

Challenges and opportunities in the removal of sulphate ions in contaminated mine water: A review



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ARTICLE INFO

Keywords:

Acid mine drainage
Mine water
Sulphate ions
Sustainability
Water contamination
Water treatment

ABSTRACT

Metal sulphates are one of the major contributors to acid mine drainage (AMD) or acid rock drainage (ARD). AMD occurs by the oxidation of metal sulphides. Sulphides produce sulphates and eventually acidify the medium by converting to sulphuric acid. As AMD contaminates the water sources downstream, the set limit of sulphates for human consumption gets compromised. Stringent standards are imposed on, to comply with limitations set by the regulatory bodies. World Health Organization (WHO) emphasises on a 250 mg/L sulphate concentration in water for human consumption. Therefore, curing AMD of sulphates has become one of the prominent issues in water research. Unregulated disposal of such drainage may cause the increase in salinity and increase in the pH, which can be detrimental to the utility of the water downstream. Corrosion, scaling and health implications are definite results of AMD. Numerous approaches are available to treat sulphates from AMD. Suitability of a specific method depends on the level of removal expected, environmental legislations, available resources, space required, economy and volume of the contaminated water. Several approaches which have demonstrated promising results in the laboratory scale, but their viability at the industrial scale is yet to be established.

This paper reviews the remediation methods which are currently in practice. It discusses the approaches in two main topics, both conventional and recent developments. While the conventional methods include lime, limestone and wetlands, the recent developments include filtration, electrocoagulation, adsorption, ion exchange and precipitation with the introduction of a certain level of novelty throughout the last few years. It is well established that lime and limestone treatment of AMD are well suited for pre-treatment processes whereas the rest of the methods can be selected upon the site specific requirements. Even though the novel methods show their potential to reduce sulphate ions greatly, these need to be tested at industrial scale in order to identify the overall effectiveness.

1. Introduction

Water has been one of the fundamental requirements of human civilisations. Although, water covers more than 70% of the earth's surface, only less than 1% is available for human consumption (National Geographic Society, 2017). Despite the shortage of potable and good quality water around the world, numerous anthropogenic activities have surged the contamination of the available water resources. Acid mine drainage (acid rock drainage or acid and metalliferous drainage) is one such source of water pollution mainly associated with mining and minerals processing activities. This phenomenon naturally occurs due to pyrite oxidation when supported by air and water, where the products are sulphuric acid and dissolved iron. The acidic streams generate consequent to AMD will dissolve heavy metals and pollute both surface and ground water resources. Countries inheriting a legacy of historical

mining, face a serious water pollution threat due to AMD. Even though the contaminated water contains species of heavy metals in addition to high sulphate levels, this work will focus on a specific aspect of AMD namely, the sulphate removal mechanisms and techniques.

Oxidation of metal sulphides is the main contributing factor for the introduction of the sulphates into the water streams. As most of the metals naturally occur as sulphides, those have become common minerals responsible for water pollution especially during mining activities. According to the World Health Organization (WHO) and US Environmental Protection Agency (EPA), water for human consumption should ideally contain less than 250 ppm of sulphates (Balintova et al., 2015; US EPA, 2017) albeit most of the mining effluents exceed the set limit. When the sulphate concentrations for human consumption are compromised, the human health concerns would naturally arise (Backer, 2000). Scaling of pipes and corrosion are major concerns in the

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industrial level once sulphate concentrations are higher and therefore the removal of sulphate from water has become a concern worth addressing (Del Ángel et al., 2014; Gomelya et al., 2014; Hong et al., 2014; Iakovleva et al., 2015; Amaral Filho et al., 2016; Ntui et al., 2016).

Remediation methods are available in different forms namely, physical, chemical and biological (Swanepoel, 2011), which are further divided into active and passive techniques, where the words merely imply the amount of maintenance required. Active methods require high maintenance and the passive methods are not completely independent of maintenance, but require a certain level of maintenance (Johnson and Hallberg, 2005). The traditional sulphate removal techniques include precipitation, but the latest research and technological advancements have helped us understand the intricacies of reactions. Several novel processes have been introduced to treat the sulphate in contaminated water and suitability of the option may depend on the requirement and the economy. For example, AMD treatment with granular activated carbon (GAC) and phytoremediation have recently captured greater attention of the scientific community. In addition, a number of hybrid methodologies have been implemented to catapult the effectiveness of the sulphate removal from AMD (Geldenhuis et al., 2003; Maree et al., 2004). Innovative processes and compounds have been recently developed in laboratory conditions capable of eliminating the sulphate contamination effectively (eg. activated carbon, Amberlyst A21 resins for ion exchange, dissolved air floatation, desilicated fly ash, unactivated attapulgite) but scaling up of those for industrial purposes has been extremely challenging (McCarthy, 2011). Therefore, it is observed that seeking for a perfect remediation method is merely impossible, however, most productive technique is acceptable to be adopted.

This paper will present a critical and a comprehensive review of the various methods in the area of sulphate removal from AMD. Both the traditional and the novel technologies will be discussed in this paper, along with their applications, effectiveness and economic suitability. This will not only present a mere description of the sulphate treatment methods but will also output comprehension of the positives and the negatives of the respective techniques to treat AMD.

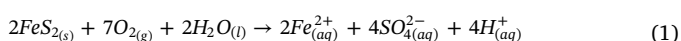
2. Sulphate ions by mining activities

Formation of AMD can take place either naturally or by anthropogenic activities. Naturally, AMD is a part of the rock weathering process, whereas industrial effluents, gas scrubbers and mining activities artificially introduce the sulphates into the eco-systems.

