REVIEW

Biosorptive treatment of acid mine drainage: a review

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Abstract

Acid mine drainage (AMD) is the biggest environmental problem of water related to the mining industry in many countries. Recently, coal mining and mineral mining industries use various AMD treatment systems. The main problem, however, is the very expensive and long retention times for treatment of AMD. For this reason, biosorption technology has been tested recently as a promising method for AMD treatment. The authors review presently available or potential biosorption methods to remove and detoxify toxic heavy metals and organic pollutants from AMD. Biosorption technology is evolving as an attractive option to supplement conventional AMD treatment. However, a literature review indicates that there is a lack of studies examining biosorption of AMD treatment. This paper examines a range of subjects, including laboratory scale experiments and the commercial application of biosorption for treatment of AMD. Our review paper will help direct future studies on the development of biosorption technology for treating AMD.

Keywords AMD (Acid mine drainage) · Biosorption · Mechanism · Heavy metal · Treatment

Introduction

Wastewaters from mining activities are a source of potential environmental problems. In mines, the target ore (gold, silver, copper, etc.) is often rich in sulfide minerals. The wastewater flows over or through sulfur-bearing materials and can generate acid mine drainage (AMD) (Johnson and Hallberg 2005; Nordstrom 2000). A general equation of the AMD production mechanism is:

$$4FeS_{2(s)} + 15O_{2(g)} + 14H_2O_{(l)} \rightarrow 4Fe(OH)_{3(s)} + 8SO_{4(aq)}^{2-} + 16H_{(aq)}^{+} \quad (1)$$
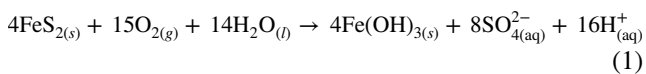
Equation (1) is a summary of a series of reactions. Subsequent reactions occur biologically and chemically. The first reaction is the oxidation of the sulfide mineral to sulfate solubilizes the ferrous iron. The next step is oxygen-dependent reactions. If the surrounding environment is sufficiently oxidizing, the Fe(II) will oxidize to Fe(III). Iron-oxidizing bacteria, many of which tend to be most active at pH 2.0

to 4.0, can increase the rate of this step (Schippers et al. 2010). Finally, when the pH of AMD is increased, either through contact with fresh water or neutralizing minerals, Fe(III) precipitates as $Fe(OH)_3$ in solution while simultaneously lowering pH. The $Fe(OH)_3$ may be harmful to aquatic systems. The precipitates reduce the amount of light that can penetrate the water, affecting photosynthesis and visibility for animal life. Furthermore, when the precipitate settles, it blankets the stream bed, smothering the bottom-dwellers and their food resources (Moses et al. 1987). Simultaneously, AMD can also dissolve other harmful metals (metalloids) and toxic substances. This phenomenon can contaminate drinking water underground. These environmental pollution problems caused by AMD affect many countries (Chen et al. 2014; Gaikwad and Gupta 2008).

To deal with these problems, a broad range of treatment technologies is available AMD. AMD treatment systems can be broadly categorized as "*source control*" and "*migration control*." Source control is a prevention technique used before the formation of AMD (Johnson and Hallberg 2005). Most source control techniques are to prevent or minimize important sources of reactants such as oxygen, water and sulfur-oxidizing bacteria in the processing of AMD (Lewis 2010; Santos and Johnson 2017). In contrast, migration control techniques are only to minimize the impact of receiving streams and rivers from AMD. In general, these techniques

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Editorial responsibility: Maryam Shabani.

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are divided into "active" and "passive" treatment (Akcil and Koldas 2006). More information related to this will be discussed in the next chapter.

One important aspect of environmental pollution caused by AMD is heavy metal contamination into receiving water bodies. If it is released into the environment without removal of hazardous heavy metals, this can cause entry into groundwater or leaching into soil and allow the negative effects to spread even further to harm all that use the stream or environment surrounding it (Hu et al. 2021; Kavehei et al. 2021; Simate and Ndlovu 2014). There are several methods for the removal of heavy metals from acid mine drainage such as chemical precipitation, ion exchange, filtration, oxidation, reverse osmosis, solvent extraction, and adsorption (Gaikwad and Gupta 2008; Hallam et al. 2021). However, these methods of treating AMD have a number of shortcomings including: extensive land utilization, production of large amounts of secondary solid waste, high capital cost and high operating costs. Among these techniques, adsorption is considered to be able to minimize the above disadvantages. Even though sorbents are highly effective in metal removal compared to other removal treatments, the high cost of sorbents is a still limitation. For this reasons, the most recent focus in sorption studies has been to investigate cheap adsorbents capable of replacing the expressive ones. Much research has focused on biosorption in recent years. Unlike bioaccumulation, biosorption technology uses dead biomass, so it is cheaper than other in the manufacture of adsorbents, and it is an economical technology because additional energy is not consumed to adsorption operate (Kratochvil and Volesky 1998; Park et al. 2010).

