "acidi mine drainage", and "AMD" were collectively treated as "acid mine drainage").

## 2.2. Methods for the Scientometric Analysis

Statistical analyses of key metrics, such as annual publications, journal, author, institution, country, the highly cited literature, and keywords, were performed using various tools, such as HistoCite Pro, VOSviewer, and CiteSpace. The bibliometric analysis software HistCite Pro 2.1 [18] and Excel 2016 were used to conduct a quantitative analysis of 2783 documents on AMD remediation published from January 1990 to December 2023. VOSviewer 1.6.18 was used to analyze the cooperative relationships between authors of manuscripts published in the WOSCC database, co-citation, and the co-occurrence of keywords to reveal the knowledge flow, reflect the similarity, uniqueness, and citation relationships among papers, and explore the dynamics and trends in global research on AMD remediation [19]. CiteSpace 6.1.R3 was used for keyword burst analyses [20]. Impact factors (IFs) were obtained from the JCR, published in 2023 (http://admin-apps.webofknowledge.com/JCR/JCR, accessed on 25 June 2024.) [21].

#### 3. Results

#### 3.1. Number of Publications during 1990–2023

The number of publications provides a comprehensive indicator of the scientific community's interest in a particular topic, which can reflect the development speed and process of the field [22]. To reveal an overview of AMD remediation research, the annual number of publications of AMD remediation research during the period of Jan 1990–Dec 2023 are shown in Figure 1. The annual number of publications has increased from 3 in 1990 to 223 in 2023. While there have been occasional declines in the number of published publications on AMD remediation research, the general trend is increasing. The development trend of AMD remediation research can be segmented into three stages: From 1990 and 2001, limited studies were conducted, resulting in a slow increase in publications. From 2002 to 2010, the growth rate accelerated slightly but displayed substantial fluctuations in the number of publications published annually. In the period from 2011 to 2023, the number of AMD remediation studies develops dramatically, with the number of publications rising from 100 in 2011 to 223 in 2023. Since 2011, researchers worldwide have acknowledged the significance of AMD remediation, leading to a dramatic increase in published studies. Consequently, AMD is expected to remain a focal point for researchers globally.



Figure 1. Annual number of publications of AMD remediation research in the WOSCC database from Jan 1990 to Dec 2023.

Year

2006

2008

2010 2012 2014 2016

#### 3.2. Characteristics of Journals

1992

1990

. 1994

1996 1998 2000

2002

Numbers of publications

A total of 515 different journals published the related articles, with 411 (79.81%) journals featuring fewer than five articles and 53 (10.29%) journals featuring more than ten articles. Table 1 shows the top 10 most productive journals, which collectively published approximately 30.87% of the articles. Mine Water and the Environment ranked first with 109 articles (3.92%), followed by Applied Geochemistry with 107 articles (3.84%), Minerals Engineering with 101 articles (3.63%), and Journal of Hazardous Materials with 100 articles (3.59% each). When ranked by TLCS and TGCS, Water Research secured the top position with a TGCS of 4933, and Science of the Total Environment had a TLCS of 1087. Additionally, the Journal of Hazardous Materials registered the highest impact factor of 13.6 in 2023.

Table 1. The top 10 most productive journals from Jan 1990 to Dec 2023.

Journal	Publications	% of 2783	IF <sub>2023</sub>	TLCS	TGCS	Average Citation Frequency
Mine Water and the Environment	109	3.92	2.1	346	1364	12.51
Applied Geochemistry	107	3.84	3.1	1019	4538	42.41
Minerals Engineering	101	3.63	4.9	714	3217	31.85
Journal of Hazardous Materials	100	3.59	12.2	716	4804	48.04
Science of the Total Environment	93	3.34	8.2	1087	4336	46.62
Chemosphere	79	2.84	8.1	458	2361	29.89
Water Research	68	2.44	11.4	863	4933	72.54
Journal of Environmental Management	65	2.34	8.0	182	1140	17.54
Water, Air, & Soil Pollution	62	2.23	3.8	159	1193	19.24
Environmental Earth Sciences	57	2.05	2.8	166	840	14.74

Notes: TLCS: total local citation score, which represents the citation frequency of the publications in the current database; TGCS: total global citation score, which represents the total frequency of citations in the WOSCC database.

