Mining, the environment and the treatment of mine effluents

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Abstract: The environmental impact of mining on the ecosystem, including land, water and air, has become an unavoidable reality. Guidelines and regulations have been promulgated to protect the environment throughout mining activities from start-up to site decommissioning. In particular, the occurrence of acid mine drainage (AMD), due to oxidation of sulfide mineral wastes, has become the major area of concern to many mining industries during operations and after site decommissioning. AMD is characterized by high acidity and a high concentration of sulfates and dissolved metals. If it cannot be prevented or controlled, it must be treated to eliminate acidity, and reduce heavy metals and suspended solids before release to the environment. This paper discusses conventional and new methods used for the treatment of mine effluents, in particular the treatment of AMD.

Keywords: acid mine drainage, high density sludge, lime neutralization, mining environment, passive treatment, sulfate-reducing bacteria.

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1 Introduction

1.1 Mining, the environment and acid mine drainage (AMD)

The potential for environmental damage from every stage of mining activities has been recognized both inside and outside the mineral industry. Guidelines and regulations have been promulgated to protect the environment throughout mining activities, from start-up to site decommissioning. Regulations and guidelines for the mining industry are determined by federal and/or provincial agencies. These regulations can be site specific and vary from site to site owing to differences in geology, mineralogy, terrain, and many other factors. Mining industries must restore the land to its former use or condition, or take protective measures to ensure the land will be suitable for use after mining operations have ceased. For instance, mining industries in Canada are obliged to prepare a Closure Plan and Financial Assurance for site decommissioning during the earliest planning stage of mining, even well before exploration starts.[1] This plan addresses details of the rehabilitation work, disposal methods and monitoring programme. The main aim is to minimize the risk of oxidation of sulfide mineral wastes to generate AMD by ensuring implementation of proper rehabilitation work to the site.

1.2 Sources of mine effluents

Mining and metallurgical processes, during operations and after site decommissioning, generate several types of acidic and toxic effluent that are treated before their discharge to the environment. These effluents include mill tailings excess decant (or reclaim tailings water), acid mine drainage and seepages, and process acid streams.[2] Reclaim tailings waters are of neutral to high pH, containing <20 mg/l total dissolved metals, and polishing with lime is usually sufficient to comply with regulatory limits. Process acid streams originate from the use or generation of acids during metallurgical processes, such as barren solutions, spent electrolyte and weak acids, and recycled process waters. Natural oxidation of sulfide minerals present in mine wastes, tailings and waste rocks results in the occurrence of AMD. AMD is characterized as low pH, high acidity effluents containing heavy metals and sulfate. Control of AMD generation resulting from mining activities is not yet possible. Consequently, treatment of AMD is necessary to meet regulated water quality standards. This paper concentrates on the treatment of AMD.

1.3 Available treatment processes

Lime as CaO or $Ca(OH)_2$ is the most common neutralizing agent used for treating mine effluents, owing to its high reactivity and abundance.[3] However, lime neutralization/precipitation processes may have some drawbacks, such as the poor quality of the final effluent and the need to dispose of a large volume of sludge.[4–6] To improve final effluent quality and sludge disposal, other neutralizing reagents such as $Mg(OH)_2$, Na_2S, NH_3, NaOH and $CaCO_3$ are also used.[7,8] New technologies are sought to minimize high process (capital and operating) costs. The processes based on the use of chemical reagents are usually referred to 'conventional' technologies.

New technologies are mainly based on either physico-chemical (ion exchange, membrane processes, solvent extraction) or biological (e.g. wetlands, biological sulfate reduction) and/or passive process application principles.[9–15] Ion exchange, solvent extraction and membrane processes (reverse osmosis, ultrafiltration, microfiltration, etc.) are employed for site-specific applications, because these methods are still not technically and economically feasible for treating AMD. Passive processes have received attention as alternatives to conventional methods for treating AMD, particularly for replacing common collection and treatment practices that would be unaffordable for low flow streams and seepages.[10–12] Major passive processes investigated to date include: wetlands, sulfate reduction and the use of sulfate-reducing bacteria (SRB); anoxic limestone drains (ALD); and biosorbents. The use of SRB has been investigated for engineered lagoons, reactors, open pits and flooded mines. They all have been found to be suitable for the treatment of low flow and low strength AMD situations. However, ALD have been used widely in the USA for restoration of abandoned mine sites and for pretreating AMD before routing to a constructed wetland.[14]

1.4 Metal removal efficiency

Lime neutralization is efficient for the treatment of common heavy metals. However, complete precipitation of all metals cannot be expected at the same pH in AMD containing a combination of metals such as Cu, Zn, Ni, Cd, Pb, and Fe, as illustrated in Figure 1.