In the past, there was metal adsorption using biomass, but it is known that the first patent application with biosorption technology was B. Volesky and M. Tsezos in 1982 (Park et al. 2010). Since then, biosorption technology has been used in various fields. The paper in which biosorption technology is used for AMD treatment is known as the uranium adsorption paper using seaweed biosorbent published by Yang J. and Volesky B in 1999 (Yang and Volesky 1999). Since then, biosorption techniques have been used to treat AMD, but have not yet been commercialized. The purpose of this review paper is to discuss recent studies on AMD treatment using biosorption technology for commercializing this technology. No attempt has been made to cover all the available literature on AMD treatment in biosorbents. For this reasons, this paper can contribute greatly to future studies on the development of biosorption technology capable of removing contaminants from a variety of AMDs. First, before dealing with AMD treatment by biosorption, the paper, an overview of the traditional technology on the treatment of AMD is presented. Secondly, the paper investigated cases applied to research in order to find out where biosorption is used. Finally, this paper deals with the case where biosorption was applied single to AMD treatment and the case where it was used in combination with other treatment technologies.

AMD treatment

Source control

Treatment of AMD may be required for many years after mining activities have ended. Results of many experiments indicate that it may be possible to prevent AMD, rather than treating the site after mining. For this reason, appropriate control methods for the site during the early stages of mining would be beneficial (Salomons 1995). Those techniques are called "source control." The source control methods include: underwater storage of mine tailings, separation and blending of waters with mineral (alkaline) materials, and control of water migration (Johnson and Hallberg 2005).

Various methods exist to prevent or minimize the generation of AMD. First, oxygen methods are commonly used for source control. AMD is almost always begun by an aerobic microbial process in the presence of oxygen. Underwater storage has been used for disposing and storing mine tailings that may prevent contact between the minerals and oxygen. Constructing wetlands represents a modification of the more traditional wet cover method, and a water depth of at least 1.5 m can reduce atmospheric oxygen diffusion into the tailings. Wetlands can be classified as aerobic and anaerobic wetlands. Self-sustaining ecosystems that mimic their natural counterparts are classified as aerobic wetlands (Sheoran and Sheoran 2006). Often these consist of shallow ditches filled with flooded gravel, soil, and organic matter to support wetland plants. An oxygen-consuming organic layer is formed by adding plants, and the water depth can be reduced. Stoltz and Greger demonstrated that the presence of plants also decreased the release of metals by increasing the pH of the substrate (Stoltz and Greger 2006).

Other control methods aim to eliminate the process of sulfide oxidation. These methods involve adding solid-phase phosphates to pyritic mine waste to precipitate iron(II) as ferric phosphate (Kim et al. 1999). This can reduce the potential of iron to act as an oxidant of sulfide minerals. Application of anionic surfactants can be used to inhibit sulfur-oxidizing bacteria. Iron- and sulfur-oxidizing bacteria play an important role in generating AMD. Anionic surfactants have been used to inhibit sulfur-oxidizing bacteria activity in mineral spoils and tailings (Kleinmann 1990).

Migration control

Source control methods cannot always prevent the formation of AMD, and appropriate treatment methods need to

be implemented when AMD generation is unavoidable. In this case, the method used to minimize environmental pollution caused by AMD is called "migration control" (Johnson and Hallberg 2005). Migration control methods of mine sites generally require pH adjustment, oxidizing or reducing (redox) conditions, and/or stabilization of wastes. Biosorption technique is applicable to this method. Migration control methods are commonly classified as either "passive" or "active" and may involve combinations of physical, biological and chemical approaches Passive treatment refers to a method of treating AMD through minimal maintenance without using electricity, chemicals, and manpower, and active treatment is a method of treating AMD by using electricity, chemicals, and manpower (Gazea et al. 1996).

Passive treatment

Passive treatment is a method that provides a restrained environment in which natural chemical and biological reactions are used to remediate AMD. Recently, several types of passive treatment systems have been developed without additional chemical reagents. For this reason, passive treatment systems are expected to provide lower costs of construction, operation, and maintenance. The primary passive methods are largely divided into biological systems and non-biological systems with inorganic materials. Table 1 lists the currently used passive treatment methods according to the systems classification.

Anoxic limestone drains (ALDs) and open limestone channels (OLCs) are representative treatment techniques using limestone. ALDs are non-biological systems constructed using buried limestone cells through which anoxic water flows. In this system, limestone supplies additional alkali AMD in the system to avoid ferrous iron oxidation and precipitation (Hedin et al. 1994). OLCs are an open analog to ALDs. These systems are favorably constructed and operated on steep slopes (Alcolea et al. 2012). ALD and OLC have benefits compared to constructed wetlands, because they generate alkalinity at a low cost. However, they only provide short-term decrease in hydroxide precipitation. Another drawback is that, although limestone is an inexpensive and effective way to produce alkalinity, it must be exploited under suitable conditions or its effects are limited.

Table 1 Methods for passive treatment of AMD

Abiotic	Biological
Anoxic limestone drains (ALD)	Aerobic and anaerobic wetlands
Open limestone channel (OLC)	Permeable reactive barriers (PRB)
Steel-slag leach bed (SLB)	Vertical-flow systems
Low-pH Fe oxidation channels	Sulfate-reducing bioreactors (SRB)

Constructed wetlands can also be an effective method for treating AMD. Aerobic wetlands, one of the typical passive treatment methods, are a method for precipitating pollutants through aeration with sufficient residence time based on storage space. The best remediation by aerobic wetlands is with mildly acidic or net alkaline waters that are filled with soil or limestone gravel in an open pond that functions as a buffer. Aerobic wetlands can also remove heavy metals. The main drawback to this treatment system is that excessive accumulation of precipitate will severely limit the remediation abilities. In contrast, anaerobic wetlands encourage alkaline water to pass through buried organic substrates, such as a bed of limestone underneath or mixed with an organic substrate that requires exclusion of oxygen and aluminum in the water. These systems neutralize acidity and reduce metals to the sulfide form in organic substrate surface layers. They have the same maintenance and operating cost advantages as other passive treatment systems, but these systems commonly require long retention times and large surface areas to be efficient due to the slow mixing of acidic and alkaline substrate water near the surface (Fabian et al. 2005).

