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Review

Acid mine drainage: Prevention, treatment options, and resource recovery: A review

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ABSTRACT

Acid mine drainage poses severe environmental pollution problems due to its high acidity, toxic metals and sulphate contents. In this review, the available prevention of acid mine drainage generation, treatment options and their importance in light of the future perspectives are briefly discussed. The possible resources to be recovered such as ferric hydroxide, ferrite, rare earth metals, sulphur and sulphuric acid and their economic benefit are discussed. Furthermore, the importance of mine tailing for stabilisation of contaminated soil and production of building materials are highlighted. Overall, this review has shown that the resource recovery and reuse is a non-debatable holistic approach to environmental sustainability and acid mine drainage pollution reduction. Finally, the future perspective and areas that deserve in-depth exploration are underscored.

Contents

1. Introduction

Acid mine drainage (AMD) is a major problem all over the world,

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especially where coal and gold mine activities are common. Once AMD is generated, it is difficult to control the process and the treatment also requires high cost (Aguiar et al., 2016; Baruah and Khare, 2010; Grande et al., 2010; Qureshi et al., 2016). AMD causes severe environmental impacts, particularly on soil, water resources and aquatic communities (Galhardi and Bonotto, 2016; Shim et al., 2015). The main source of AMD is oxidation of sulphide mineral ores, which are initially exposed to the environment by intensive mining activities. In particular, among the metal sulphides, pyrite ore (FeS_2, commonly known as fool's gold) is one of the main mineral responsible for generation of AMD due to its ease of oxidation when exposed to oxygen, water, and microorganisms (Blodau, 2006; Chen et al., 2014b; Han et al., 2015; Pierre Louis et al., 2015; Plante et al., 2014, 2012). The pyrite oxidation is represented by different reactions under different conditions. For example, reactions (1)–(6) represent the major and common processes for pyrite oxidation. Reaction (1) shows oxidation of pyrite under molecular oxygen in the presence of excess water at neutral pH (Neculita et al., 2007). The overall reaction for pyrite oxidation (reactions (1)–(3)) is represented by reaction (4). Reaction (5) shows complete oxidation of pyrite where ferric acts as the oxidizing agent, and is also deemed a faster reaction than reaction (1) (Pierre Louis et al., 2015). Similarly, the complete oxidation of pyrite in the presence of low water content is represented by reaction (6) (Chen et al., 2015).

$$FeS_2 + 7/2O_2 + H_2O \rightarrow Fe^{2+} + 2SO_4^{2-} + 2H^+ \tag{1}$$

$$Fe^{2+} + 1/4O_2 + H^+ \rightarrow Fe^{3+} + 1/2H_2O \tag{2}$$

$$Fe^{3+} + 3H_2O \rightarrow Fe(OH)_3 + 3H^+ \tag{3}$$

$$FeS_2 + 15/2O_2 + 7/2\ H_2O \rightarrow Fe(OH)_3 + 2SO_4^{2-} + 4H^+ \tag{4}$$

$$FeS_2 + 14\ Fe^{3+} + 8H_2O \rightarrow 15Fe^{2+} + 2SO_4^{2-} + 16H^+ \tag{5}$$

$$FeS_2 + 15/4O_2 + 1/2H_2O \rightarrow Fe^{3+} + 2SO_4^{2-} + H^+ \tag{6}$$

AMD poses a severe pollution problem to current and future generations, especially due to low pH, high concentrations of potentially toxic dissolved metals, metalloids, and sulphate (Anawar, 2015; MacIngova and Luptakova, 2012; Modabberi and Alizadegan, 2013; Ouyang et al., 2015). Among the dissolved metals, Fe(II) is the most abundant and common in most of AMD. Fe(II) in AMD reacts with dissolved oxygen to produce iron oxide precipitates, which is commonly called “yellow boy,” and can smother life all along the way by embedding on stream or ocean beds. Consequently, small aquatic life that feeds from the bottom of the ocean or streams can be severely affected and may finally die out (Han et al., 2015). Further, the problem does not end with small aquatic life; it has also a negative impact on the food chain. Furthermore, formation of ferric hydroxide precipitate, aggravates the condition by lowering the pH and damage most of the microorganism existing in it (Agrawal and Sahu, 2009). Due to the corrosive nature, AMD interacts with rocks containing different types of mineral ore and easily provoking the solubility of toxic metals. The generated AMD water elevates the level of dissolved metal in the receiving surface water stream and negatively affects the stream biota.

The degree of environmental pollution by AMD is dependent on its composition and pH, which in turn may vary depending on the geology of the mine sites or sources. For example, water draining at Maurliden mine in Sweden is highly acidic (pH 2.3) and rich in Zn (~460 mg L^{-1}) and iron (~400 mg L^{-1}), and contains smaller concentrations (0.3–49 mg L^{-1}) of other metals such as Mn, Co, Cd, Mn, Ni and As (Hedrich and Johnson, 2014). In South Africa, the Fe(II) is very high; however, source dependent variations in concentration from place to place are reported. For example, the Fe(II) concentrations detected in coal and gold mines were 2135 mg L^{-1} and 835 mg L^{-1}, respectively (Kefeni et al., 2015a). A recent report from Spain showed a presence of 2040 mg L^{-1} Fe(II) and 194 mg L^{-1} Al(III) in AMD (Carrero et al., 2015). A predominance of Fe(II), Al(III) and magnesium sulphate from the Tharsis mining area of the Iberian Pyrite Belt (SW, Spain) (Valente et al., 2013), and variations in concentrations of Fe(II) (694–845 mg L^{-1}) and sulphate (2853–3622 mg L^{-1}) in Tinto Santa Rosa AMD (SW Spain) (Asta et al., 2010), have also been reported. These results clearly show the wide variation of mineral content and dissolved ions in AMD according to the geological strata of mining areas. Therefore, ensuring long-term environmental sustainability will require effective and efficient technology that can minimize the negative impacts of AMD (Bussiere, 2009). Some severe AMD impacts on the environment are presented in Fig. 1.

Treatment of AMD which is composed of several dissolved toxic metals is too complex and expensive. If AMD is not managed properly, it causes considerable environmental degradation, water and soil contamination, severe health impact on nearby communities, biodiversity loss and aquatic ecosystem (Albanese et al., 2014; Anawar, 2015; Durand, 2012; Hakkou et al., 2009; Khalil et al., 2014; Lghoul et al., 2014; Luptakova et al., 2012; Mulopo, 2015; Nejeschlebová et al., 2015; Ngure et al., 2014; Niane et al., 2014; Roychoudhury and Petersen, 2014; Vaněk et al., 2013). These negative environmental impacts pass from generation to generation. Therefore, in order to preserve and protect the environment and enhance ecological sustainability, proper prevention of AMD generation should be one of the important preconditions. However, once AMD is formed, immediate remediation and a rehabilitation paradigm shift become imperative. Innovative remediation of AMD in recent years has included sequential or selective removal of soluble metals and other toxic environmental pollutants. Among all remediation options developed for AMD until now, limited numbers are cheap and sustainable while most are expensive and even unaffordable (Anawar, 2015). Some of the existing treatment methods of AMD mainly focus on neutralising, stabilising and removing pollutants through various physical, chemical and biological processes (MacIngova and Luptakova, 2012). There are several available cost-effective innovative methods of AMD remediation in the literature such as biological passive treatment using sulphate-reducing bacterium (SRB), environmental available materials (eg. dairy manure, bentonite (aluminium silicate clay), lignite and zeolite), magnetic nanoparticles (MNPs), membrane technology etc., however, the traditional neutralisation treatment of AMD is still widely used.

In this review, more emphasis is placed on prevention and recent treatment options of AMD. The available prevention of AMD generation and details of the advantages and disadvantages of the conventional and the most recent treatment options of AMD are highlighted and summarised. The necessary precautions required and future perspectives are presented. In general, the main objective of this review is to provide up-to-date information on prevention of AMD generation and treatment options in comparison with the traditional neutralisation techniques. Finally, the possible resources to be recovered from AMD, a future perspective and research needs are addressed to inspire further innovative research in AMD generation prevention and remediation.

2. Methods

This review is devoted to provide up-to-date prevention of AMD generation and treatment options. A literature search in the Scopus