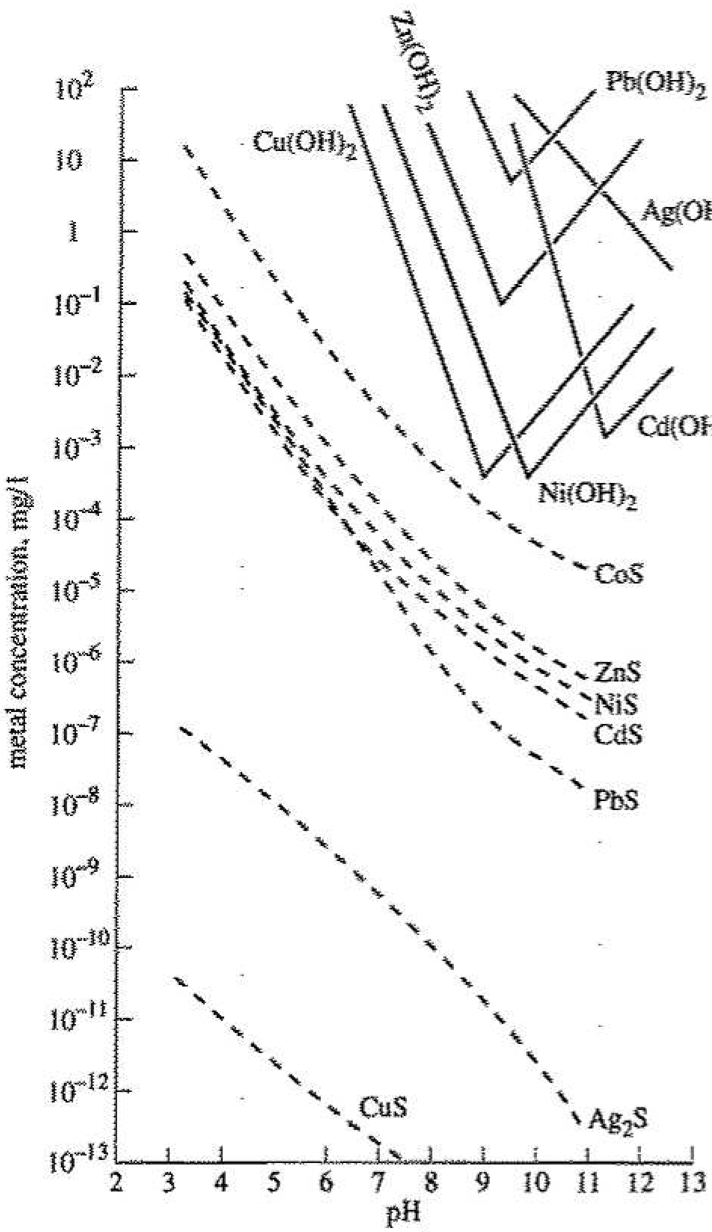

Figure 1 Solubilities of metal hydroxides and sulfides as a function of pH.[16,17]

For metal hydroxides, there is an optimum point at which the solubility limit is achieved and metal hydroxides precipitate out of solution. Above and below the optimum pH, metal hydroxides are more soluble. For instance, at pH 9–10, Cu and Zn can be reduced to levels of <0.1 mg/l and Pb and Fe^{3+} are reduced to µg/l.[15] Levels of Cd and Ni below 1 mg/l can be achieved at pH values above 10, unless the water contains high levels of iron. Cr can be lowered to below 0.5 µg/l at pH 7–8, following reduction of Cr^{6+} to Cr^{3+}. Mn removal requires strong oxidation followed by liming at pH greater than 10. Other contaminants, such as As, Sb, Mo and Se, require additional chemicals (H_2O_2, $FeCl_3$ or $Fe_2(SO_4)_3$, Na_2S, CO_2) as adjuncts to the lime process. A common method for removing Hg is by sulfide precipitation, resulting in an effluent of 10–12 µg/l. Co-precipitation with iron lowers Mo from 4 mg/l to 0.5 mg/l. Ion exchange appears to be an alternative method to achieve low Hg and Mo levels, 1 to 5 µg/l and 2mg/l, respectively. Oxidation of As^{3+} to As^{5+} is necessary to remove As from effluent prior to lime, sulfide and ferric iron precipitation. In some cases, pH adjustment is ineffective, or not quite effective. Therefore, other emerging technologies (ion exchange, membrane separation, solvent extraction, etc.) for metal removal should also be considered to meet the required standards.