Among other passive treatment methods, permeable reactive barriers (PRBs) have gained recent attention. This method blocks AMD groundwater from underground water flow. PRBs are installed by burying layers of reactive barrier materials, such as limestone, zero valent iron, and organic matter. These materials can promote microbial sulfate reduction reactions and precipitation of iron and other metal sulfide minerals. During this process, these materials remove contaminants from AMD through a variety of reactions, such as adsorption, precipitation, and biological mechanisms. Therefore, the choice of organic matter in barriers is the most important factor in this process (Benner et al. 1999).

Biological reactors, which are referred to as sulfate-reducing bioreactors (SRBs), may be used to reduce sulfate in PRB or wetland systems. SRBs are based on their capacity for reducing sulfate to sulfide as an electron acceptor. As a result, bacteria precipitate as metal sulfides, resulting in increase in pH and alkalinity. For this reason, SRB is an economical treatment system for simultaneously removing metals and sulfides in AMD. However, the removal mechanisms of metals have not been identified, and efficient carbon source selection problems remain (Luptakova and Kusnierova 2005; Zagury et al. 2006).

Alkalinity-producing systems have also been called successive alkalinity-producing systems (SAPS) and reducing and alkalinity-producing systems (RAPS). The systems typically consist of a drainage layer, an organic layer, and a limestone layer. The difference between SAPS and RAPS is that SAPS pass AMD through each layer sequentially, while RAPS pass the material through a combined layer of organic material and limestone. The fundamental concept of these systems is related to the wetland method. These systems

have been used to treat high acidity and high concentrations of Fe^{3+}, Al^{3+}, and DO. These systems can also encourage metal precipitation in difficult treatment conditions that include low flow (Barton and Karathanasis 1999).

Active treatment

Passive treatment has many advantages related to maintaining the system. However, most passive treatments require high start-up cost and a large area. Those reasons can result in a move to active treatment methods, which (in its basic concept) is a low technology approach for AMD treatment. Active treatment systems use chemical and physical processes, such as precipitation, biological mediation, sorption, sulfate reduction, electrochemical concentration, and flocculation. Information on the various active treatment systems is summarized in Table 2.

Among active treatment methods that can treat AMD, inorganic alkaline precipitation treatment is the most common and cost-effective method. A wide variety of natural by-product alkaline reagents or manufactured chemical reagents can also be used, determined by availability, cost, and performance. These treatment methods involve addition of chemical neutralizing agents including lime, caustic soda, and calcium carbonate. These reagents increase the pH for precipitation of heavy metals as mainly hydroxide complexion to treat AMD. The reagents vary in cost and effectiveness. Thus, selecting the right reagents for the site is the most important factor for this treatment. For example, sodium hydroxide is less expensive and more effective than limestone. Although active treatment can provide an effective method of treating AMD, it has the disadvantages of sludge disposal problems and continuous operation costs.

Active treatment methods are divided into fixed plant and *in-situ* methods. Fixed plant treatment of AMD occurs in a fixed location and uses the AMD pump for the plant. The process includes adding and mixing reagents in one or more reactor tanks, collecting treated sludge, and discharging the treated water. Typically, the main advantage of stationary plants is that they can be designed to handle and achieve water quality targets and unexpected contaminants. An important limitation of a fixed plant is that, regardless of AMD source, the affected water must be delivered to the plant. Therefore, there is a significant additional cost when the AMD source is far from the plant. On the other hand, *in-situ* treatments have a relatively low capital cost regardless of distance, because they use portable and simple systems. All *in-situ* systems have storage and supply abilities to use similar reagents, such as limestone and calcium hydroxide. However, *in-situ* systems are limited by reagent, power supply, maintenance parts, and sludge disposal.
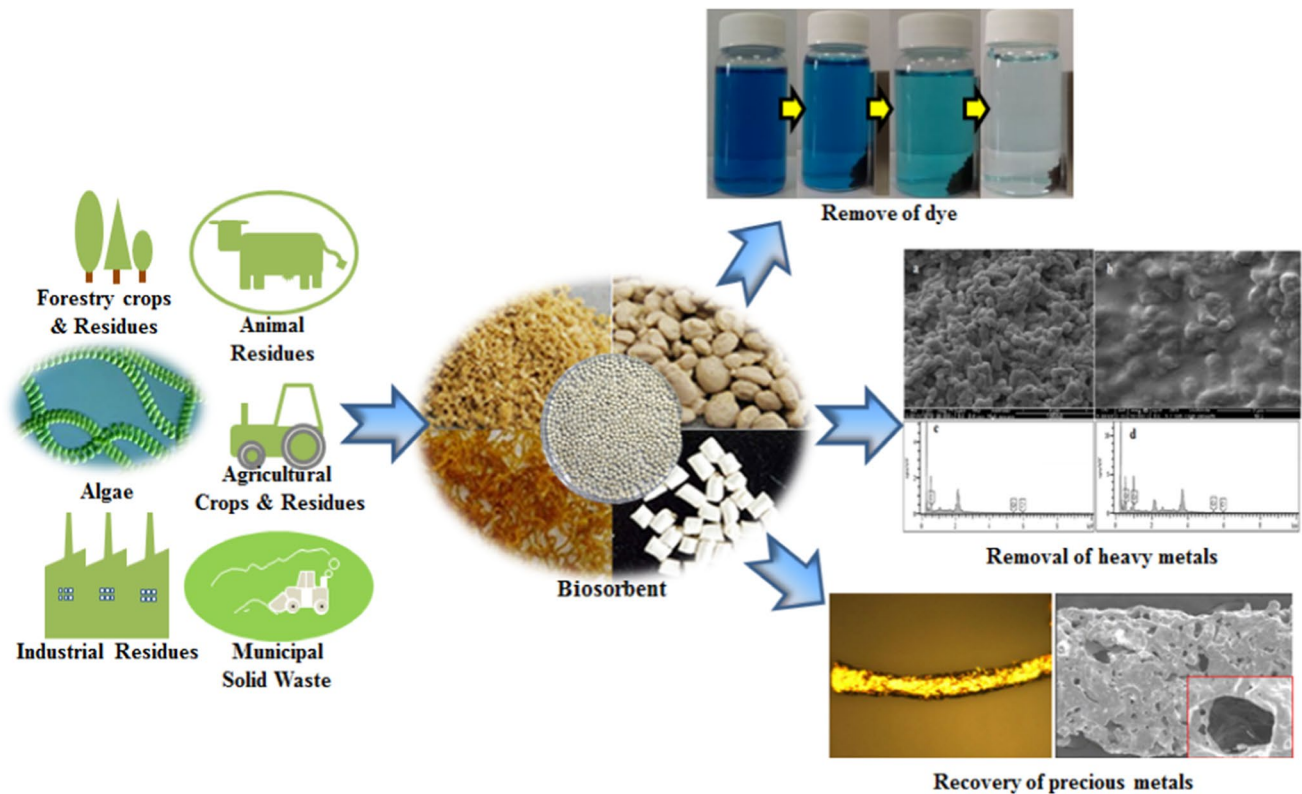
Biosorption