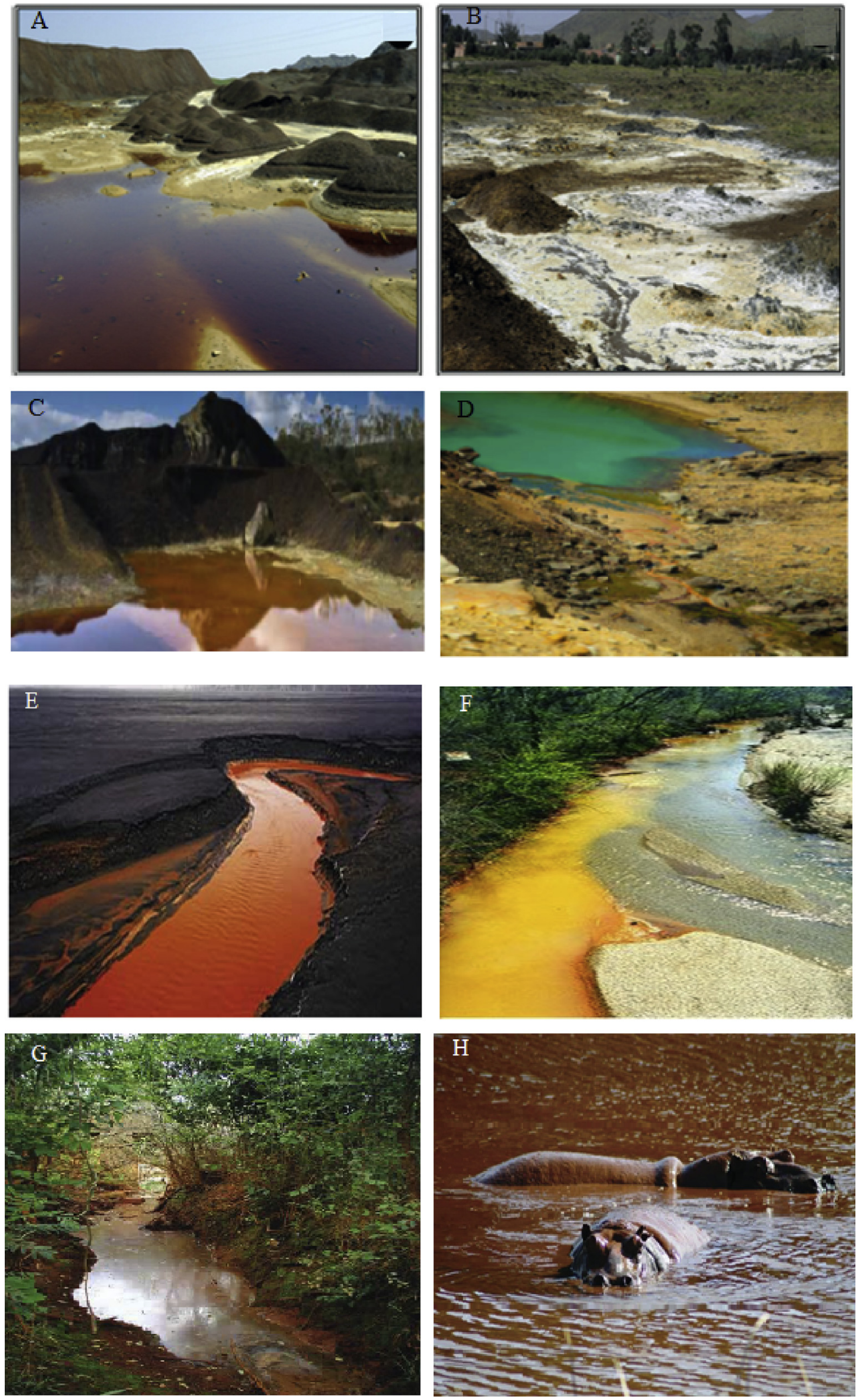

Fig. 1. Illustration of pollution caused by AMD: (A) AMD effluents, (B) blooms of secondary efflorescent minerals in Morocco (A and B are reproduced with permission from Ref. (Khalil et al., 2014), copyright 2014), (C and D) AMD in South Africa (reproduced from Ref. (Clay et al., 2013), copyright 2013 Deloitte & Touche), (E) Cyanide leach, (F) Sulphide deposits in Canada generated by mining (E and F, reproduced from Ref. (Edward Burtnyski, 2007)), (G) Stream affected by AMD in the United States reproduced with permission from Ref. (Schaider and Hauri, 2009) copyright 2009, (H) AMD originated from gold mining in South Africa at Hippo Dam of the Krugersdorp Game Reserve. Picture credited to Stephan du Toit, copyright 2011.

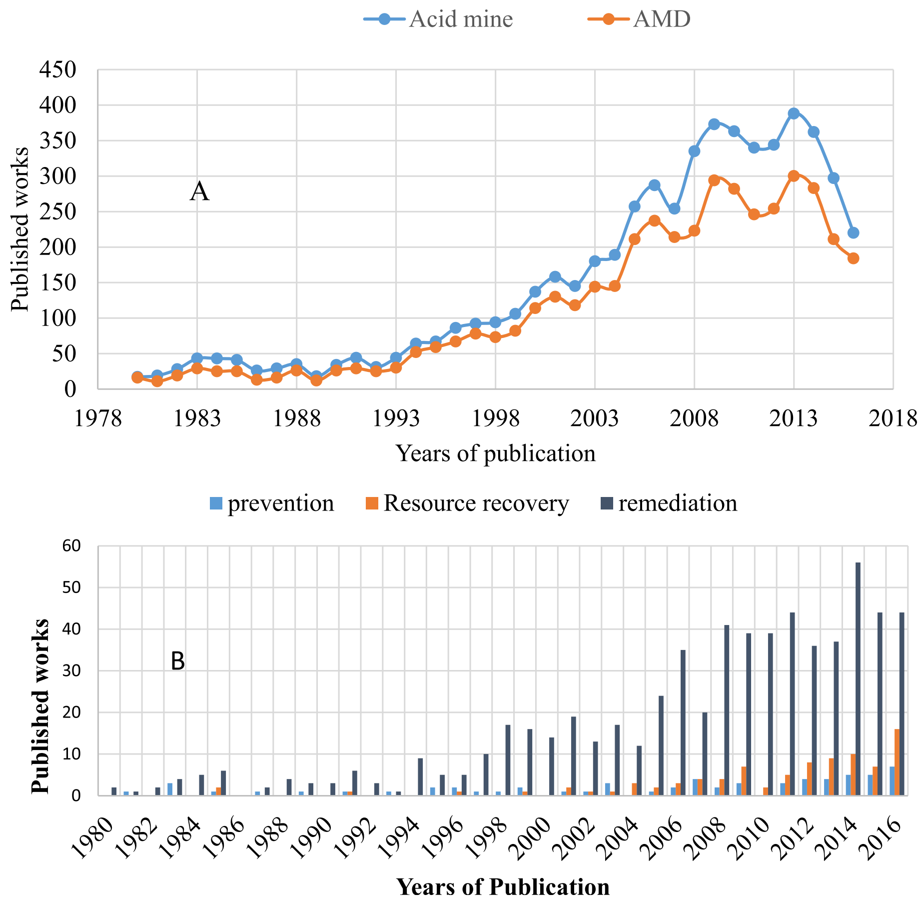

Fig. 2. Number of papers published since 1980 to 30 August 2016 in scopus database: A. with "acid mine" and "acid mine drainage" keyword search. B. Trends of number of papers published in AMD prevention, remediation and resource recovery.

database for "acid mine drainage" and "acid mine" in the keywords from 1980 to 30 August 2016 resulted in a total of 4303 and 5590 articles, respectively. Among these, papers published before 1994 accounts for only 8%, in addition, there was no significance differences in publications per year and also the number of publications per year were less than 50. The extracted numbers of published papers since 1980 to 30 August 2016, excluding papers in press are presented in Fig. 2A. In order to screen the appropriate articles to this review, articles dealing with real AMD treatment issues, removal of contaminants, prevention of AMD generation, AMD remediation or resource recovery were selected based on their title and abstract. To keep the consistency, the screening was done only by corresponding author. In order to observe the trends of different categories of AMD topics considered for this review, relevant terms such as [acid mine drainage and prevention, backfilling, alkaline cover, inhibition, flood wastes or oxygen barrier], [Acid mine drainage and remediation or treatment] and [Acid mine drainage and recovery] were used to search in the title, abstract and keywords. The generated references were thoroughly checked and unrelated published papers were eliminated. These references are presented in supplementary material (S1) while the corresponding data are presented in Fig. 2B. Overall, the analysis of the data has shown high increment in publication from 1994, particularly with rapid increment in AMD remediation relative to resource recovery. The generated data has also shown insignificant differences in the number of papers published in the area of AMD prevention.

In order to address up-to-date AMD treatment options, more emphasis has been given on the latest papers published in between 2010 and 2016. The screened electronic peer reviewed articles in Scopus database were easily accessed from Elsevier (sciencedirect.com), Taylor & Francis (tandfonline.com), IEEE (ieeexplore.ieee.org), Springer (springerlink.com), Wiley (onlinelibrary.wiley.com) or Google scholar. Overall, 200 references are included in the current paper, of which 79% are those published since 2010. Under each subtopic the authors' insights are included from the experience and observation of the real AMD prevention and remediation options. The authors' believe that this review is important in order to design and look for an alternative best technology for prevention of AMD generation and remediation in the future.

3. Prevention of AMD generation

Prevention of AMD generation mainly requires protection of sulphide minerals from air, water and bacteria. This can be done by several management techniques of rock wastes and tailings. Among the available management techniques, mine backfill using a mixture of waste materials such as mixtures of mine tailings and soil, quarried and crushed aggregate, sand materials and Portland cement and other binders are used to minimize acid formation. The backfills are placed into underground-mined voids and serve to improve the underground conditions, provide sufficient alkaline to neutralize acidity (Benzaazoua et al., 2002). Backfills are classified into dry, cemented, hydraulic and paste backfill. Particularly, the use of paste backfill is commonly used in Canada. Backfilling and sealing of the mine waste are intended to prevent air reaching the rocks, thus limiting the oxidation of sulphides (Villain et al., 2013).

Above the ground, several prevention options of AMD generation could be used such as dry cover, water cover, oxygen consuming cover and subaqueous tailing disposal could be used as oxygen barriers to protect AMD generation (Blowes et al., 2014; Demers et al., 2015). Prevention of AMD generation is an important task in order to protect the environment risks in the future (Lu et al., 2014). In Sweden, the two most common methods to prevent sulphide oxidation are dry cover and water cover. In a dry cover, low-sulphide content tailings, clay sub-soils, oxide wastes, alkaline substrates, organic wastes, soils and neutralising materials are commonly used to prevent AMD generation (Lottermoser, 2010; Olds et al., 2012; Smart et al., 2010; Taylor et al., 2006; Zinck et al., 2010). In a study conducted over 500 days in laboratory and 4 years in field, soil-sludge mixture covers has been proven to be an efficient oxygen barriers to prevent AMD generation from waste rock and tailings impoundments (Demers et al., 2016). Recently, the application of alkaline industrial by products for covering rock wastes and tailing are commonly observed. For instance, the small scale trial conducted at the Stockton mine observed that high alkalinity generation from a mixture composed of cement kiln dust to granite in 1:4 ratio by volume. The alkalinity generation rate stabilised at approximately 500 mg $CaCO_3$ eq. L^{-1} after 31 weeks (Olds et al., 2012). The produced alkaline leachate expected to offset the acid production from potentially acid forming wastes and believed to be one of the important methodologies in prevention of AMD generation (Smart et al., 2010; Taylor et al., 2006). In a study where seven waste cover options (waste rock, red mud-waste rock, limestone-waste rock, lime-waste rock, red mud, limestone and lime) were evaluated for 100 days for prevention of AMD generation, application of mixture of red mud-waste rock was found one of the best alternative to restrict AMD generation (Abreu et al., 2012). In fact, one has to pay proper attention during selection of dry covers. Because in case the alkaline cover produces high alkalinity, metal mobilisation from the waste may occur. For example, fly ash used for tailing waste cover produced high alkalinity and resulted in weathering of the tailing waste (Lu et al., 2014). Similarly, a water cover is used as an oxygen diffusion barrier. Because under normal condition, the solubility of oxygen is low and ranges from 8 to 13 mg L^{-1}, and the oxygen diffusion coefficient is approximately 1.12×10^4 times less in water than in air. This phenomenon has been exploited to limit the rate of sulphide mineral oxidation when mine wastes are covered with water (Blowes et al., 2014). This shows that once tailing wastes are covered with water, sulphide oxidation could be almost eliminated (European Commission, 2009).