# 3.3. Characteristics of Authors

Over the past 34 years, 7596 authors have contributed to the AMD remediation research, with Table 2 presenting the top 10 most productive authors. Among them, four are from Spain, two from Canada, two from China, one from South Africa, and one from France. Ayora C from the CSIC, Spain, has contributed the most publications with 42 articles, followed by Nieto JM (40 articles), Dang Z (37 articles), Grande JA (37 articles), and Neculita CM (33 articles). Ranked by the TGCS, Ayora C (2207 times) holds the highest number, trailed by Nieto JM (1925 times) and Casiot C (1239 times). Furthermore,

2022

2020

2018

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according to the TLCS, Ayora C (715 times) possesses the highest value, succeeded by Nieto JM (547 times) and Neculita CM (376 times), indicating that Ayora C and Nieto JM have significantly influenced this research field.

Table 2. Top 10 most productive authors during Jan 1990–Dec 2023.

Labs	Institution/Country	Publications	TLCS	TGCS	Average Citation Frequency
Ayora C	CSIC/Spain	42	715	2207	52.55
Nieto JM	Univ Huelva/Spain	40	547	1925	48.13
Dang Z	South China Univ Technol/China	37	142	719	19.43
Grande JA	Univ Huelva/Spain	37	208	653	17.65
Neculita CM	UQAT/Canada	33	376	919	27.85
Benzaazoua M	UQAT/Canada	30	161	750	25
De La Torre ML	Univ Huelva/Spain	28	198	606	21.64
Lu GN	South China Univ Technol/China	28	135	600	21.43
Masindi V	Univ South Africa/South Africa	28	160	547	19.54
Casiot C	Univ Montpellier/France	27	233	1239	45.89

Notes: UQAT: University of Quebec in Abitibi-Temiscamingue, CSIC: Consejo Superior de Investigaciones Científicas.

## 3.4. Major Institutions and International Cooperation

A total of 2224 institutions contributed to articles on AMD remediation research, with Table 3 presenting the top 10 institutions. The University of Huelva in Spain is the leading institution, having published 113 articles, followed by the Chinese Academy of Sciences in China and the CSIC in Spain with 69 and 54 articles, respectively. Regarding the number of TGCS, the University of Huelva holds the highest number at 3396 times, followed by the CSIC and US Geological Survey with 2471 and 2201, respectively. Considering three indicators, including publications, TGCS, and TLCS, the University of Huelva, US Geological Survey, and CSIC exert a comprehensive influence in the field of AMD remediation research.

Table 3. Top 10 most productive institutions from Jan 1990–Dec 2023	j.
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Institution	Publications	% of 2783	TLCS	TGCS	Average Citation Frequency
Univ Huelva	113	4.06	928	3396	30.05
Chinese Acad Sci	69	2.48	113	1317	19.09
CSIC	54	1.94	674	2471	45.76
US Geol Survey	54	1.94	430	2201	40.76
Univ Witwatersrand	52	1.87	206	1125	21.63
Penn State Univ	46	1.65	290	1624	35.30
South China Univ Technol	39	1.40	108	680	17.44
Univ Johannesburg	37	1.33	114	523	14.14
CSIR	33	1.19	230	633	19.18
Cent South Univ	30	1.08	4	288	9.60

Notes: CSIR: Council for Scientific and Industrial Research.

A total of 99 countries (regions) participated in AMD remediation research studies. Table 4 lists the contributions of the 10 most productive countries (regions). Among the top 10, there are three European, three American, two Asian, one Oceanian, and one African country. The USA leads the field with the highest number of publications (526 articles, 18.90%), TGCS (16985 times), and TLCS (3004 times), followed by China (502 articles, 18.04%), South Africa (264 articles, 9.49%), Spain (231 articles, 8.30%), and Canada (226 articles, 8.12%). These results indicate that the USA plays a critical role in this area, while China's research ranks high in terms of publication numbers yet demonstrates relatively low TLCS and TGCS values.

Country	Publications	% of 2783	TLCS	TGCS	Average Citation Frequency
USA, North America	526	18.90	3004	16,985	32.29
China, Asia	502	18.04	1147	10,203	20.32
South Africa, Africa	264	9.49	1009	4562	17.28
Spain, Europe	231	8.30	1485	6910	29.91
Canada, North America	226	8.12	1552	7800	34.51
Australia, Oceania	151	5.43	656	4278	28.33
Brazil, South America	151	5.43	406	2856	18.91
UK, Europe	141	5.07	1276	5621	39.87
South Korea, Asia	108	3.88	408	1835	16.99
France, Europe	99	3.56	511	3888	39.27

Table 4. Top 10 most productive countries/regions during Jan 1990–Dec 2023.