Definition of biosorption

Biosorption is a type of sorption technology and can be defined as passive uptake of ionic pollutants (heavy metals, dyes, and precious metals) by dead or inactive materials derived from biological sources (Volesky 2007). Figure 1 shows the overall process of biosorption. The mechanisms of biosorption are due to a number of metabolism-independent processes in the cell wall. The cell wall of the biomass is mainly composed of polysaccharides, proteins and lipids. The biomass cell wall has many functional groups, which are able to engage in physico-chemical interactions with pollutants ions. Functional groups most commonly involved in such interactions include carboxylate, hydroxyl, amine and phosphoryl groups present within the cell wall components (e.g., polysaccharides, lipids and proteins) (Naja and Volesky 2011). Therefore, the pollutant uptake will differ according to the biomass type.

A number of biological sources, such as algae, bacteria, fungi, industrial wastes, natural residues, bio-industrial wastes, and other biological compounds, have been investigated as cheap adsorbents capable of replacing well known but more expensive commercial adsorbents. When selecting a biomass for biosorption, one primarily considers cost effectiveness and the efficiency of ionic pollutants of the biomass. Many researchers have studied new biomass types with low cost and high efficiency. Furthermore, development of methods for pretreatment of biomass and physical or chemical surface-modification has been of interest for enhancing biosorptive capacity in recent years (Vijayaraghavan et al. 2008). In general, biomass pretreatment basically aims to clean up biomass to enhance biosorption capacity. Common chemical pretreatments involve acidic and basic pretreatments. Other chemical pretreatments include alkaline, ethanol and acetone treatments of the biomass

Table 2 Methods for active treatment of AMD, including fixed plant and *in-situ*

Methods	Sort	Principle
Low density sludge plants	Fixed plant	pH control / precipitation
High density sludge plants	Fixed plant	pH control / precipitation
Pulsed carbonate reactors	Fixed plant	pH control / precipitation
Electrochemical treatment	Fixed plant	Electrochemical concentration
Microbial reactor systems	Fixed plant	Biological mediation / redox control
Process water ponds	*In-situ*	pH control/precipitation
Heap leach ponds	*In-situ*	pH control/precipitation
Tailing dams	*In-situ*	pH control/precipitation
Channels, creeks, rivers	*In-situ*	pH control/precipitation

Fig. 1 Schematic of different processes for using biomass in various biosorption technologies

(Khosravihaftkhany et al. 2015; Puranik and Paknikar 1999; Vijayaraghavan et al. 2008). Several researchers also studied enhancing/modifying a particular functional group of the biomass (Deng and Ting 2005). Carboxyl, amine, phosphonate, sulfonate and hydroxyl functional groups present in the biomass were specifically studied. The various methods for enhancing/modifying biosorbents include the removal of interfering sites, replacing interfering sites with binding sites and the addition of ionic polymer coatings and/or grafts.

Application of biosorption

The advantages of biosorption are its eco-friendly products, absence of secondary pollution, and low cost. Thus, it has achieved significant attention for remediating industrial wastewater. In addition, biosorbents allow removal of contaminants by dead organisms. Thus, the biosorption process is simpler and less expensive than using living biomass that requires supply of nutrients and energy. Since the first study on biosorption that focused on copper adsorption by fungi, various types of biomass have been reported to remove heavy metals (metalloids) and radionuclides. In addition, biosorption studies and applications have broadened to include removal of organic pollutants and recovery

of precious metals including gold, platinum, palladium, and silver (Park et al. 2010).