Organic materials such as wood wastes and other waste forms of organic carbon have been applied to cover surface of mine wastes and act as oxygen scavenger. Consequently, prevent oxygen entry into the mine wastes. However, they have a short life span to be used. Similarly, the subaqueous tailing disposal aids to minimize oxygen contact with reactive materials available in tailings, particularly to prevent the oxidation of sulphide (Blowes et al., 2014). It is commonly practiced in Canada as a protocol for mine reclamation (Jennings et al., 2008). Generally, the literature survey clearly shows a decline of research in the area of new prevention options of AMD generation.

4. AMD treatment options

There are two major categories of AMD treatment options for clean-up of AMD, namely active and passive. The active treatment methods include the application of alkaline chemicals to precipitate metals, and other techniques such as adsorption (Fu and Wang, 2011; Motsi et al., 2011, 2009), ion exchange (Gaikwad et al., 2010), and membrane technology (Alkhudhiri et al., 2012; Ricci et al., 2015). Among the conventional active chemical treatment options, the neutralisation of AMD using alkaline industrial chemicals such as calcium hydroxide ($Ca(OH)_2$) or limestone ($CaCO_3$) is a common method that has been widely applied for the removal of metals as metal hydroxide precipitates and sulphate as gypsum ($CaSO_4.2H_2O$) sludge (Olds et al., 2013; Tolonen et al., 2014). More detailed discussions and examples are presented under Section 4.1 and 4.2. Another good example of active treatment options of AMD is the alkaline barium calcium (ABC) desalination process. This is one of the best recent technology in which both metals and sulphate are reduced below the toxic level. The interesting thing about this technology is that low levels of sludge are disposed of after useful chemicals or metals are recovered, the detailed study of this technology is presented by Mulopo (2015). In general, the ABC desalination process has three major steps, namely the neutralisation part for metal removal, sulphate removal and sludge processing. In fact, for this technology to provide drinking water quality further research is needed.

Since the early 1990s, the passive treatment of AMD is often used to treat AMD (Yadav and Jamal, 2015). It involves biological treatment with constructed wetlands, chemical treatment with limestone drains and sulphate-reducing bioreactors (Tolonen et al., 2014; Zipper and Skousen, 2014). A permeable reactive barrier is also one of the most commonly used passive treatment options, which make use of either chemical or biological process (Neculita et al., 2007). The passive treatment option is more appropriate for application at abandoned mines than the continuous flow of AMD water (Lukacs and Ortolano, 2015; Song and Choi, 2015), as it has benefits of low operational cost and maintenance. The choice of passive treatment to remediate AMD is dependent on the capacity of particular system of producing alkalinity and its efficiency of metal removal. For example, metals such as Zn and Mn are not easily removed at lower pH < 6, therefore, passive treatment in which limestone drains is used, is not the method of choice for the removal of high concentration of Zn and Mn. For the successful removal of high concentration of these metals, application of MgO or combination of limestone and MgO could be more preferred as an alternative method in passive treatment of AMD (Caraballo et al., 2009; Macías et al., 2012; Rötting et al., 2008). In addition, Apatite II™, a biogenic hydroxyapatite, which easily react with acid water releasing phosphate and increasing pH up to 6.5–7, is another alternative for removal of the divalent metals such as Zn(II), Pb(II), Mn(II) Cu(II), Cd(II) and Fe(II) as metal–phosphate precipitates during passive treatment of AMD (Oliva et al., 2012, 2010).

On comparison bases, passive treatment option is viewed as an economic alternative to active treatment because it does not require continuous chemical inputs and also prone to high cost of sludge disposal (Johnson and Hallberg, 2005). In relative terms, the passive methods of treatment are much cheaper than the active ones. The waste produced are denser, less voluminous and more stable compared to the sludge produced during chemical treatment (Neculita et al., 2007). In addition, passive treatment has lower over whole environmental impacts because environmentally relevant materials are usually used during the treatment. It has also been clearly stated that appropriately used passive treatments are very important for ecological restoration due to their sediment generating capacity as the result of bio-mineralisation (Kalin, 2004). However, there are several drawbacks, which limit their use in modern mining industries. For example, passive treatments are usually less effective than active methods and require longer process time for effective remediation of AMD (Iakovleva et al., 2015). The results of detailed life cycle assessment analysis of five various passive and two active AMD treatment approaches conducted at a major coal mine in New Zealand also confirmed similar fact that the active treatment option has greater life cycle assessment impacts compared to the passive treatment approaches (Hengen et al., 2014). This is mainly due to the expensive chemicals used and frequently energy requirement during the treatment time. Generally, the main distinction between the two treatment options is application of continuous chemical inputs for neutralisation of AMD in the active treatment case while no continuous chemical inputs are required in passive treatment but natural occurring chemicals and biological processes are used to cleanse AMD.

The choice of AMD treatment options is dictated by a number of environmental, AMD composition, pH and economic factors. Overall, there is no universal technology for AMD treatment, as the composition of AMD differs depending on its sources, the choice of AMD treatment and the waste produced during AMD treatment also differs. However, some researchers recommended that the dispersed alkaline substrate technology for treating AMD with high metal concentrations. This technology relies on the use of fine-grained alkaline reagents such as limestone sand or MgO powder mixed with pine wood shavings to supply a high porosity to the reactive mixture (Caraballo et al., 2009; Macías et al., 2012). For example, the passive treatment of AMD that employed dispersed alkaline substrate technology at Mina Esperanza (SW Spain) by Caraballo et al. (2011), after 20 months of proper operation in between March 2007 to October 2008, reported complete removal of Al, As, Cd, Cr, Cu, Ti and V, and reduction of Fe concentration from the range of 755–1100 mg L^{-1} to 170–620 mg L^{-1}. Despite its high metal removal capacity, no appropriate treatment options have been suggested to treat the waste generated by this technology.

Recently, an interesting report from Poland on the history of an acidic mine lake where neither the passive nor the active treatment options were involved, but natural processes influenced and neutralised the acidic mine lake (Sienkiewicz and Gąsiorowski, 2016). Pictures of a real acidic mine lake and the present status of the same lake are shown in Fig. 3. The neutralisation of acidic mine lake was accompanied by the disappearance of acidobiontic/acidophilous species and an increase in phytoplankton communities. The study is very helpful for predicting the recovery of other similar lakes situated in that area. In addition, it helps to further our understanding of the natural process of AMD neutralisation so that a similar experimental setup may be designed for remediation of the existing AMD.

Currently, the interest of reducing the cost of treatment motivated the researches to look for low cost AMD treatment options. For example, the applications of naturally available materials, industrial by-products, MNPs and organic substrates have recently emerged as low cost AMD treatment options. The details and relevant literature are presented in the following subsections. Furthermore, the advantages and disadvantages of these treatment options and others are presented in Table 1.

4.1. Alkaline industrial chemicals

Over the past 50 years, several efforts have been made in order to reduce the impact of AMD on the environment through removal of metals and sulphate by neutralisation, particularly using alkaline industrial chemicals. The most popular alkaline industrial chemicals normally used for neutralisation include limestone ($CaCO_3$), slaked lime ($Ca(OH)_2$), soda ash ($NaCO_3$), caustic soda ($NaOH$), ammonium hydroxide (NH_4OH), magnesium hydroxide ($Mg(OH)_2$) and calcium oxide (CaO) (Watten et al., 2005). Most of these chemicals are commercially produced and quite expensive. In addition, when slaked lime or limestone is used for AMD treatment, a large amount of sludge with a high water content is produced (Tolonen et al., 2014), and the sludge has no economic value due to the difficulty of recycling the metals from the waste (Chen et al., 2014a). Therefore, the waste needs to be disposed of in landfills that occupy large areas of land, requiring special designs to avoid the re-dissolution and subsequent migration of toxic trace metals (McDonald et al., 2006). In addition, it requires costly disposal and it can overshadow the operational costs of an AMD treatment plant and make the process unsustainable (Herrera et al., 2007). These chemicals are also ineffective in terms of sulphate removal capacity. On the other hand, treatment of AMD using $Mg(OH)_2$, NH_4OH and NaOH are more preferred for separate precipitation of metals while leaving sulphate in solution (Akinwekomi et al., 2016; Kefeni et al., 2015b). In addition, among the aforementioned alkaline industrial chemicals, NaOH has high capacity of metal removal. This is mainly related to its complete dissociation that enables to achieve high pH value, however, it is quite expensive compared to others.

In the presence of alkaline chemicals such as $CaCO_3(s)$, $MgCO_3(S)$ or NH_4OH, aeration by compressed air or oxygen is also commonly used to facilitate the oxidation of Fe(II) to Fe(III) and its removal in the form of ferric hydroxide precipitate (Akcil and Koldas, 2006; Clyde et al., 2016; Potgieter-Vermaak et al., 2006). The rate of metal removal from AMD is influenced by the chemical composition of the AMD, the aeration rate and the type of alkaline substance used for changing the pH of the solution.

4.2. Alkaline industrial by-products

Alkaline industrial by-products which would otherwise be treated as wastes have recently been utilised for AMD remediation by several researchers. The most common alkaline industrial by products tested for AMD treatments are cement kiln dust, lime kiln

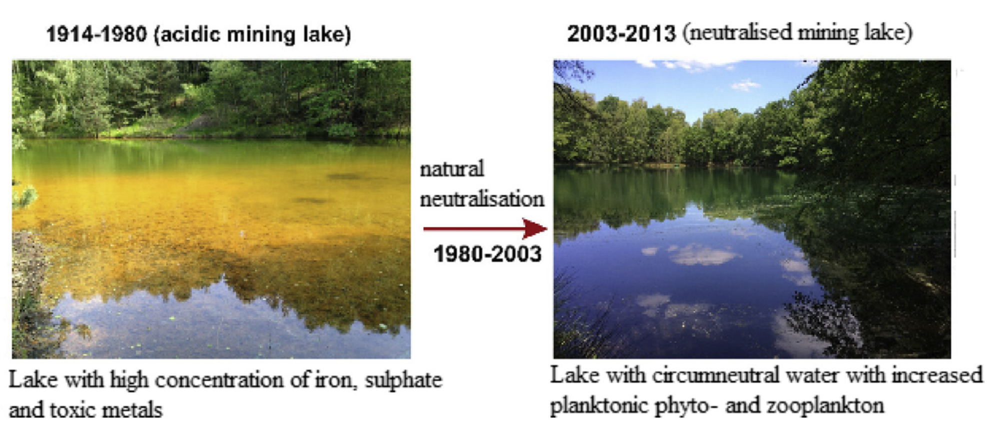

Fig. 3. Natural neutralisation of AMD (reproduced with permission from Ref. (Sienkiewicz and Gąsiorowski, 2016), Copyright 2016).