The relationship of national cooperation network is shown in Figure 2. China has cooperated with the USA, Canada, India, Japan, South Korea, and other countries or regions. The USA and China, as the countries with the first and second largest number of publications, have extensive cooperation with various countries. In addition, Portuguese-speaking and Spanish-speaking countries such as Brazil, Portugal, Spain, and Chile have maintained some cooperation. Strengthening international scientific research cooperation has gradually become an important trend in the field of global research. However, differences between multiple languages may hinder the speed and effect of national scientific research cooperation.



**Figure 2.** Cooperation between major countries/regions of AMD remediation research from January 1990–December 2023.

## 3.5. Highly Cited Publications

Citation frequency partially reflects the attention a research paper receives, as well as its importance and standing in academic communication [23]. Highly cited publications offer insights into influential authors and significant research topics within a field over time. Table 5 presents the top 10 cited publications in this field. Johnson DB's "Acid Mine Drainage Remediation Options: A Review", published in 2006 in *Science of the Total Environment*, holds the highest citation frequency. The article discusses the mechanisms of action, advantages, and disadvantages of chemical and biological techniques for HMs removal from AMD and classed the abiotic and biological systems as "active" and "passive" [24]. Ranked second in citation frequency is Acero P's 2006 study, "The Behavior of Trace Elements during Schwertmannite Precipitation and Subsequent Transformation into Goethite and Jarosite", published in *Geochimica et Cosmochimica Acta*. The study investigates

the formation mechanism of schwertmannite, a ubiquitous mineral formed by AMD, in the abandoned Monte Romero mine. Schwertmannite formation primarily arises from the precipitation of Fe(II) in supersaturated solutions through oxidation to Fe(III) and its subsequent transformation into pinnate and yellow potash ferrite [25]. "Trace Metal Adsorption onto an Acid Mine Drainage Iron (III) Oxyhydroxy Sulfate", by Webster JG and published in *Environmental Science & Technology* in 1998, ranks third in citation frequency. The paper compares the adsorption behavior of goethite-bearing iron(III) oxyhydroxy sulfate and schwertmannite-bearing iron(III) oxyhydroxy sulfate deposited in AMD in New Zealand, as the adsorption properties of AMD-associated Fe-rich precipitates differ from those of pure hydrous iron(III) oxides [26]. The publication of these papers has significantly contributed to advancing the field.

Title	Fist Author	Journal	Year	TLCS	Reference
Acid mine drainage remediation options: a review	Johnson DB	Science of the total environment	2005	518	[24]
The behavior of trace elements during schwertmannite precipitation and subsequent transformation into goethite and jarosite	Acero P	Geochimica et Cosmochimica Acta	2006	103	[25]
Trace metal adsorption onto an acid mine drainage iron (III) oxy hydroxy sulfate	Webster JG	Environmental Science & Technology	1998	100	[26]
Biological treatment of acid mine drainage under sulphate-reducing conditions with solid waste materials as substrate	Chang IS	Water Research	2000	91	[27]
Characterization and reactivity assessment of organic substrates for sulphate-reducing bacteria in acid mine drainage treatment	Zagury GJ	Chemosphere	2006	84	[28]
Schwertmannite precipitated from acid mine drainage: phase transformation, sulphate release and surface properties	Jönsson J	Applied Geochemistry	2005	83	[29]
Hydrogeochemistry and microbiology of mine drainage: An update	Nordstrom D K	Applied Geochemistry	2015	83	[30]
Removal of sulfate and heavy metals by sulfate reducing bacteria in short-term bench scale upflow anaerobic packed bed reactor runs	Jong T	Water Research	2003	77	[31]
The chemistry of conventional and alternative treatment systems for the neutralization of acid mine drainage	Kalin M	Science of the total environment	2006	74	[32]
Environmental impact and remediation of acid mine drainage: a management problem	Gray N F	Environmental Geology	1997	66	[33]

Table 5. Most cited publications of AMD remediation research from January 1990–December 2023.

## 3.6. Keyword Analysis

3.6.1. Keyword Co-Occurrence Analysis

Keywords usually reflect research themes and the core content of manuscripts; accordingly, statistical analyses of keywords can provide insight into research hotspots and future research directions [34]. After extracting keywords from papers on AMD remediation published from 1990 to 2023 and filtering and merging redundant terms, keyword clustering and analyses were performed using VOSviewer, and a knowledge map was constructed to visualize hotspots and dynamic trends in the field. In the keyword co-occurrence network map for AMD remediation research, there were five clusters (Figure 3, Table 6).