Dye removal from dye wastewater

Wastewater containing dyes may be a serious hazard and can be toxic, carcinogenic, and even mutagenic to aquatic systems. Efficient dye removal from textile industry wastewater is one of the most urgent environmental challenges. Dyes are often difficult to remove from wastewater because they generally have a synthetic origin and a complex aromatic molecular structure (Aksu 2005). Biosorption has been studied as an effective method for treatment of dye wastewater. Various types of biomass have been used for biosorption of a wide range of dyes and are sorted as anionic (direct, acid, and reactive dyes), cationic (basic dyes), and nonionic (disperse dyes) (Fu and Viraraghavan 2001). Table 3 shows the biosorption of dyes by various biomass materials under various conditions. The main mechanism of dye biosorption is physico-chemical interactions, such as adsorption, deposition, and ion-exchange. Earlier studies that focused on removal of dye via biosorption reported many biosorbents available on a laboratory scale. However, many dye biosorption studies use synthetic dye wastewater, which does not consider behavior of these sorbents with different

Table 3 Biosorption of dye using different biosorbents

Dye sort	Dye	Biosorbent	pH / Time (h)	Uptake (mg/g)	Reference
Anionic dyes	Methyl orange	Rice husk ash	3 /10	252	Hosseinzadeh and Mohammadi (2016)
	Congo red	*Penicillium janthinellum*	5 / 24	344.8	Wang et al. (2015)
	Drimarine black	Peanut husk	1 / 3	48.54	Noreen et al. (2013)
	Reactive blue 19	Chitosan hollow fibers	3.5 / 1.5	454.5	Mirmohseni et al. (2012)
Cationic dyes	Methylene blue	Salvia mitiorrziza	7 / 2	100.0	Zhao and Zhou (2016)
	Methylene blue	Eucalyptus saw dust (Biochar)	7 / 2	178.6	Sun et al. (2015)
	Malachite green	Eggshells	- / 24	243.2	Podstawczyk et al. (2014)
	Acridine orange	Food waste (Hydrothermal carbonization)		60.24	Parshetti et al. (2014)
Nonionic dyes	Disperse red 1	*Aspergillus niger*	4 / 48	5.59	Fu and Viraraghavan (2002)

competitors or with physico-chemical parameters that can differ considerably in real conditions. For this reason, future research should be conducted using actual dye wastewater more actively.

Removal of heavy metals from industrial wastewater

Pollution of water by heavy metals occurs by various industrial and human activities and is one of the most important environmental problems of recent times. Conventional methods, such as precipitation, coagulation, oxidation–reduction, ion exchange, membrane, and adsorption, are expensive for removal of toxic heavy metal ions from wastewater due to the non-regenerable materials used and the sludge treatment costs (Fu and Wang 2011). The use of biosorption to remove heavy metals from aqueous solutions is one of the most recent developments in environmental technology. The main advantage of this technology is that the production and operating costs are less than those of other technologies. In addition, it has high efficiency, minimal chemical sludge, allows regeneration of biosorbent, and has a possibility of metal recovery. For these reasons, many researchers have studied the biosorptive capacities of various biomasses for removal of heavy metals.

Heavy metal biosorption occurs due to specific functional groups on the cell wall. Carboxyl groups and amine groups of the cell wall are the major functional groups used for removing metal ions. Carboxyl groups actively participate in the binding of cationic metals, and amine groups remove both cationic metals and anionic metals via electrostatic interactions or hydrogen bonding. The use of inexpensive and efficient materials for heavy metal biosorption is summarized in Table 4. Furthermore, biosorption requires a structural biosorbent, adsorption of multiple metals, desorption, mechanistic modeling, enhancement of biosorption capacity through modification of biosorbents, and continuous reactor studies. However, information on these studies is insufficient for process scale-up and for design of real plants.

Recovery of precious metals

Precious metals are broadly used in various industries due to their unique physical and chemical properties. However, the limited availability of precious metals has led to excessive increases in their prices. Therefore, recovery of precious metals from aqueous solutions is economically attractive.

Various methods have been employed to recover precious metals. The main technologies used for recovering precious metals from wastewater include solvent extraction, ion exchange, precipitation, electrolytic recovery, and the oxidation–reduction method. Different processes have demonstrated various efficacies for different metals. Some traditional methods for recovering precious metal are not only inefficient, but they also damage the environment. Therefore, advanced recovery technologies, such as electrochemical and bio-metallurgical, have recently been used to overcome the drawbacks of these traditional methods. Currently, biosorption is a rising technology for recovering precious metals from aqueous solutions. For this reason, research has focused on biosorption to replace conventional methods to recover precious metals. Table 5 summarizes the various biosorbents used for precious metal biosorption.

As a result of analyzing a number of papers, the current precious metal recovery research using biosorption technology is focusing on improving the performance of the biosorbent by using various surface modification methods. Research is also being conducted to apply the developed high-performance biosorbent to pilot-scale continuous processes.