Table 1
Summary of AMD treatment options and their advantages and disadvantages.

Treatment options	Advantages	Disadvantages	References
Passive: carbonate, lime, marble, fly ash	cost-effective relative to active treatment	Overdose may mobilise contaminants	Pérez-lópez et al. (2009)
Bentonite clay composite	Effective at small scale	Requires shaking	Masindi et al. (2015)
Biochar	Reduce acidity and metal toxicity	Insignificant, difficulty of resource recovery	Jain et al., 2014; Kim et al. (2014)
Bone meal	Cost effective Effective passive treatment of AMD	Need to be heated at 500 °C, cleaned, crushed, boiled and dried; difficult to achieve higher pH	(Payus et al., 2014)
BOS sludge	Cost effective relative to alkaline industrial chemicals	Insignificant	Jafaripour et al. (2015)
Cellulosic waste + SRB	Cost-effective, relative to alkaline chemicals	Slow rate of metal removal	Choudhary and Sheoran (2012)
Chicken manure + SRB	Effective than dairy manure and sawdust	Add high organic loads	Zhang and Wang (2014)
Coal fly ash	efficient and cost-effective relative to lime or limestone	not suitable for recovery of metals from the waste due to elevated concentration of a radioactive element	Madzivire et al. (2014)
Crushed seashell	Cost-effective	Not available as required; needs to be crushed	Masukume et al. (2014)
Dairy manure compost	Generate minimum toxic sludge, cost effective relative to alkaline chemicals	Requires pH adjustment for selective metal removal	Zhang (2011)
Dead Bacillus drentensis sp. In polysulfone polymer	No continuous nutrient supply required, effective at lower pH for toxic metal removal from AMD affected groundwater	Insignificant	(Kim et al., 2014a)
Dunite	Cost effective, removes most of the metals from AMD	Need to be crushed and sieved	(Demetriou et al., 2012)
Electrochemical neutralisation	"No chemicals added to remove chemicals" Form sulphuric acid	Cathode corrosion may occur in the presence of excess Fe(III) in AMD	(Bejan and Bunce, 2015)
Electrodialytic	More effective than the conventional lime treatment	Expensive, fouling	Rojo and Hansen (2010)
Fe_3O_4 nanoparticles	Fast and effective for metal removal, reuse of nanoparticles	insignificant	Kefeni et al. (2015a)
Filamentous green algae	Cost-effective relative to alkaline industrial chemicals; could be reused	pH dependent adsorption	Bakatula et al. (2014)
Food based waste compost and Zeolite	Effective for prevention of AMD generation from tailing	Insignificant	(Hwang et al., 2012)
Lignite	Nontoxic and cost-effective compared to alkaline industrial chemicals	Used for selective metal removal, over dosage may add additional contaminants	Mohan and Chander (2006)
Lime nanoparticles	Fast and effective than conventional lime treatment	expensive	Roy and Bhattacharya (2010)
Manure + SRB	Efficient and cost-effective, compared to active treatment	Insignificant	Choudhary and Sheoran (2012)
Marble stone powder (calcite tailing) + SRB	Effective for passive neutralisation of AMD	The treated AMD water, could be used only for irrigation	(Barros et al., 2009; Martins et al., 2010)
Membrane	Almost complete contaminant removal	Expensive, membrane fouling, brine generation	Buzzi et al., 2013; Martí-Calatayud et al. (2014)
Pig slurry and marble waste	Effective for metal removal (Cd, Pb, and Zn)	Poor Cu removal	Zornoza et al. (2013)
Spent mushroom substrate	Effective for passive treatment of coal mine drainage	Used only for pH greater than 3, and not effective for the removal of Mn and dissolved ferric	(Stark and Williams, 1994)
SRB	Metals removed as sulphide precipitate	Takes longer time for complete removal of contaminants	Castillo et al. (2012)
Steel slag leaches beds	Improved alkalinity	Decrease of alkalinity over time	Goetz and Riefler (2014)
Wood ash mixed with sand	Effective for removal of high concentration of Fe(II) from sulphate reducing passive bioreactors effluent	Requires more than 3 months for almost complete removal of Fe(II) as $Fe(OH)_3$ precipitate	(Genty et al., 2012)
Zeolite	Good metal binding capacity and readily available	Low removal efficiency	Salam et al. (2011)
Zero-valent iron nanoparticles	effective for both organic and inorganic removal	No significant disadvantage	Klimkova et al. (2011)

dust, red mud bauxite, coal fly ash, and blast furnace slag. For example, by-products from quicklime manufacturing were used instead of commercial quicklime or slaked lime which is traditionally used as a neutralisation chemical in AMD treatment (Tolonen et al., 2014). Four by-products (partially burnt lime stored outdoors, partially burnt lime stored in a silo, kiln dust and a mixture of partly burnt lime stored outdoors and dolomite) were studied and the results were compared with commercial quicklime and hydrated lime. According to the authors, all tested by-products removed over 99% of Al, As, Cd, Co, Cu, Fe, Mn, Ni, Zn and approximately 60% of the sulphate from AMD. Of the four tested by-products, partly burnt lime stored outdoors and partly burnt lime stored in a silo showed a higher capacity for metal removal and are expected to be among the most promising by-products to be used as an alternative to quicklime or slaked lime for AMD treatment. Such growing interest in the utilisation of industrial by-products that would otherwise be treated as waste is both ecologically and economically very important due to reduced amounts of waste and waste disposal costs. Similarly, the utilisation of paper and pulp mill by-products as alkaline chemicals for AMD treatment has been studied by Alakangas et al. (2013); cement and lime kiln dust utilisation by others (Doye and Duchesne, 2003; Mackie and Walsh, 2012). The study results obtained have shown that the importance of alkaline by-products in order to circumvent the AMD formation. However, the cost-saving capacity and its sustainability, need to be evaluated through the treatment of active AMD at plant scale.

Residual gas sludge from blast furnace slag, steel slag, mill scales and many other fine solid particles recovered after wet scrubbing of the gas generated from basic oxygen furnace (BOF/BOS) in sludge form are known as waste gas sludge. These types of sludge have been reported as among the most effective materials for AMD treatment (Jafaripour et al., 2015). Nevertheless, the regenerated BOS sludge adsorption capacity was reduced and not as effective as the fresh sludge, perhaps due to the neutralisation of alkalinity by the sulphuric acid used for regeneration. In another study, a similar observation was reported and a decrease in the removal capacity of the contaminant was related to the destruction and distortion of the BOF sludge structure by acid during regeneration time (Zhou and Havnes, 2010; Zhou and Haynes, 2010). For instance, the batch experimental study on the simulated AMD (Simulated Witwatersrand gold basin AMD of pH 2.5 with composition of 5000 mg L^{-1} sulphate and 1000 mg L^{-1} soluble Fe(II)) through the addition of basic oxygen slag increased the pH to 12.1 and removed 99.7% of Fe(II) and 75% of sulphate in a reaction time of 30 min (Name and Sheridan, 2014). Based on the results, the authors suggested that basic oxygen slag is one of the candidates to replace lime for AMD treatment. In fact, the possibility of leaching out of the metals and the means of metal recovery from the sludge or about the fate of the sludge produced after AMD treatment by the addition of BOF slag was not reported by the authors, and requires proper attention in future research.

The application of coal combustion by-products such as boiler slag, bottom ash, fly ash and flue gas desulphurization materials in mine site rehabilitation has recently been reviewed by Park et al. (2014). This review addressed the important of the aforementioned residue for AMD attenuation, however, in order to fully utilise the coal combustion by-products, the author recommended the need for effective guidelines, proper regulations and further research. Others also investigated coal fly ash as one of the potential industrial by-products for removing toxic metal contaminants (Fe, Al, Mn), radioactive metals and sulphate from AMD (Madzivire et al., 2014). In addition, the quality of water obtained after treatment has been reported as being suitable for irrigation purposes. The report has also shown the importance of coal fly ash for removing radioactive materials from mine water and as an aid to obtain water of a standard suitable for drinking. Especially, AMD treatment using fly ash resulted in 90% removal of radioactive metals such as thorium, uranium, radium and lead in the form of their metal oxide precipitate (ThO_2, UO_2, RaO and PbO/PbO2). In particular, coal fly ash acted as the sink of the radioactive materials in AMD. However, its application for treatment of AMD has been used without leaching of the radioactive metals showing that fly ash and contaminated gold mine water can be mixed to produce good-quality water thereby reducing the costs associated with AMD treatment.

In another study, different recycled concrete aggregates and fly ashes in column leach tests were investigated for possible AMD remediation (Jones and Cetin, 2017). The test results have shown concentration reduction of Cr, Cu, Fe, Mn, and Zn in AMD. In addition, sulphate concentrations decreased significantly after being treated by recycled concrete aggregates while sulphate concentrations in the AMD increased when treated by fly ashes. The increase in sulphate concentration in treated AMD is an indication of sulphate leaching from fly ashes, which might be an advantage to be taken into consideration, because at the same time fly ashes could be used as a source of sulphate and remediation of AMD.

The use of cryptocrystalline magnesite tailings which are the by-products of gold mining for removal of elevated toxic chemical species from AMD, has recently been reported (Masindi, 2016). The treated AMD water was within the prescribed legal framework, for water use in the agricultural and industrial sectors in South Africa. The crystalline silicate phase present in iron tailing can generate calcium silicate hydrates, which could be utilised in the production of autoclaved aerated concrete blocks (Ma et al., 2016). These findings clearly show the possibility of reducing environmental impact of tailing through utilisation for different industrial purposes.

Overall, it is well-known that AMD treatment requires addressing the issues concerned with low pH, high dissolved metals and acidity. Therefore, for long-term sustainability and reduction of environmental pollution, addressing these issues must be accompanied by suitable technology that can help to recover valuable resources from AMD. The application of industrial by-products is important and cost-effective; however, the sludge should not be disposed of in the usual way. There must be a proper design for further processing and recovery of the metals from the waste.

4.3. Naturally available resources

There are many naturally available resources which have been tested for AMD treatment and found to be effective such as crushed seashell, lignite, attapulgite, zeolite and bentonite. In addition to their effectiveness, the application of these naturally available resources aid to decrease the cost of treatment. For example, AMD has been treated using crushed seashells as an efficient and cost-effective adsorption medium in batch and column studies (Masukume et al., 2014). Seashell is a hard material in which the main constituent is calcium carbonate and it can be crushed to small pieces for increased capacity to adsorb trace metals. The results of AMD treatment by means of crushed seashell revealed that the seashell-derived adsorbent has great potential as an alternative low-cost material in such treatment.