**Figure 3.** Keyword co-occurrence network view of AMD remediation research from January 1990 to December 2023.

Clusters	Keywords
Red cluster	Acid mine drainage; pollution; sediments; geochemistry; transport; tailings;
Red cluster	river
Yellow cluster	Iron; oxidation; schwertmannite; kinetics; pyrite; mechanisms
Blue cluster	Sulfate-reducing bacteria; sulfate reduction; reduction; performance;
Dide ciustei	reducing bacteria
Green cluster	Removal; adsorption; resource recovery; biosorption; recovery; fly ash

Table 6. Core keywords in different keyword clusters.

The red cluster represents environmental risk assessments. The oxidation of sulfide minerals (e.g., pyrite) promotes the generation of sulfuric acid, which subsequently promotes the release of metals, contributing to the high acidity and dissolved metal ion contents in AMD [35]. The extremely low pH of AMD makes it corrosive. The concentrations of metal ions in the aqueous environment increase [36]. AMD originates from the oxidation of sulfide minerals accompanied by the activation, release, and migration of heavy (analogous) metal elements (Fe, Cd, Cr, Pb, Cu, and As). HMs in the environment easily interact with organic or inorganic ligands to form complexes, which can be adsorbed by soil colloids, reducing their mobility; they are not easily dissolved and degraded by leaching and microbial degradation [37].  $Fe^{2+}$  is the most common metal ion in AMD. It is oxidized to generate  $Fe^{3+}$ , which is oxidized by dissolved oxygen in the water and precipitated into iron hydroxide, causing the bottom and sides of the water body to appear red, affecting environmental aesthetics [38]. When untreated, AMD is discharged into water bodies, such as rivers, lakes, and soils; the HMs in AMD then inhibit or prevent the growth and development of bacteria and microorganisms by inhibiting enzymatic activity, destroying cell structure, generating reactive oxygen species, etc. This leads to alterations in the physiology and morphology of plants, affecting growth, purification of the water body, biodegradation in the soil, and destroying the ecosystem [39]. HMs and other elements accumulate in the food chain, enter the human body through food intake and skin contact, and accumulate in vital organs and glands, such as the heart, brain, kidneys, bones, and liver, hindering brain development, inhibiting cellular metabolism, causing respiratory distress, and inducing tumor generation [40].

The yellow cluster represents AMD research focused on environmental geochemical cycling. Sulfide minerals in tailings are oxidized under the action of microorganisms, releasing a large number of toxic and harmful HM ions. Furthermore, complex physicochemical changes (e.g., oxidative evaporation, dissolution, and neutralization) lead to the production of secondary minerals, usually by the oxidation of pyrite, magnetic pyrrhotite, chalcopyrite, and other sulfide compounds. When the pH of the AMD environment is <3 and concentrations of sulfate and monovalent cations are high, pyrrhotite-like minerals are readily formed; when the pH is increased, schistosome minerals, hydromagnetite, and acicular iron ore can form [41]. Secondary minerals affect the physicochemical properties of AMD through the adsorption and co-precipitation of toxic metals and sulfates, which are closely related to the evaporation, oxidation, hydrolysis, and neutralization of AMD. These secondary minerals can control trace metal pollution through surface adsorption and co-precipitation. They play an important role in the migration and passivation of HMs in the environment and are the main determinants of trace elements in river surface sediments [37]. Advanced techniques, such as isotope analyses, atomic force microscopy, transmission electron microscopy-selected area electron diffraction, and high-resolution transmission electron microscopy, have been utilized to study the physical evolution of secondary minerals and the passivation of HMs.

The blue cluster is related to the application of sulfate-reducing bacteria (SRB) for AMD remediation. SRB is a group of anaerobic prokaryotic organisms commonly found in natural environments; more than 220 species belonging to 60 genera have been discovered to date [42]. Under anaerobic conditions, SRB can take sulfate, sulfite, and other oxidized forms of sulfur as the electron acceptor and organic matter (lactic acid, methanol, ethanol, etc.) as the electron donor and carbon source, reducing the sulfate in AMD to hydrogen sulfide. The hydrogen sulfide continues to react with metal ions in the wastewater, resulting in insoluble metal sulfide precipitation; thereby, the metals are effectively recycled and utilized while simultaneously generating alkaline substances to reduce the acidity of AMD [43].