Table 4 Biosorption of heavy metals using different biosorbents

Metal	Biosorbent	Uptake (mg/g)	Condition	Reference
Cd(II)	Risk husk (NaOH-treated)	11.39	pH 6	Li et al. (2015a)
	Chestnut bur	34.77	pH 4	Kim et al. (2015a)
	Brown macroalgae (*Lessonia nigrescens*)	109.5	pH 3.7 / 5 days	Gutiérrez et al. (2015)
	UV-mutant *Bacillus subtilis*	208.08	pH 5	Wang and Sun (2013)
	Crambe abyssinica Hochst seeds	19.34	pH 6	Rubio et al. (2013)
	Moringa	7.86	pH 7	Meneghel et al. (2013)
	Spent coffee grounds	19.32	pH4-8	Kim and Kim (2020)
Cr(III)	Peach palm sheath modified (*Agaricus Blazei*)	19.42	pH 5	Massocatto et al. (2015)
	Spent grains residual	16.7	pH 5	Ferraz et al. (2015)
	Fugal biomass (*Termitomyces clypeatus*)	24.84	pH 4 / 60℃	Fathima et al. (2015)
	Garden grass	19.4	pH 4	Sulaymon et al. (2014)
	Algal biomass (*Ulva* sp)	3.97 (mmol/g)	pH 4.5	Vijayaraghavan and Joshi (2014)
Cu(II)	Bagasse pith	21.57	pH 6.5 / 323 K	Liu et al. (2016)
	Potato leaf powder (chemically treated)	42.74	pH 6.5	Moyo et al. (2016)
	Algal biomass (*Cystoseira crinitophylla*)	160	pH 4.5	Christoforidis et al. (2015)
	Alginate immobilized bentonite	131.58	pH 5	Tan and Ting (2014)
Pb(II)	Rhodophyte (*Eucheuma denticulatum*)	81.97	pH 5	Rahman and Sathasivam (2016)
	Fir bark (Modified by sodium hypochlorite)	0.966 (mmol/g)	pH 5	Su et al. (2015)
	Porous lignin-based sphere	31.8	pH 5	Li et al. (2015b)
	Sewage-sludge	98.45	pH5	Seo et al. (2020)
	Brown algae	1.363 (mmol/g)	pH 3	Moghaddam et al. (2013)

Treatment of AMD using biosorption

Single system

As mentioned in the previous chapter, biosorption technology is increasingly being applied in various fields. However, there are very few cases of biosorbents that have succeeded in industrialization due to the lack of research on actual wastewater treatment (Beni and Esmaeili 2020). Thus, much work in this area tries to demonstrate its possibilities on actual wastewater treatment systems. One of the targets of the study to the actual waste water is AMD. As mentioned in the previous chapter, acid mine drainage is different from general wastewater pollution as it is represented by low pH and high content of heavy metals and since the pollution generation scale is very large, it requires a lot of cost to treat it. For this reasons, research is being conducted to treat AMD using biosorption technology directly, which has an advantage in terms of cost and is known as an eco-friendly technology.

The technology that treated AMD using biosorption technology can be divided into a technology that uses only biosorbents and combines technology with other treatment technologies. Treatment of AMD using biosorption single systems is summarized in Table 6. Normally, a series of batch treatment studies and lab-scale continuous column studies is conducted to optimize the design parameters and conditions for real-scale treatment processes. Batch experiments generally tested the factors influencing biosorption, which are important in the assessment of the biosorption potential of any biomaterial. Batch experiments must be used to evaluate the required fundamental information including optimum experimental conditions, biosorbent efficiency, biosorption rate, and the potentiality of biomass regeneration. Various batch studies were conducted to treat AMD by biosorbents. The biosorption of AMD using chitosan (or chitin) is one of the more frequently reported approaches for

Table 5 Biosorption of precious metals using different biosorbents

Metal	Biosorbent	Uptake (mg/g)	Condition	Reference
Gold	Chitin	58	Computer microprocessor leachate solutions	Côrtes et al. (2015)
	Shewanella putrefaciens (Heat treated cells)	749.14	pH 3	Varia et al. (2014)
	PEI-modified bacterial biosorbent fiber	421.1	pH 5.5	Park et al. (2012)
	Bacillus cereus	153.41	–	Mohd Bahari et al. (2013)
	PEI-modified C. glutamicum biomass	361.76	pH 3.0	Kwak and Yun (2010)
Lithium	Ca—Alginate beads	0.562	Neutral pH	Song et al. (2013)
Silver	Verde-lodo clay	0.570 (mmol/g)	283 K	Cantuaria et al. (2016)
	Sulfonated gum mushroom biocomposite	140.8	pH 6.0	Das et al. (2015)
	Ag^+—imprinted chitosan gel beads	119.25	298.15 K	Zhang et al. (2015)
	Waste yeast	41.8	pH3, 288 K	Zhao et al. (2015)
	Chemically modified chitosan resin	2.1 (mmol/g)	–	Donia et al. (2007)
Platinum	Persimmon powder (glycine-modified)	1.28 (mmol/g)	pH 1.0	Gong et al. (2016)
	Escherichia coli biomass	45.65	pH 1.2	Kim et al. (2015b)
	Indian almond (tannin)	22.50	pH 2.0	Ramakul et al. (2012)
	Poly(allylamine hydrochloride)-modified E. coli	348.8	pH 2.5	Mao et al. (2010)
	PEI-modified E. coli	108.8	Pt-containing ICP wastewater	Won et al. (2010)
Palladium	2-aminobenzaldehyde modified chitosan Schiff's base	275 ± 0.4	pH 5.0	Monier et al. (2016)
	Methanogenic granular sludge	1.62	30 °C	Pat-Espadas et al. (2016)
	Ion-imprinted chitosan fibers	326.4	pH 1.0	Lin et al. (2015)
	Polyethylenimine-grafted spun-fiber made of cellulose nanofibril	26.5	pH 1.0	Hong et al. (2020)
	Escherichia coli biomass	38.87	pH 1.2	Kim et al. (2015b)