In another study, Falayi and Ntuli (2014) used inactivated attapulgite [attapulgite is a magnesium aluminium phyllosilicate with formula $(Mg,Al)_2Si_4O_{10}(OH).4(H_2O)$] as an adsorbent for the removal of trace metals from the AMD of a gold mine. The results showed that the removal of metal ion after 4 h reaction time was 100% for Cu(II) and Fe(II), 93% for Co(II), 95% for Ni(II) and 66% for Mn(II) with a 10% (w/v) attapulgite loading. They also reported that the removal of sulphate was low due to the application of inactivated attapulgite, and suggested that the activation of attapulgite may improve sulphate removal and reduce the amount of adsorbent loading. Others also used lignite as an adsorbent to remove metals such as Fe(II), Fe(III), Mn(II), Zn(II) and Ca(II) in single- and multi-component columns setup in down-flow mode from AMD (Mohan and Chander, 2006). The observed results showed that lignite has a high capacity for adsorption of metals even in the presence of interfering metal ions and could be used for the removal of metals from AMD. Furthermore, the authors reported that the recovery of metal ions through the desorption mechanism using 0.1 M HNO_3 (aq.) was found to be about 100% effective. However, nothing was stated about the possible separation techniques of metals from HNO_3 (aq.) and the fate of HNO_3 (aq.). The separation of metals from HNO_3 (aq.) and recovery of HNO_3 (aq.) requires additional cost. . If the disposal option is selected the nitrates are one of the major environmental contaminants and cause drinking water contamination and results in high health impact. Thus, under such experimental condition, the complete processes of both metal and HNO_3(aq) recovery and cost analysis may help in order to evaluate its economic feasibility as well as its sustainable environmental pollution reduction aspect.

Recently, ball-milled bentonite clay was used for the neutralisation of acidity and the removal of toxic metals from AMD at a laboratory scale. The toxic trace metal removal was enhanced through an increase in its surface area to volume ratio. The smaller the amount of bentonite used, the better the water quality as measured by the standard guidelines for discharge (Masindi et al., 2015; Masindi and Gitari, 2016). Despite its importance as good adsorbent of contaminants, bentonite has lower capacity of dissolution compared with commonly used chemicals for active AMD treatment such as limestone and slaked lime. Therefore, there is a need in order to confirm the effectiveness of bentonite for active treatment of AMD in a large scale. Generally, the application of naturally available resources for AMD remediation is encouraging; provided that their application in large-scale in a sustainable condition and valuable resource recovery from the adsorbent is possible under cost effective manner. Otherwise, if one attempts to dispose of the sludge in the usual way, the adsorbed metals and other contaminants can easily leach out, particularly from the adsorbents lacking strong binding capacity. Consequently, the side effects may be worse than the impacts of AMD. In addition to their high metal adsorption capacity, for the selection of naturally available resources for AMD treatment, there is a need to consider either their suitability in terms of cost effectiveness and easy of metal recovery through desorption after treatment or their strong toxic metal immobilisation capacity.

4.4. Plant and animal based wastes

Several plant and animal based wastes have been tested for AMD remediation such as dairy manure compost, cellulosic waste, rice husk, spent coffee grounds and biochar. For example, dairy manure compost has been used as an efficient bio-sorbent for the removal of trace metals from simulated AMD (Zhang, 2011). In this study, the removal of three metals, Pb(II), Cu(II) and Zn(II) from simulated AMD was tested. The result showed that the adsorption affinity order of the three metals was Pb > Cu > Zn. The adsorption efficiency of these metals was pH-dependent, and the maximum adsorption was found to occur at around 3.5, 4.5 and 5.5 for Pb, Cu and Zn, respectively. The regeneration experiments have also shown that the adsorbent could be regenerated and reused at least three times without a significant decrease in adsorption capacity. Similarly, the application of carbon-rich by-products synthesised from the plant and animal-based biomass by thermal decomposition under a limited supply of air/oxygen, which is commonly known as biochar has also been and used for remediation of AMD (Jain et al., 2014; Kim et al., 2014b). For example, the application of spent coffee grounds and charred spent coffee grounds (biochar) has been reported as one of effective methods for the remediation of AMD (Kim et al., 2014b). However, relatively speaking, uncharred spent coffee grounds are not suitable for AMD treatment due to an excessive amount of dissolved organic carbon released that induces biological toxicity. These experimental results clearly demonstrated the importance of plant and animal based wastes for the effective remediation of AMD. For example, it has been reported that biochar is important for metal immobilisation in both water and soil. The stabilisation of the metals is possible through formation of either metal complexes (bond formation between d-orbitals of the metals with delocalised π-electrons of carbon (C=C)) or their corresponding carbonate, hydroxide or phosphate precipitate. Consequently, toxic metals and other contaminants adsorbed onto biochar from AMD do not easily leached out. This implies, biochar is an environmental relevant materials and its application for AMD remediation appears more promising. In addition, amendment of soil with biochar improves the soil quality and promoting bioatic activity and soil fertility (Uchimiya et al., 2010; Yuan et al., 2011).

4.5. Limestone based passive treatments and others

Passive treatments mainly based on limestone are well studied and employed for AMD treatment. Some of commonly used include, anoxic limestone drains (ALD), oxic limestone drains (OLD), open limestone channels (OLC), and limestone diversion wells. These treatment options are generally limited by slower rates of neutralisation and metal contaminant removal relative to active treatment (Yadav and Jamal, 2015). The ALD comprised of trenches filled with limestone, through which the AMD percolates. The ALD often have a pre-treatment stage to remove oxygen from the

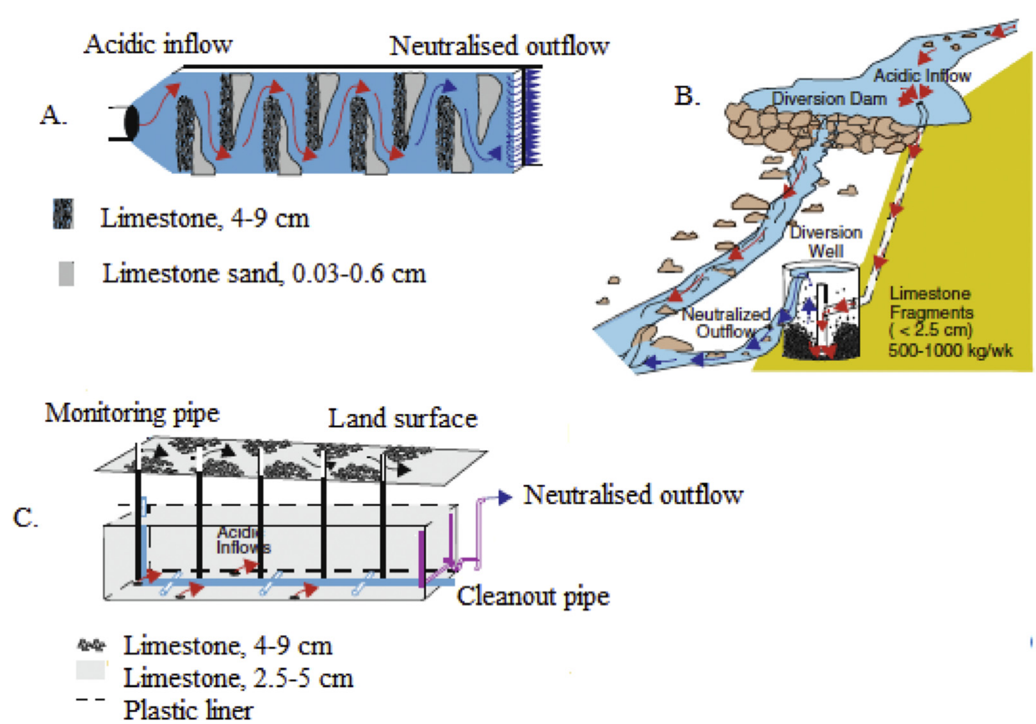

Fig. 4. Schematic illustrations of passive treatment systems: A. open limestone channel B. Limestone diversion well, and C. Anoxic or Oxic limestone drain type (Reproduced by permission from reference (Cravotta III, 2010), Copyright 2010).

Table 2
Selected exiting biological PRB installed in worldwide.

Country and year of installation	Composition of PRB	Dimensions (m)	t_R(d)	pH	Concentration of contaminant in the influent (mg L^{-1})		pH	% removal		Ref.
					Metal	SO_4^{2-}		Metal	SO_4^{2-}	
Spain, Aznalcóllar (2000)	Vegetal compost (35–40%) Sewage sludge (0–5%) Limestone (60%) Iron (0–5%)	W = 110 T = 1.4 D = 3.0–7.5	1–2	4	Al(15) Cu(1) Zn(20)	1000	5	50–98	0–43	Gibert et al. (2011)
UK, Shilbottle (2003)	Horse manure & straw (25%) Green compost (25%) Limestone (50%)	W = 180 T = 2 D = 3	0.4–1.7	<4	Al(16–862) Fe(10–800) Mn (5.4–227)	8700	–	10–87	40	Caraballo et al. (2010)
USA, South Carolina (2002)	Zero-valent Fe (20%), pea gravel (45%), limestone (5%)	W = 7.9 T = 1.8 D = 4.1	–	3.2–4.2	Al (69–2710) As(0.26–206) Cd(0.003–1.4) Fe(83–10500) Ni(0.05–2.12) Pb(0.03–4.08) Zn(2.6–1060)	1800–49500	6.9–9.3	>98	83–99	Ludwig et al. (2009)
USA, Florida	Limestone (7–10 mm particle diameter)	W = 6 T = 0.9 D = 4.6	–	6.46	Fe (30) Mn(1.62)	– –	8.7	91[a], 64[b] NR	–	Wang et al. (2016)
	Crushed concrete (70–150 mm particle diameter)	W = 6 T = 0.9 D = 4.6	–	6.46	Fe(30) Mn(1.62)	–	8.7	95[a], 61[b] NR	–	Wang et al. (2016)

Dimensions: W = width, T = thickness and D = depth.
NR = not reported.
[a] After one year functioning.
[b] After three year functioning.

influent AMD, in most cases, the initial section of the trench could be filled with organic materials for prior removal of oxygen. Furthermore, the trenches are designed to exclude oxygen entrance, thereby inhabiting metal hydroxide precipitation and acidic influent of AMD is neutralised by dissolution of limestone (Bejan and Bunce, 2015; Hedin et al., 1994; Santomartino and Webb, 2007). The effluent is directed into settling pond to allow pH adjustment and metal precipitation before discharge to water system. The main drawbacks of ALDs after the long term use include clogging with particulates and metal hydroxide precipitates (Bejan and Bunce, 2015). In addition, ALD techniques are less efficient in order to remove high metal concentrations, thus it is not a method of choice for treating AMD with high metal concentration, unless combined with other passive treatment options such as SRB. Alternatively, OLD or OLC could be used for neutralisation of AMD influents that does not meet criteria for an ALD (Cravotta III, 2008).