Theoretically, SRB has various advantages for AMD treatment. However, in natural remediation, the lack of available organic waste as electron donors limits the growth of microorganisms, and the remediation effect is often not obvious. Therefore, it is necessary to enhance SRB treatment by adding a suitable carbon source and electron donor [44] or Fe0 [45] as a reinforcing agent to reduce sulfate. Researchers have designed a variety of bioreactors to gradually overcome the problems of complete mixing of sludge and AMD, including the complete utilization of biological sludge for wastewater treatment and the biofilm method for treatment of low loads, short flow, clogging, etc. Active and passive bioreactors (including microfiltration membrane bioreactors, upflow anaerobic sludge blanket reactors, and anaerobic baffled reactors) are promising treatment systems for AMD. SRB biochemical processes are susceptible to external factors, including temperature, sulfate and HM concentrations, carbon sources, AMD pH, redox potential, hydrogen sulfide toxicity, and hydraulic retention time, and variation in these factors can therefore affect the performance of the bioreactor [3]. In the future, it is necessary to focus on the regulatory mechanism underlying the remediation effects of SRB on AMD under the influence of multiple factors to achieve the maximum degree of transformation of a variety of HMs into sulfide minerals. Molecular biological approaches should be used to analyze microbial community succession and functional bacteria involved in the crystallization of metal sulfides in the sludge in the sulfate reduction system as well as to clarify interactions between metals and biological systems and to identify synergistic effects of SRB and other processes (e.g., constructed wetlands).

The green cluster is related to the application of adsorption in AMD remediation. Adsorption is recognized as an effective, efficient, and economical method for removing/binding inorganic contaminants and is promising for AMD remediation. The adsorption method can be divided into physical and chemical adsorption and can be classified according to the underlying mechanism, e.g., ion exchange, surface complexation, and

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precipitation [46]. Common AMD adsorbent materials include diatomaceous earth, clay minerals, bentonite, zeolite, seafoam, fly ash, and a variety of newly modified materials with layered structures and good adsorption properties [47].

Fly ash is a by-product of the coal-fired power generation process and is a potential low-cost adsorption material owing to its strong alkalinity, flocculation, large specific surface area, and porous structure. Analyses in different regions of the United States and India showed that fly ash can effectively neutralize AMD but can also lead to the secondary contamination of treated wastewater with oxyanion-forming elements [48]. As fly ash contains large quantities of stable silica, alumina, and other components, its scope of applications and effects is limited. Therefore, the surface modification or structural modification of fly ash is often necessary to enhance its activity, further improve the adsorption performance, and realize the comprehensive utilization of high-value-added materials. Low-grade fly ash can be used as a raw material to synthesize many different types of zeolites and geopolymers, which can be further applied to AMD remediation. Cardoso et al. synthesized Na-P1 molecular sieves using fly ash as a raw material for the removal of pollutants from acid mine wastewater under mild conditions. After treating acid mine wastewater for 30 min with synthesized zeolite, the rates of arsenic, nickel, calcium, copper, iron, and manganese ion removal were high, and ammonia, magnesium, potassium, and zinc ions also exhibited high removal rates [49]. Most recent studies of the adsorption performance of fly ash on HMs in AMD have been conducted in laboratory batch mode. Further studies should focus on the development of new fly ash-based lowcost adsorbents at the industrial level, the determination of the mechanism of action of fly ash-based adsorbents, and the demonstration of practical applications in real wastewater.

### 3.6.2. Keyword Burst Analysis

Salient terms can indicate trends and emerging research directions in a discipline. The basic principle of analyses of salient terms is to determine keywords in a field based on increases in frequency in the titles, abstracts, and keywords of documents [50]. To discover keywords that do not reach the established frequency threshold but contribute to research in the field, burst detection was further used to identify changes in the determined hotspots (Figure 4).



**Figure 4.** Top 50 keywords with the strongest citation bursts from January 1990 to December 2023 (with the periods from 1990 to 2012 and 2013 to 2023 shown on the left and right, respectively).

Based on an analysis of the top 50 keywords, from 1990 to 2001, beyond "acid mine drainage" and "acid rock drainage", the main keywords in this period were "oxidation", "acidification", "*Thiobacillus*", "*Thiobacillus ferrooxidans*", "solubility", and "geochemistry", indicating that research during this period was focused on the mechanism and regulation of AMD production and the geochemical characteristics of AMD. From 2002 to 2010, the main keywords were "sulfate-reducing bacteria", "bioremediation", "bioleaching", and

"temperature", indicating that there was a shift in focus to the bioremediation of AMD, especially the application of SRB and the influence of external factors, such as temperature, on the remediation effect of SRB. From 2011 to 2023, the main keywords were "adsorbent", "activated carbon", and "nanoparticle", indicating that research at this stage was mainly related to the bioremediation of AMD, especially the application of SRB in AMD repair and the effect of external factors. The co-occurrence of the keywords "adsorbent", "activated carbon", and "nanoparticle" reflects the extensive use of adsorption, with the large amount of research on activated carbon and other adsorbent materials, in AMD remediation. The further development of adsorbent materials that are highly efficient, low-priced, and reusable will be an important research direction. In recent years, "separation", "heavy metal ion", "metal recovery", and "rare earth element" have appeared in the literature. Resource recovery in AMD, especially the recovery of HM ions and rare earth elements (REEs), will be a key research object in the future.