removal of pollutants in batch experiments First, chitosan is one of the most plentiful and inexpensive types of biomass. Second, it presents a significant number of functional groups in the biomass cell wall, such as amino/acetamido and hydroxyl groups. Functional groups in the chitosan have helped to remove various metallic ions from effluents. Furthermore, natural chitosan has been modified by many techniques to not only improve its sorption capacity, but also its application potential in support materials for immobilization. Due to its high adsorption capacity, biosorbent research has increased in recent years (Sarode et al. 2019). Most recently, magnetic chitosan and bentonite composites were synthesized and used to treat AMD for heavy metal removal. The sorbents containing chitosan showed excellent performance for actual remediation of AMD containing heavy metals (Cd, Cr, Cu, Fe Zn, Ni, and Pb). The percentage biosorbent removal of heavy metals from actual AMD was greater than 84% (Feng et al. 2019). Robinson-Lora and Brennan (2009) also studied chitin as a biosorbent for AMD treatment in a continuous system and indicated that chitin can play a major role in neutralization of AMD and removal of metal contaminants, such as sulfate (Robinson-Lora and Brennan 2009). Moreover, chitosan is naturally associated with $CaCO_3$ and proteins, allowing gradual release of alkaline species and nitrogen into the aqueous system. As a result, the previous neutralization treatment is not required to control the pH of the wastewater, thus reducing costs and treatment time. Laus et al. (2007) noted that chitosan could remove Fe(II), Al(III), and Cu(II) from AMD with less than 7 g/L of biosorbent, and the pH increased from 2.58 to 6.20 (Laus et al. 2007). Some researchers have studied the performance of crab shell products in removing pollutants from actual AMD in batch-scale study(Daubert and Brennan 2007; Pinto et al. 2011).

According to the literature, there have been many studies on the use of industrial or agricultural biowaste as biosorbents for removal of heavy metals from AMD. The availability and low cost of these types of biowaste make them attractive as biosorbents (Park et al. 2010). Rice husks are an example of biowaste used for biosorption of Cu, Fe, and Zn from AMD (Chockalingam and Subramanian 2006). Bark was also assessed as a biosorbent in the treatment of AMD.

Table 6 Results from the literature on treatment of AMD using biosorption single system

Target AMD		System	Biomass	Capacity	Reference
Synthetic	Fe(II), Al(III), Zn(II), Mn(II), Ni(II), Cu(II), and Cd(II)	Continuous	Peat biofilter	$Cd < Mn < Zn < Al < Ni < Cu < Fe$	Champagne et al. (2008)
	Zn(II), Cu(II)	Continuous	non-viable activated sludge	Zn(II): 3.4 mg/g, Cu(II): 5.9 mg/g	Utgikar et al. (2000)
	^{226}Ra	Batch	Sargassum sp., Penicillium sp., Saccharomyces cerevisiae and Monoraphidium sp.	Sargassum sp.: 99.6% Penicillium sp.:99% Monoraphidium sp.: 98% Saccharomyces cerevisiae: 86%	Nascimento et al. (2006)
Real	–	Batch	*Ectodermis* of *Opuntia* (H_2SO_4-treated)	Fe: 6.43 mg/g, Zn: 0.57 mg/g	Barrera et al. (2006)
	Southwestern United States	Batch	Chitin products	Fe: 60 mg/g, Pb: 0.5 mg/g, Zn: 39.5 mg/g	Pinto et al. (2011)
	Ingaldahl copper mines (Karnataka, India)	Batch	*Eucalyptus tereticornis* (Smith) bark	Fe: 96%, Zn: 75% Cu: 92%, SO_4^{2-}: 41%	Chockalingam and Subramanian (2009)
	Coal mine (Siderópolis, Brazil)	Batch	Chitosan microspheres	Fe(III), Al(III) and Cu(II)	Laus et al. (2007)
	Ingaldahl copper mine (Karnataka, India)	Batch	Rice husk	90% Fe(II) 89% Zn(II) 75% Cu(II)	Chockalingam and Subramanian (2006)
	Rand uranium mine (Randfontein, South Africa)	Continuous	Sea shell	Fe: 22.5 mg/g Mn: 1.98 mg/g	Masukume et al. (2014)
	Metallic mine (Korea)	Batch/Continuous	Iron-rich sludge	As(V): 98% As(III): 2%	Yang et al. (2014)
	Malan Mining (Shanxi, China)	Batch/Continuous	Chitosan	Cu, Ni, Zn, Cd and Pb ≥ 89% Fe ≥ 84%	Feng et al. (2019)
	Snow Shoe area (Pennsylvania, USA)	Batch/Continuous	Crab-shell chitin	-	Robinson-Lora and Brennan (2009)

The bark was able to remove 96% of iron, 92% of copper, 75% of zinc and 41% of sulfate from AMD (Chockalingam and Subramanian 2009).

A majority of the available data on treatment of AMD by biosorbents has been produced by batch experiments. However, batch-scale results are challenging to apply to real AMD, because the volume of mine effluents requiring treatment is on a huge scale. It is more advantageous to use a continuous reactor type system that is easier to maintain and can continuously treat pollutants than a batch reactor type for the industrialization of AMD treatment system. Therefore, a continuous system is needed for real AMD treatment applications. Despite the ample research on biosorption with batch testing, there are a few biosorbents employed in continuous systems. Yang et al. (2014) studied iron-rich sludge as a biosorbent for treating arsenic in AMD in a continuous system through a column reactor. In that study, the breakthrough point of arsenic was 200 bed volumes, and the sorption capacity was 9.3 mg/g. The main mechanism of As removal in this sludge is adsorption onto iron oxides. This

paper showed that not only the surface of the sludge, but also the substances constituting the sludge can be involved in the adsorption of heavy metals in AMD. (Yang et al. 2014). Non-viable activated sludge (non-living sludge) was evaluated in the presence of a synthetic solution of copper and zinc (which closely mimicked AMD composition) using packed-bed flow columns (Utgikar et al. 2000).