OLD is a system physical similar to ALD installed to treat oxic mine water. In OLD system, Fe^{3+} and Al^{3+} are removed through precipitation within OLD and large precipitation of metal hydroxides retained in suspension, so that they exist the OLD without contributing to clogging (Younger et al., 2002). Schematic illustration of ALD, OLD, OLC and limestone diversion wells are presented in Fig. 4.

Other treatment options such as constructed wetlands and permeable reactive barriers (PRB) are also among one of the passive treatment technologies commonly used. The constructed wetlands utilise organic substrates to support bacterially mediated sulphate reduction and dissolved metal removal from AMD (Ayora et al., 2016). However, due to its high space demand, more compact alternative design which is mainly based on the neutralisation of acidity using limestone dissolution is more preferred. The readers also can found more information with respect to water quality and design requirements for different passive treatment options in Ziemkiewicz et al. (2003).

The permeable reactive barrier (PRB) was emerged in the last two decades as a promising alternative compared to the conventional pump-and-treat approaches, particularly for groundwater treatment. It consist of reactive materials buried in a narrow trench such that contaminated water could be treated as it follows through buried reactive (Caraballo et al., 2010; Gibert et al., 2011; Henderson and Demond, 2007; Ludwig et al., 2009; Pagnanelli et al., 2009; Wang et al., 2016). The selection of reactive materials dependent on the type of AMD to be treated and contaminants to be eliminated. Currently, there are several PRB installed worldwide for the removal of different type contaminants from the ground water. In most cases, the composition of reactive materials include zero-valent iron (ZVI), and less often modified zeolites, limestone, activated carbon, organic substrates with SRB etc. In addition to the type of filling materials, flow rate and residence time within the PRB mainly influences the contaminant removal capacity. For example, it has been observed that higher flow rate and low residence time resulted in lower rate of contaminant removal (Gibert et al., 2011). Table 2 presents the detailed information with respect to composition of reactive materials used as filler in PRB, retention time and other important parameters. An example of the integration of combination of each approaches for AMD remediation is presented in Fig. 5.

4.6. Microorganisms

Biological passive treatment using SRB has recently emerged as one of the most promising alternative technologies for AMD remediation. The feasibility of passive treatment options is evaluated on the basis of the capacity of a particular system to produce the required alkalinity and the removal capacity of dissolved metals

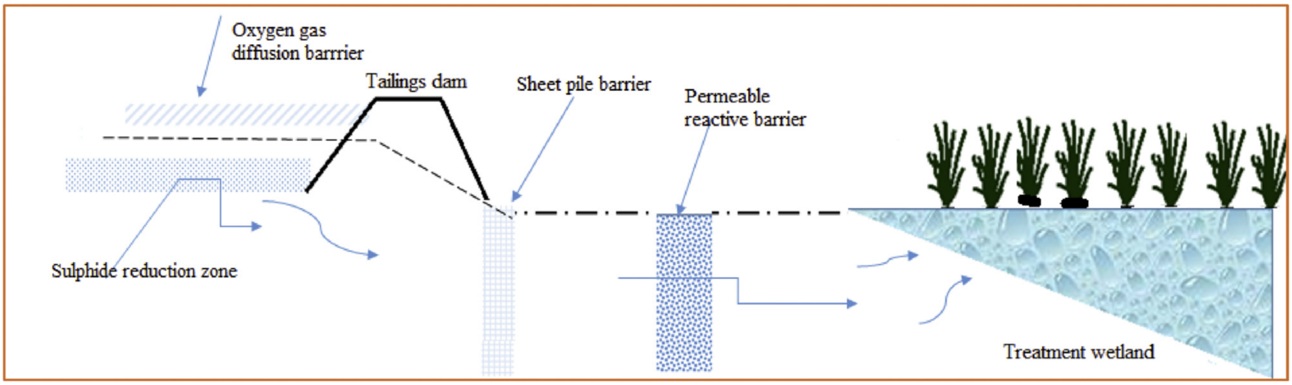

Fig. 5. Schematic illustration of mine waste impoundment with a combined treatment approach: A cover to prevent and water ingress, mixing to stabilize waste, PRB in aquifer to treat subsurface drainage and wetland for surface treatment of drainage, adapted from Ref. Blowes et al. (2014).

(Kalin et al., 2006). A number of experimental tests approved SRB as an effective AMD treatment option and have been reported by several researchers (Bai et al., 2013; Castillo et al., 2012; Clyde et al., 2016; Deng et al., 2016; Hao et al., 2014; Johnson, 2014; Kieu et al., 2011; Sahinkaya et al., 2015; Zhang and Wang, 2016, 2014). The utilisation of SRB for the remediation of industrial sulphate containing sewage and sulphur contaminated groundwater and their environmental preferences with regard to bioreactors on different operational parameters has recently been reviewed by Hao et al. (2014). The review summarised the advantage of SRB over the conventional method of wastewater treatment in sulphur bioconversion systems for simultaneous removal of carbon and toxic trace metals existing in industrial wastewater. Furthermore, the utilisation of SRB results in minimal biological sludge production and greenhouse gas emission relative to conventional carbon cycle based treatment technologies.

There are a number of microorganisms which survive and flourish in extremely harsh environments of AMD that could be used for AMD remediation. However, before the SRB are used for AMD treatment, their adaptability or tolerance to the specific AMD should be studied. Because, according to the recent study that applied a bar-coded 16S rRNA pyrosequencing technology analysis results of 59 AMD samples collected from 14 physically and geochemically diverse mining sites across Southeast China revealed that contemporary environmental variation as the major factor explaining community differences and their survival rate in these harsh environments rather than geographical distance (Kuang et al., 2013).

Remediation of AMD by means of SRB is commonly based on the use of mixtures of salts ($CaCl_2$, K_2HPO_4, NH_4Cl, $MgSO_4.7H_2O$, $CaCO_3$, $(NH_4)_2Fe(SO_4)_2$) and locally available organic substrates such as manures, sawdust, sugarcane waste, yeast extract, wood chips, spent mushroom compost and other carbon sources for the bacterial metabolism. It should be also noted that higher sulphate reduction is achieved with reactive mixtures containing more than one organic component sources (Gibert et al., 2011, 2004; Muhammad et al., 2015; Neculita et al., 2007; Waybrant et al., 1998; Zagury and Neculita, 2007). In addition, increasing or decreasing the substrate mixtures have significant impact on the efficiency of SRB. Therefore, in order to achieve optimum sulphate reduction and metal removal, optimization of the best proportion mixture of substrate is required (Muhammad et al., 2015; Neculita et al., 2007). The substrate mixtures, besides their being used as nutrients for microorganisms, they are also important in the reduction of the adverse effects of toxic metals on SRB through the removal of the metals via adsorption and the buffering of the solution's acidity (Zhang and Wang, 2014). The organic substrate, which is composed of herbaceous and woody materials are potentially important as carbon sources to promote SRB activities for biological remediation of AMD while the wood chips aid to increase the permeability of the media in the bioreactor. SRB greatly contributed to enhancing the removal of sulphate, iron and trace metals (i.e., Cu, Cd, Zn, Ni) through microbially mediated reaction. First sulphate is converted into hydrogen sulphide (H_2S (g)), while organic matter ("CH_2O") is converted into hydrogen carbonate. The H_2S (g) reacts with some metal cations and form insoluble metal sulphide precipitates, usually metals with $+2$ oxidation states such as Zn, Fe, Cu, Ni, and Cd (Lewis, 2010; Schaider and Hauri, 2009), and the metals are removed through sulphide precipitation. SRB is an integrated biological approach to the treatment of metal-rich mine water and at the same time, selective recovery of metals has recently been reported (Hedrich and Johnson, 2014). In this study, the two major metals available in AMD at the Maurliden mine referred to earlier (Zn ~460 mg L^{-1} and iron ~400 mg L^{-1}) were removed by controlled bio-mineralisation of Zn as ZnS(s) in sulfidogenic bioreactors and iron as schwertmannite by microbial iron oxidation and removed as ferric hydroxide precipitate. The organic substrate based reactors were found to be more efficient than only limestone reactors (Lefticariu et al., 2015). The results have shown the importance of anaerobic organic substrate bioreactors as one of the promising technologies for the remediation of AMD generated by coal mining. Further, the authors suggested improving the organic substrate input and the pre-treatment of AMD aids in order to reduce the high concentration of Fe and Al content and enhance the efficiency of contaminant removal. Some of the advantages of using SRB over metal hydroxide precipitate is that metal sulphides have a lower solubility over a broad pH range (Choudhary and Sheoran, 2012). Generally, the treatment of AMD by means of SRB is among the more cost-effective treatment options for abandoned mines. Furthermore, the application of SRB completely removes Zn and reduces the mobility of all metals available in AMD (Castillo et al., 2012), which are commonly left in the solution after passive treatment of AMD by means of limestone. However, as it is a passive treatment, more time is required for the complete removal of the pollutants in the form of metal precipitates and the recovery and reuse of valuable chemicals are difficult except in bioreactors. This means, the fate of the metal sulphide precipitates is also not well known.

Recently, toxic metal-resistant immobilized SRB granules were used for the treatment of a synthetic AMD containing high concentration of metal ions (iron 463 mg L^{-1}, Mn 79 mg L^{-1}, Cu 76 mg L^{-1}, Cd 58 mg L^{-1} and Zn 118 mg L^{-1}) and sulphate

3706 mg L^{-1} using an up-flow anaerobic packed-bed bioreactor (Zhang and Wang, 2016). This work demonstrated the efficient removal of metals from AMD with a removal efficiency of 99.9% except for Mn (42.1–99.3%) and sulphate (80.2%). Similarly, a system has been developed in which coal mine-derived AMD flows as a sheet 0.5 to 1 cm-deep over the terrestrial surface. This developed system enhanced ease of aeration of AMD and the activities of Fe(II) oxidizing bacteria and gave rise to excessive Fe(III) hydroxide-rich deposits which are referred as an iron mounds. These iron mounds were developed with no human intervention, indicating that microbiological activities associated with iron mounds may be exploited as an inexpensive and sustainable approach to the removal of Fe(II) from AMD. Soil-associated with microbial communities which enhance the oxidation of Fe(II) to Fe(III) was used on coal-mine-derived AMD. The result showed the continuous exposure of soil to AMD-induced progressively greater rates of Fe(II) bio-oxidation (Brantner and Senko, 2014). Generally, the experimental result indicates that the possible restoration and remediation of soil affected by AMD using similar application.