# 4. Other Highly Investigated Remediation Technologies

To control AMD pollution, extensive research around the world has evaluated the causes of AMD and remediation methods. A large number of remediation technologies have been developed, and these can be divided into two major categories: end treatment and source control technologies. End-of-life pipe treatments and remediation technologies for AMD can be classified into active and passive treatments. The primary treatment technologies include neutralization, adsorption, iron oxidation, membrane separation, and electrochemical methods. Passive treatments include constructed wetlands, permeable reactive barriers, and sulfate-reducing bioreactors [51].

#### 4.1. Active Treatment

#### 4.1.1. Neutralization

Traditionally, the most common method for AMD treatment is neutralization, in which the pH is increased by the addition of neutralizing agents (e.g., calcium (hydr)oxide, calcium carbonate, sodium carbonate, sodium hydroxide, and magnesium oxide). This approach neutralizes acids and causes metal ions to form sulfides, hydroxides, and carbonates, thereby removing HMs from AMD [52]. Neutralization methods for AMD remediation produce a large amount of sludge rich in water and metals, cause the deposition of particles in the pipeline and equipment, require a large landfill for disposal, and cause secondary pollution; furthermore, HMs are transferred to the waste sludge and therefore are not completely removed [53]. It is, therefore, necessary to develop new low-cost and readily available neutralizers to reduce sludge production and improve the safe disposal and comprehensive utilization of HM waste. Iizuka et al. prepared a new neutralizer, PAdeCS, using concrete sludge and compared its effect with that of calcium hydroxide, a conventional AMD neutralizer. Although the amount of PAdeCS required to neutralize the same amount of AMD was 1.5 times higher than that of Ca(OH)<sub>2</sub>, the amount of sludge produced by PAdeCS was much smaller and the coagulation and sedimentation performance of PAdeCS was better than that of  $Ca(OH)_2$  [54].

#### 4.1.2. Membrane Separation

Membrane separation utilizes the selective permeability of membranes (nanofiltration and reverse osmosis membranes) to separate particles of different sizes driven by external pressure. Commonly used membrane separation technologies include nanofiltration, ultrafiltration, electrodialysis, reverse osmosis, and membrane distillation, among which nanofiltration and reverse osmosis are most common in the treatment of AMD [55]. López et al. evaluated the recovery efficiency of three different types of nanofiltration membranes for the treatment of AMD, with a poly(piperazinamide) active layer (NF270) showing high permeate fluxes, good retention of HMs and REEs, and a gradual increase in permeate flux with increases in the iron (III) concentration and acidity [56]. Membrane separation technology facilitates the selective separation and recovery of metal ions, reduces sludge generation, and produces high-quality wastewater. However, the selectivity of membrane separation technology is relatively low, the approach is only applicable to acidic media, serious membrane contamination can occur, and the lifespan is short.

#### 4.1.3. Ion Exchange

The ion-exchange method effectively removes and recovers HM ions from AMD through the reaction of inorganic ion exchangers (e.g., natural zeolites, synthetic zeolites, and sulfonated coal) and organic ion exchangers (ion-exchange resins) with the HM ions dissolved in AMD [57]. Ion-exchange resin, a kind of functional polymer material with a reticulated structure and special functional groups, is the most widely used ion-exchange material in AMD remediation. Fu et al. prepared a molybdenum sulfide-modified chelating resin with high selectivity and adsorption capacity for Hg(II), Cd(II), and Cu(II) in actual AMD [58]. Cheng et al. designed and synthesized a polyacrylonitrile-based ion-exchange fiber with polyamine and amidoxime groups with an adsorption capacity of 200.1 mg g<sup>-1</sup> for U(VI) in uranium-containing wastewater [59]. The repair and recovery of HMs and sulfate in AMD by ion-exchange technology is not only simple but also has benefits such as a high efficiency, large volume of treated water, and good quality of effluent; in addition, it does not cause secondary pollution of the environment. However, the resin used for ion exchange needs to be regenerated in large quantities, and the regeneration cost is high, which increases the cost of wastewater treatment.