Combined systems

Each treatment processes for AMD has unique advantages. Often, treatment requires a combined strategy to minimize the effect of variability on treatment performance. For example, SAPS combines treatment concepts from both wetlands and ALDs. Biosorption for treating AMD can also be performed with existing treatment systems such as precipitation (Santos et al. 2004), bioreactors (Choi and Lee 2015; Hurtado et al. 2018) and various passive treatment systems (Groudev et al. 2008; Jeen and Mattson 2016). These combined systems compensate for the defects in the existing

treatments of AMD. Table 7 lists treatment facilities that combine biosorption with various AMD processing systems.

Precipitation using alkaline reagents is the most commonly used active treatment method for removing metals from AMD. It also has found its wide application for metals removal. Research has been conducted to combine precipitation and biosorption to more effectively remove heavy metals present in AMD (Holub et al. 2018). Although the process is cost effective, this method generates large volumes of hazardous, concentrated, mixed-water sludge. It can also be extremely difficult to filter and requires large and expensive solid–liquid separation units. To solve this problem, biosorption treatment was used to produce an easily filterable pulp and a solution containing metals. The study concluded that addition of biomass (grape stalks or cork powder) to synthetic AMD removed iron from the solution and improved the rate of sedimentation and filtration operations (Santos et al. 2004).

Choi and Lee (2015) studied the effects of a combined biosorbent and bioreactor system for treatment of AMD. The reactor consisted of two parts, calcined eggshell powder as a biosorbent and supernatant of organized microalgae. In this reactor, the first part of the process removes heavy metals using biosorption. In addition, the calcined eggshell neutralized the acidity from the AMD in this combined system. The combined reactors using the eggshell and microalgae hybrid system showed total heavy metal removal values from AMD effluents of 99.66% for Fe, 99.47% for Cu, 99.90% for Zn, 99.81% for Mn, 100% for As, and 100% for Cd (Choi and Lee 2015).

Heavy metals are a problem, but high sulfate concentration is also a problem in AMD. Thus, SRB, a biological reactor, has recently been used for efficient removal of metals and sulfide. A study using biosorption technology to increase SRB efficiency has also been reported (Hurtado et al. 2018). According to that study, it was possible to remove metal and sulfate from AMD through the synergistic combination of a biosorption treatment system with an SRB to eliminate copper.

Jeen and Mattson (2016) studied column experiments for passive treatment systems of AMD from a waste rock storage area. They evaluated biomaterials such as chicken manure, mushroom compost, and straw in comparison to limestone for use in the column reactor. All the column reactors had the following results: sulfate was reduced to sulfide, pH was increased, alkalinity was generated, and heavy metals were removed from the pollutants. These results revealed that the reaction mechanism for heavy metal removal is sulfate reduction followed by precipitation of sulfides, secondary carbonates, and hydroxides. As a result, precipitation and biosorption onto a biomaterial contributed to removal of metals from this system (Jeen and Mattson 2016).

PRB can also be used in a combined system with biosorption for treatment of AMD. Groudev et al. (2008) treated AMD during a 10-year period using different passive systems including ALD, wetlands, a rock filter, and permeable reactive multi-barriers. These methods were used separately or in different combinations. In their study, biosorption technology was combined with permeable reactive multi-barriers. A section of the multi-barriers was filled with a mixture of biodegradable solid organic substrates, such as cow manure, plant compost, and straw. In that study, much of the As, U, and Ra and most of the heavy metals were removed by increase in pH and biosorption (Groudev et al. 2008).

Conclusion

In this review, we presented several biosorption technologies that have been employed for AMD treatment. Relative to biosorption literature, the treatment of AMD via this process is still in its infancy. As discussed in this review, various biosorbents have been used for AMD treatment, which can occur through many mechanisms and can target various contaminants. Furthermore, it may be possible to reduce the cost of biosorbent use and improve the design of the process. Treatment of AMD is not an easy task due to the large scale. Therefore, economics can be the most important factor in

Table 7 Results from the literature on treatment of AMD using biosorption combination systems

Target AMD	System	Biosorbent	Performance	Reference
Yongdong mine (Gangneung city, Korea)	Calcined eggshell and microalgae hybrid system	Waste eggshell (Treated calcium carbonate)	99.47 to 100% (Fe, Cu, Zn, Mn, As, and Cd)	Choi and Lee (2015)
Copper mine (Andean Mountains, Chile)	SRB + Biosorption	Bacillus sp.	Until 23 days	Hurtado et al. (2018)
Metal (gold) mine	Layered and mixed passive treatment	Straw, chicken manure and mushroom compost	Until 175 days	Jeen and Mattson (2016)
Uranium deposit (Curilo, Bulgaria)	Wetlands, ALD, PRB and a rock filter	Cow manure, plant compost, straw	Until 10 years	Groudev et al. (2008)

the choice of technology. In most cases, expensive technology cannot be applied in a low-income country. Biosorption represents a technique for using economical, biological materials for treatment of AMD. This review paper can help in guiding future studies on the development of biologically-based sorbent materials capable of removing contaminants from a variety of AMDs.

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