The remediation of AMD by SRB is a promising alternative to the conventional method of treatment (Bai et al., 2013). In order to enhance the efficiency of SRB the addition of small amounts of Fe(0) is important. For example, the addition of Fe(0) improved the activity of SRB, and more than 61% of sulphate, 99% of Cu(II) and 86% of Fe(II) were removed effectively during the AMD treatment (Bai et al., 2013).

One of the drawbacks of the application of SRB is that the low pH of AMD seriously affects the growth of microorganisms and impairs the bioreactor performance (Hao et al., 2014; Sánchez-Andrea et al., 2014). The problem can be reduced through addition of $CaCO_3$ and the organic substrate. Especially, $CaCO_3$ aids to increase the pH of AMD while both chemicals improve the proper performance of SRB by removing certain toxic trace metals and at the same time can be used as nutrients for microorganisms. Another drawback of the application of SRB is the metal sulphide precipitates are no longer recovered and used, at the end, after a long time they may cause environmental pollution. However, recent reports in some of the literature stated that the possibility of recover and reuse of metal sulphides, which are precipitated from aqueous solutions in bioreactors (Costa et al., 2013, 2012; Xingyu et al., 2013). The recovery is possible through careful control of the reaction conditions and obtains the sulphide precipitate at nanoparticle size. The recovered sulphide can be used for further precipitation of other metal sulphides in bioreactors (Costa et al., 2013, 2012). The recovery and reuse of metal sulphides for precipitating another metal sulphide have an economic advantage in that the application of SRB eliminates the use of expensive or toxic chemicals or any other sophisticated apparatus (Vitor et al., 2015). In general, application of SRB for AMD is important if the aforementioned drawbacks are improved through advanced research.

4.7. Magnetic nanoparticles as seed

Application of MNPs for AMD treatment as seeds is one of the best options and has many advantages over chemical precipitation and adsorption methods. For example, the process is cost-effective due to several times reuse of the MNPs by recovering them using an external magnetic field without filtration or centrifugation (Foroughi et al., 2015). The metal ions from AMD could be removed either through adsorption on the surface of MNPs or incorporated into the MNPs. The adsorbed metals on the surface of MNPs could be easily recovered via desorption process using appropriate aqueous solutions of either sodium hydroxide or strong acids (Gómez-pastora et al., 2014; Lata et al., 2015). Unlike the lime or limestone methods which generates a large volume of sludge, the sludge produced from this process is compact and less toxic (Lou and Chang, 2007). In addition, MNPs could also be applied directly for simultaneous removal of various cations and anions that contain toxic metals such as $Cr_2O_7^{2-}$, CrO_4^{2-}, and MnO_4^- through adsorption route.

The AMD with high iron content is different in that the iron can be easily recovered in the form of magnetite and industrially significant for paint production and various other purposes. Generally, application of MNPs for AMD treatment reduces sludge waste and produces valuable mineral resources (de Almeida-Silva et al., 2012). For example, the iron recovered in the form of magnetite at pH 3.6 allowed the recovery of 96% of the iron in acidic coal mine drainage, which would reduce the overall volume of sludge by 70%. The magnetite seeds are also valuable for separate removal of iron under controlled conditions (Lahav et al., 2003). In general, the application of MNPs as a seed in order to recover metals available in the AMD is economically important and reduces the cost of waste disposal. However, application of MNPs for AMD treatment is not well explored and less emphasis is given in the investigation of practical applicability of MNPs in a continuous operation that resembles industrial application. In addition, after several recycle and reuse, there is a point at which MNPs are exhausted and might be disposed. In fact, there is no proper procedure of disposal and effective management technology developed for disposal controlling for MNPs. Thus, their toxicity is a concern, which requires detailed investigation before their application in industrial scale at a wide range.

4.8. Membrane technology

Application of membrane process is the most promising technologies to reduce effluent discharge, and minimize water requirement through wastewater reclamation (Liu et al., 2011). AMD treatment using membrane technology is not common due to the relatively high cost of the membrane and high membrane fouling due to the susceptibility of membrane systems to low pH of AMD; however, application of nanofiltration (NF) and reverse osmosis (RO) processes for AMD treatment recently attracted more researchers due to their high capacity of salt and metal retention. In recent study, high membrane fouling and risk of performance failure was reduced by pre-treatment of AMD using sand filtration equipped by rice husk-ash and coal fly-ash adsorbent columns followed by ultrafiltration (UF) and RO (Nasir et al., 2016). The pre-treatment increased the final pH permeate of RO in the range of 6.0–6.8, and also removed 98.00, 94.11, and 95.8% sulphate, iron, and Mn, respectively. Furthermore, the small-scale AMD plant was capable of producing high quality of permeate and met the environmental standard.

Recently, the applicability of RO and NF for gold AMD treatment was evaluated; in comparison, the NF had higher permeate flux and satisfactory solutes retention efficiency than RO (Aguiar et al., 2016). Furthermore, this study achieved the maximum water recovery rate of 60% with the NF270 membrane from gold AMD for a single step treatment at the optimised conditions, after which the permeate flux decrease was sharper, indicating higher membrane fouling. Other researchers also suggested the NF as one of the preferable membrane separation process for effluent treatment due to its higher fluxes at lower applied pressure (Al-Zoubi et al., 2010; Mullett et al., 2014). Similarly, in a laboratory scale study, in north of Spain, Hg mining has been identified as a potential source of trace elements such as As, Sb, Pb, and Hg. Some of these contaminants are dissolved in AMD and polluted, particularly Los Rueldos mine site. This AMD has been treated using FILMTEC™ NF-2540 membrane by means of NF to remove some of its pollutants. The result has shown the effectiveness of membrane used in removing the pollutants even at low pH and moderate pressure (Sierra et al., 2013).

Another good example of membrane technology for AMD treatment is mining activity at the Zijinshan plant in China which was being treated with a capacity of 3300 to 3600 m^3 $(days)^{-1}$ (Renman et al., 2011). In fact, both RO and NF are one of the more recent technologies applied to AMD. However, as with many other treatment options, in addition to its high cost, this process has one major drawback – it generates brines rich in sodium sulphate. The treatment and disposal of brine are very expensive because the brine produced often has total dissolved solids of a high concentration, ranging from 20,000 to 35,000 mg L^{-1} (Mulopo, 2015). Therefore, along with improving membrane separation technology for AMD treatment, means of treating or possible reuse of the brine needs to be developed.

In another study, anion- and cation-exchange membranes were evaluated for AMD treatment (by means of an HDX 200 anion-exchange membrane and an HDX 100 cation-exchange membrane). The results have shown that electrodialysis is suitable for recovering water from AMD, with a contaminant removal capacity greater than 97%. However, the precipitation of the iron at the surface of the cation exchange membrane created a problem by blocking the membrane through the membrane fouling and reduced the process efficiency (Buzzi et al., 2013). The use of the membrane for AMD treatment has also been reported for obtaining pure sulphuric acid (Martí-Calatayud et al., 2014). In general, despite the good separation of pure water from contaminants, the question of cost and membrane fouling may reduce the application of membrane technology for the treatment of AMD. In fact, the pretreatment of AMD using microfiltration (MF) or UF prior to NF and RO treatments can remove fouling components and reduce the severe damage to the membranes. For example, in one of the recent study, in order to reduce membrane fouling, 2.0 μm ceramic compact rotating disc MF membrane prepared from α-Al_2O_3 was used to treat AMD, which composed of high concentration of iron and other toxic metals (Meschke et al., 2015). The results of this study have shown 99.9% iron retention for the AMD waters pretreated with lime milk. The rotary motion ceramic disc membrane reduced the layer formation on the membrane surface, minimized membrane fouling and improved flux. In general, application of membrane for AMD treatment seems a promising technology, however, for full application of membrane technology researchers should look into appropriate fouling resistance membrane and also developing technology that treats the final waste produced as brine is necessary.

5. Possible resources to be recovered from AMD

Although the metals and sulphate which exist in AMD are considered environmental pollutants, they may also be valuable resources (Buzzi et al., 2013; Chen et al., 2014a; Kefeni et al., 2015a). In the recent AMD treatment, most of the AMD constituents are considered as valuable resources. Consequently, more emphasis has been placed on the recovery and reuse of the valuable chemicals. For example, the most common valuable resources obtained at laboratory scale are ferric hydroxide, ferrite, barium sulphate, gypsum, rare earth metals, sulphur and sulphuric acid. Recovery of these resourceful chemicals from AMD is one of the alternative way of sustainable mining and reduction of environmental pollution. These recovered resources from AMD could be saleable and at least help to cover the cost of treatment. For example, the BioteQ plants which is operating in China, Canada and in the U.S. mining sites has been recognized to treat AMD, leach solutions, industrial wastewater, water in mineral processing and metallurgical operations, and contaminated ground water and convert into a useful resource (Gleason, 2009). The valuable metals recovered from AMD include Cu, Zn, Ni and Co, by selling these metals new income could be generated. In addition, toxic metals such as As, Sb, Pb, Cd and Mn are also removed from the water by the same plant. In another study, the pilot test conducted for six months on metal resource recovery from real AMD by fractional precipitation process in continues flow plant confirmed that the possible recovery of major metal ions such as Fe, Cu, Zn and Mn available in AMD (Yan et al., 2015). The recovery rates of Fe, Cu, Zn, and Mn were 82%, 79%, 83%, and 83%, respectively. In addition, the cost-analysis based calculations also demonstrated that the recycled metals were able to pay for the cost of metal reagents used.

Recently, rare earth elements and yttrium, which are very important raw materials in modern technological development such as permanent magnets, light emitting diodes, alloy in rechargeable batteries and liquid crystals are found in higher concentrations in AMD than in naturally occurring water bodies (Ayora et al., 2016). Thus, AMD is considered as one of an important new source of these rare earth metals and yttrium.