# 4.2. Passive Treatment

# 4.2.1. Constructed Wetlands

A constructed wetland is a shallow water area constructed to imitate natural wetlands and is a comprehensive ecosystem with substrate-plants-microorganisms as the core. Adsorption, retention, filtration, precipitation, oxidation-reduction, microbial degradation, plant absorption, and other processes exert synergistic effects. HM ions are adsorbed in plant tissues through the root system and transformed into corresponding sediments. Microbial degradation and oxidation effectively remove HM ions from AMD, raising the pH value of the wastewater and improving the quality of the water body. Given the different constituents of AMD, it is possible to optimize the configuration of plants and microorganisms to improve the treatment efficacy of the constructed wetland [60]. Singh et al. evaluated the effectiveness of a horizontal submerged flow constructed wetland (HSSF-CW) with cow dung and bamboo chips as substrates and *Cyperus rotundus* as aquatic plants for the remediation of AMD; the pH increased from 2.1 to 6.4 in 6 months, while HM ions and sulfate removal rates reached 44–75% [61]. Compared with physical and chemical processes, constructed wetland systems for AMD treatment have the advantages of low investment costs, easy operation, easy maintenance, and low energy input. However, they also have disadvantages; for example, the substrate exhibits frequent clogging, covers a large area, and is sensitive to climatic and other factors, such as a large land area and long treatment time.

#### 4.2.2. Permeable Reactive Barriers

Permeable reactive barriers (PRBs) are filled with a combination of alkaline slowrelease materials, adsorption materials, catalytic oxidation materials, and other filler materials based on neutralization, oxidation, precipitation, filtration, adsorption, ion exchange, and other principles to remove HM ions from AMD; they are mainly used in tailing ponds or waste rock piles [62]. Sanchez-Ramos et al. (2023) evaluated the effectiveness of different industrial and agricultural wastes, such as sugar foam, paper mill sludge, drinking water sludge, and olive mill waste, as PRB fillers for the nominal removal of HMs in AMD. The removal of HM ions by paper mill sludge and drinking water sludge reached 55–99% and 55–95%, respectively [63]. The remediation effect of PRBs on AMD is affected by the type of filler material, flow rate, residence time in water, and other factors. With the chemical precipitation of pollutants and the growth of biofilms during the operation of PRBs, the porosity and hydraulic residence time are significantly reduced, resulting in a reduction in the removal of HMs and sulfate ions from AMD [64].

Currently, most AMD remediation and treatment measures have been evaluated individually and cannot comprehensively reduce the impact of AMD on the surrounding environment. While continuously developing new treatment technologies (such as electrochemical technology and microbial fuel cell technology), it is necessary to effectively couple active and passive treatments for the characteristics of AMD, focus on reasonable and effective combinations of different remediation measures, integrate the advantages of various processes and methods, strengthen the remediation effect of remediation measures, enhance the management of AMD remediation processes after the generation of waste, and research the resourceful reutilization of AMD remediation processes.

#### 5. Discussion

#### 5.1. Current AMD Remediation Technologies and Research Directions

In accordance with the composition of wastewater, concentration, emissions, sources, discharge characteristics, and site-specific conditions, controlling the source of pollution is more important than end treatment. The basic principle of source control is to control iron oxidation. This is mainly achieved by inhibiting the growth of iron-oxidizing bacteria, engineering coverage technology, and passivation treatment [65]. Microorganisms play an important role in the formation of AMD, and their ability to accelerate the conversion of Fe<sup>3+</sup> promotes AMD production. Therefore, by inhibiting or killing microorganisms, especially Thiobacillus ferrooxidans, bactericidal methods can effectively inhibit the production of AMD [66]. However, because the effective fungicide period is not long and is easily affected by the natural environment and climate (e.g., rain), some fungicides themselves may act as pollutants. Covering technology, to a certain extent, prevents contact between sulfur-containing minerals and O<sub>2</sub>; however, the cover layer is subject to rainfall, snow, and ice and can be damaged by the growth of plant roots. Long-term isolation cannot be achieved, regular maintenance is required, and the material is prone to drying out, thereby inhibiting the oxidation of tailings. The geographic and hydrological condition requirements are more particular. Passivation technology is currently one of the most promising methods. The basic principle is to form a layer of insoluble, inert cover film on the surface of sulfide mineral particles through chemical or physical reactions (such as organosilane-based coatings, biochar, anti-photo-corrosive materials, microencapsulation, carrier microencapsulation technology, and self-repairing coatings), resulting in the formation of a dense layer of inert protective film on the surface of the sulfide mineral to reduce or prevent contact between oxygen, water, microorganisms, and metal sulfide minerals [67]. Most research on surface passivation technology is in the laboratory stage and is based on pure pyrite, which does not reflect natural conditions. There are limitations concerning conditions, and the practical application of passivation technology is an important direction for the future of AMD remediation technology.