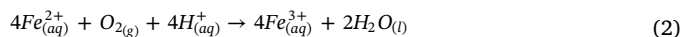
Some of the metals are currently extracted from their respective sulphide ores, for example, iron (pyrite), zinc (sphalerite), copper (chalcopyrite) and nickel (pendanlite) (Keith and Vaughan, 2000; Johnson and Hallberg, 2005). Oxidation of these metal sulphides thus results in the production of AMD (Evangelou, 1995; Howell, 2004).

The ores are chemically stable under in-situ or saturated environments without the contact of oxygen and water, but once exposed to water and atmospheric oxygen, the sulphide oxidation will commence to form AMD (Blodau, 2006). Sulphide oxidation is a natural phenomenon, where the causes are identified to transcend the beliefs of oxygen and water being the usual culprits. Additional sulphide oxidizers are identified as hydroxyl ions and CO_2 in different environmental conditions (Evangelou and Zhang, 1995).

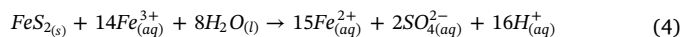
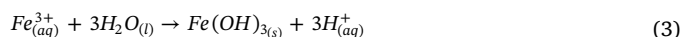
Sulphide oxidation occurs due to both abiotic and biotic reasons. Abiotic oxidation refers to the involvement of an inorganic chemical compound whereas biotic reactions occur due to microorganism involvement (Evangelou and Zhang, 1995; Van der Zee et al., 2003; Johnson and Hallberg, 2003, 2005). Firstly, emphasising on the abiotic oxidation, mining of sulphide ore introduces the stable ore to both air and moisture (Eq. (1)).



Both direct and indirect oxidations are possible in the abiotic process. Eq. (1) presents the direct oxidation reaction where oxygen, along with moisture, is responsible for the production of sulphates. Oxygen dominates further in oxidizing the dissolved ferrous and results in ferric (Eq. (2)).

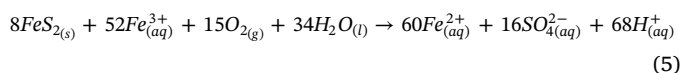


Ferric typically precipitates as $Fe(OH)_3$ at pH values between 2.3 and 3.5 (Akcil and Koldas, 2006, Eq. (3)). Afterwards, the oxidation takes place indirectly and the resulting ferric oxidizes the pyrite (Eq. (4)).



Banks et al. (1997) presented that pyrite oxidation leads to the production of more acid, than it consumes as illustrated in above reactions (Eqs. (1) and (4)) and the medium thus remains acidic. Consequently, the ferric initially precipitates as $Fe(OH)_3$ and the leftover ferric reacts with pyrite and generates an additional sulphate concentration (Johnson and Hallberg, 2003; Akcil and Koldas, 2006).

The overall reaction of sulphate generation (when additional ferrous is available to oxidize pyrite) is given in Eq. (5) (Akcil and Koldas, 2006):



Eqs. (1)–(5) are related to iron based sulphide minerals and the same equations can be inferred for any other mineral inclusive of sulphides as well. The inference can be justified as most of the ores exist in association with pyrite (Spears et al., 1994; Banks et al., 1997). In addition, AMD occurs if 1–5% of sulphur content is available in the form of pyrite in the substance (Tiwary, 2001).

In open-pit mining, water draining from stockpiles of ore, washers, processing areas and waste, augments sulphate concentration of the eco systems (Kuyucak, 1999). However, in underground mining operations water is a major distraction (Rapantova et al., 2007; Mudd, 2008; Altun et al., 2010). Therefore, water is constantly pumped out in order to maintain the water table at a lower level. The process is carried out to hinder the reaction between the minerals and moisture. As the mining activities cease, free access is granted to complete the reaction between water and the ore along with the air. The contaminated water eventually reaches the groundwater sources and contaminates the complete water body.

In addition to the chemical reactions, microorganisms are also capable of oxidizing the sulphide minerals (biotic processes). Prokaryotic microorganisms are well documented as both iron oxidizers/reducers and sulphur oxidizers (Norris et al., 1996; Cha et al., 1999; Johnson and Hallberg, 2003; Rawlings, 2005; Johnson, 2006; Akcil and Koldas, 2006; Vidyalakshmi et al., 2009). The energy released from the sulphide oxidation process is utilized by the microorganisms to remain metabolically active at difficult conditions (Vishniac and Santer, 1957; Johnson and Hallberg, 2003). The main microorganism families are explained thoroughly by Bosecker (1997) as thiobacillus, leptospirillum and thermophilic bacteria. Sub divisions of the above families carry the prefixes T-, L- and Th-respectively. Microorganisms such as Leptospirillum ferrooxidans, Acidithiobacillus (“At.”) ferrooxidans, Acidimicrobium (“Am.”) ferrooxidans and Sulfolobus metallicus are well known for the ability to oxidize the metal sulphides (Johnson and Hallberg, 2003). While the oxidation takes place, sulphuric acid is extensively produced, which lowers the pH and eventually leads to the dissolution of the solid sulphides (Bosecker, 1997) increasing the overall sulphate concentration. However, this characteristic behaviour of microorganisms is utilized and targeted in the area of bio-mining (bio-leaching) for the extraction of gold and base metals (eg. copper)

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