The possible recovery options of sulphuric acid from AMD and its reuse have recently been reviewed by Nleya et al. (2015), which showed the possible recovery of sulphuric acid with good quality that could meet the demand of various sulphuric acid consumers. Particularly, among several methods for sulphuric acid recovery options from AMD, the authors recommended acid retardation and crystallization as the most promising technology; based on cost-effectiveness and technical feasibility of sulphuric acid recovery from AMD.

In order to reduce the AMD sludge discharge and environmental pollution, special efforts have been made in recovering iron oxides from AMD for preparation of catalyst, pigment and ferrite nanoparticles (Andersen et al., 2012; Cheng et al., 2011; de Almeida-Silva et al., 2012; Flores et al., 2012; Sun et al., 2015a, 2015b). Recently, fuel cell technology is becoming one of an important area of study for recovery of metals from AMD. For example, by applying the fuel cell technology the iron in AMD is selectively recovered, oxidised in the presence of air and then calcinated to form Fe_3O_4/carbon composite which is an effective catalyst in the electron-Fenton reaction (Sun et al., 2015a). While the generation of electricity and the simultaneous extraction of goethite (α-FeOOH) nanoparticles using new fuel cell technology during the treatment of AMD have also been demonstrated by Cheng et al. (2011). The size of goethite nanoparticles obtained during this process ranged from 120 to 700 nm, which is appropriate for pigments in paints and other applications. The preparation of brown inorganic pigments from the residue of AMD treatment after being mixed with pure metal oxide has been reported by Marcello et al. (2008). When only the residue pigment obtained from AMD was used, a faded brown colour was obtained, which was inadequate quality to be used for ceramic glazes; however, mixing with pure metal oxide improved the quality. Michalkova et al. (2013) reported the recovery of ferric and aluminium hydroxides above 90% and also several other metals from low- to high-percentage recovery. Such initiatives of developing new technology for the recovery and reuse of chemicals may offer economic benefits while also providing solutions to environmental problems.

Recent reports on field assessment for As immobilisation in soil amended with iron rich AMD sludge showed that soil amended with AMDs could prompt As immobilisation in soil and prevent As transfer from soil to crops (Ko et al., 2015). The mobility of As was prevented by the AMD added into the soil, and the concentration of As in the field soil and grain was found to be similar. AMD has also been used for biomass recovery of two morphologically different microalgal species (Scenedesmus Obliquus and Chlorella Vulgaris). Thus, AMD could be an effective option for the economic harvesting of microalgal biomass coagulation/flocculation within 20 min at

Table 3
Summary of recovery methods and chemicals recovered from AMD.

Method	Solution content	Recovered chemicals	Amount recovered (%)	Application	Ref.
Diffusion dialysis	H_2SO_4 (61.7), Fe (11.2) and V (4.60) g/L	H_2SO_4	84	Industrial	Wei et al. (2010)
Neutralisation using NaOH(aq.)[a]	Cations: Fe, Al, Cu, Zn, Ni, Mn, Ca, Mg, Na, and anions: Cl^- and SO_4^{2-}	Fe Al Cu Zn	97.2 74.9 66.9 89.7	Industrial	Park et al. (2015b)
Electrochemical		Fe Al Cu Zn	99.4 89.9 54.3 70.3	Industrial	Park et al. (2015a)
Co-precipitation	Cations: Fe, Al, Co, Cu, Zn, Ni, Mn, Ca, Mg, Na	Fe recovered as Fe_3O_4 and α-Fe_2O_3	84 and 15.6	adsorbent	Kefeni et al. (2015a)
Co-precipitation Adsorption, Rice Husk	Cations: Fe, Cu, Zn, Na, K, Ca Anions: SO_4^{2-}	Fe(III) Fe(II) Cu(II) Zn(II)	>95[b] >95 >98 >98	Industrial	Chockalingam and Subramanian (2006)
Roasting Pb-Zn sulphide tailings	S, Fe, Pb, Zn, Cu, Cr, Cd, Ag and Ga	Sulphur as sulphuric acid	94.7	Chemical industries	Lei et al. (2015)
Scheil-titration	Fe, Al, Co, Cu, Zn, Ni, Mn, Ca, Mg, Na	Fe (pH = 2.6–3.6)[c] Al(pH = 3.5–5) Cu(pH = 4.9–8) Zn(pH = 4.9–8) Co(pH = 4.9–8) Mn(pH = 7.7–10) Mg(pH = 10–11)	97 94 94 96 72 62 68	Could be used for several purposes [d]	Salminen et al. (2015)
Oxidation and precipitation for Fe, only Precipitation for Cu, Zn and Mn	Al, Ca, Mg, Mn, Cr, Fe, Co, Ni, Cu, Zn, Cd, and Pb	Fe(at pH = 3.8) using H_2O_2 Cu (using Na_2S)[e] Zn (using Na_2S) Mn– using CaO	82 79 83 83	Could be used in industries for different purpose	Yan et al. (2015)

[a] The pH adjusted to 3.5, 5.0, 6.0 and 8.0 for the removal of Fe, Al, Cu and Zn, respectively.
[b] All metals were recovered by using 1 M HCl (aq.).
[c] All recovered as metal hydroxide, except Mg which recovered as mixtures dolomite and metal hydroxide and Mn as $MnCO_3$(s).
[d] Fe-for water purification application, adsorption and agent, Al-as reagent in ettringite, $Ca_6Al_2(SO_4)_3$ precipitation for sulphate removal from mine waters, metal such as Cu, Co, Zn, Mg, and Mn-could be further processed and used in chemical industries.
[e] Cu is precipitated prior to Zn and Mn based on the solubility product of metal sulphides.

initial suspension pH levels of 7 and 9. The application of AMD for the recovery of microalgae biomass is a cost-effective method; which might further allow the reuse of a flocculated medium for algal cultivation, thereby contributing to the economic production of biofuel from microalgal biomass (Salama et al., 2015). The treatment of AMD at a controlled pH range of 7–8.5 at a temperature of 60 °C through the co-precipitation method in the presence of 25% NH_4OH (aq.) produced mixtures of MNPs with a major composition of Fe_3O_4, γ-Fe_2O_3, Mn_3O_4, MnO_2 and ZnO (Kefeni et al., 2015a). These MNPs could be used as a seed to treat fresh AMD in order to facilitate the rate of metal contaminant removal; also, they are important as additives in pigments. Summary of methods used and valuable recovered chemicals from AMD are presented in Table 3.

6. Future perspective and research needs

Generation of AMD could be reduced by following proper procedure. For example, soil samples of the area anticipated for mining operations should be pre-analysed in order to determine all chemical composition of the minerals. Based on the composition determined, the acidity generation potential of the samples must be tested and decision of mining should be made based the analysis results. On the other hand, avoiding mining of sulphide ores may be difficult because they are most often associated with the mineral resource of interest. In this case, possible prevention of AMD generation should be employed. Overall, possible waste management strategy should be in place at all stages, such as pre-extraction, appropriate extraction procedures, sludge disposal, recovery of possible resources, etc. and even as far as appropriate closure procedures and rehabilitation.

One of the problems that aggravate the impact of AMD is seeping down of toxic chemicals from mine tailings, which are commonly considered as one of AMD source (Hansen, 2015). The impact of mine tailing could be reduced by the development of appropriate technology that utilises mine tailing as raw materials for building or other purposes. For example, fly ash mixed with mine tailing has been used for the production of geopolymer concrete (Aldred and Day, 2012). Mine tailing can also be used as an additive in the preparation of building materials such as bricks. Other methods that improve tailing managements such as paste and thickened tailings, tailing reuse, recycling and reprocessing are also valuable (Edraki et al., 2014). If not used, tailing wastes should be covered as stated under Section 3, in order to prevent sulphide oxidation and generation of AMD. In another study, sludge produced from AMD has been utilised as an effective adsorbent for phosphate removal from dairy waste water (Wang et al., 2014). Unfortunately, nothing was stated about the fate of phosphate loaded AMD sludge, which requires proper attention in order to reduce environmental pollution.

To date, there is no reliable method for the treatment of AMD. Therefore, prevention and prediction are the best strategies for handling the mine drainage problem and minimising its environmental impact (Abrosimova et al., 2015; Parviainen et al., 2014; Wei et al., 2014). The recovery and reuse of resources from AMD will require universal and suitable technology. The required technology should be designed to enable ease of recovery and reuse of the resources while also solving the environmental problem caused by

AMD. In addition, the technology should take into consideration that when the recovered resources are sold, they should not only cover the cost of treatment but also provide further benefits. The technology, which may aid in the recovery of MNPs, sulphuric acid and pure water in sequential order are extremely important. Recovery and reuse will increase the rate of employment and decrease environmental pollution.

7. Conclusions

The conventional method of treatment of AMD with lime or limestone demands more landfill space for waste disposal. Furthermore, the leaching of toxic metals into the soil and ground water may threaten the environment. Therefore, the occurrence of such problems highlights the need for effective AMD treatment methods in which the utilisation of waste is possible through recycling and reuse. Among methods developed for AMD treatment, the application of MNPs seems very valuable due to ease of recovery and reuse of MNPs. In addition, the recover and reuse of resources from the waste results in cost effective treatment of AMD, clean environment and it is the best option for long term sustainability issue. However, there is a lack of comparative experimental results among the conventional AMD treatment options (neutralisation and precipitation) and modern technologies, particularly MNPs and membrane technologies. Comparison of these active treatment technologies based on the cost effectiveness and quality of the final AMD effluents may aid to identify the best technology to be used in the future.

AMD remediation technologies designed to reduce the amount of waste and enhance the recovery and reuse of the resources that bring financial returns are greatly preferred. However, there are no truly effective treatment options currently in existence. Therefore, it seems to be a long way to go before better, cost-effective and environmentally sound AMD treatment options emerge. Furthermore, properly optimised methods in which the resources are fully recovered and used will guarantee sustainability issue with respect to AMD remediation and reduction of environmental pollution. Thus, proper attention should be given to some of the modern technologies, which capable of recovering resourceful chemicals from AMD. In addition, the following points should be taken into consideration in efforts to minimize generation of AMD and to preserve clean environment.

i. A possible management strategy should be devised so that waste disposal at the mining site can be controlled, by using any possible prevention of AMD generation.
ii. AMD tailing waste should be reused, recycled and reprocessed for utilizations in different industries.
iii. Researchers need to look at the possible combination of SRB and appropriate organic substrate and alkaline chemicals to facilitate toxic metal removal and at the same time reduce the acidity of AMD.
iv. Resource recovery and reuse from AMD should get proper attention.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jclepro.2017.03.082.

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