Traditional acidic wastewater treatment technologies have problems such as high treatment costs and low treatment efficiency. The release of AMD is a long-term process. It presents a huge safety hazard to the tailings pond, and long-term collection and treatment are expensive. Therefore, approaches from the end of treatment to source control are fundamental for resolving AMD pollution. Additionally, future AMD remediation technologies should represent a range from low efficiency and high cost to high efficiency and low cost. A combination of source control and end-of-pipe treatment technology should be used to effectively address AMD pollution.

# 5.2. Avenues for Future Research

Despite recent progress in AMD remediation technology, current methods only reduce the acidity of AMD and remove HM ions and sulfates, requiring continuous investment in chemicals and energy. The high costs, incomplete treatment, and secondary pollution (with the potential to impact the ecological environment) are major issues [68]. AMD is rich in valuable compounds that can be recycled, such as reclaimed water, sulfuric acid, HMs, and REEs. Recycling tailings as building materials and geopolymers can significantly reduce AMD formation [69].

There are various potential recyclable resources and recycling technologies for AMD. Recovering these resources from AMD for reuse can effectively reduce the cost of wastewater treatment and create economic and environmental benefits, realizing sustainable development and opening up new avenues for AMD treatment. The recovery of REEs from AMD is an important research topic. REEs, mainly the 15 lanthanides (atomic numbers ranging from 57 to 71), scandium, and yttrium, play essential roles in many high-tech industries and are key components of a wide range of advanced industrial materials, including superconductors, lasers, optical fibers, and aerospace alloys [70]. Currently, chemical precipitation, solvent extraction, ion exchange, and adsorption are the most widely researched techniques for recovering rare earth elements from solution [71]. However, the chemical composition of AMD is complex, containing a large number of other HM elements; therefore, the recovery of a single rare earth element is inefficient. Integrated processes must be developed to remove other HMs before precipitation, ion flotation, or adsorption processes to improve the purity of REEs in the resource recovery process. Rare earth element recovery technologies, such as magnetic separation, ionic liquids, and cloud point extraction, have broad application prospects and feasibility; however, several key challenges remain.

#### 6. Conclusions

With continuous economic and social development, mineral resources have become an essential part of human life. Inevitably, some pollution is generated during the development of mineral resources, and AMD is the biggest issue in the water environment in many countries. The remediation of AMD is often complex, expensive, and challenging and the cost and efficacy depend on the site conditions, composition of the wastewater, and treatment technologies. Therefore, the development of efficient, sustainable, environmentally friendly, and low-cost AMD remediation technologies is essential. In this study, the WOSCC database was used for a review of the literature on AMD remediation published between 1990 and 2023. The salient findings of this manuscript can be summarized thusly:

- (1) This analysis showed that research on AMD remediation has increased worldwide. The USA, China, and South Africa have the most publications in this field. Among the most prolific authors in the field, Ayora C, Nieto JM, and Dang Z were the most influential. The top three research institutions concerning publication number were the Univ Huelva, the Chinese Acad Sci, and the CSIC. Publications in this field were mainly published in *Mine Water and the Environment, Applied Geochemistry*, and *Minerals Engineering*.
- (2) The highly co-cited literature, keyword co-occurrence, and keyword burst analysis revealed that "ecological risks of AMD", "environmental geochemical cycling of AMD", "sulfate-reducing bacteria remediation", and "adsorption technology" were dominant research topics in the field of AMD remediation. Source control technology and the recycling of resources in AMD, especially the recovery of REEs, will be among the main focuses of research for AMD remediation in the future. Future AMD remediation technology should shift from end-of-pipe treatment to source control, with the simultaneous adoption of a combination of technologies and continued investment in the development of low-cost and efficient AMD remediation methods.
- (3) In this study, the current state and trajectory of AMD remediation research were meticulously evaluated through the application of quantitative bibliometric techniques. Notwithstanding, the present study solely utilized the WOSCC database, leaving room for future expansion through incorporating additional databases such as Scopus, Google Scholar, Dimensions, and PubMed for article databases or Derwent Innovations Index for patent databases. This integrated approach would consequently offer a more all-encompassing dataset and furnish robust theoretical underpinnings for the continued advancement of AMD remediation technologies.

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