2 Conventional processes for treating acid mine waters

2.1 *Lime neutralization*

In lime neutralization processes, acids are neutralized and metals, such as Fe, Zn, Cu, Al, and Pb, are precipitated in the form of metal hydroxides. The mixture of $CaSO_4$ (gypsum) and metal hydroxide is called sludge. The principal reaction in lime neutralization can be expressed as follows.

$$Ca(OH)_2 + M^{2+}/M^{3+} + H_2SO_4 \rightarrow M(OH)_2/M(OH)_3 + CaSO_4 + H_2O \qquad (1)$$

Air is frequently used to oxidize ferrous to ferric iron during precipitation, because ferric iron sludge is chemically more stable than ferrous iron sludge. The sludge produced is allowed to settle in clarifiers/thickeners. When the solid content is less than 1 mg/l, sand-bed filters are employed for polishing, to meet the required level of suspended solids in the final effluent.[18,19] The supernatant is then discharged to the receiving stream, and the settled sludge is disposed of in specifically designed ponds.

2.2 *Lime neutralization processes and critical process parameters*

Depending on site factors, lime neutralization facilities can vary greatly in degrees of sophistication. They range from the simple addition of lime to the tailings pipelines to plants consisting of reactors, clarifiers, and sludge dewatering equipment, as depicted in Figure 2.

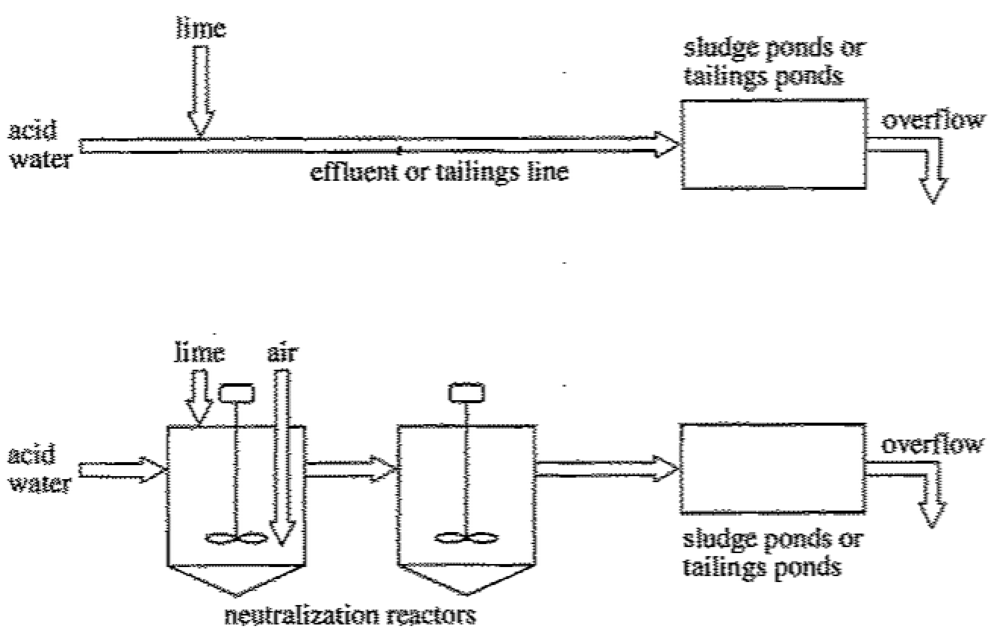

Figure 2 Two types of lime neutralization process.

The sludge densities vary from 1% to 30% solids, depending on the metal concentration of the water and the sophistication of the treatment process. Because the formation of a voluminous sludge is undesirable, the process parameters are set to obtain dense sludge. Major process parameters affecting sludge characteristics are the rate of neutralization, the rate of oxidation, the Fe^{2+} to Fe^{3+} ratio, the concentration of ions, ageing, the recycling of settled sludge, the temperature and crystal formation. The current state-of-the-art lime neutralization process for treating AMD and other acidic waters is called the high density sludge (HDS) process and is capable of producing more compacted sludges than traditional methods of liming.[20] In the HDS process, more than one reactor is used to perform the neutralization (Figure 3). A mixture of sludge, recycled from the clarifier underflow, and lime is used as the alkaline reagent in the first reactor. Both reactors are aerated to oxidize Fe^{2+}, and the pH is continuously monitored. The neutralized AMD with metal precipitates is then flocculated with a polymer, and a clarifier/thickener is used to facilitate solid/liquid separation. The solid content in the resulting sludge is significantly higher (e.g. 10–30%) than when sludge recycling is not performed.[4] A two-step lime neutralization process can produce denser and less voluminous sludges.[21] In this process, the pH of the influent is raised to 4–5 with recycled sludge in the first reactor, and in the second reactor the pH is set to 9–10 and aeration is provided (Figure 4).

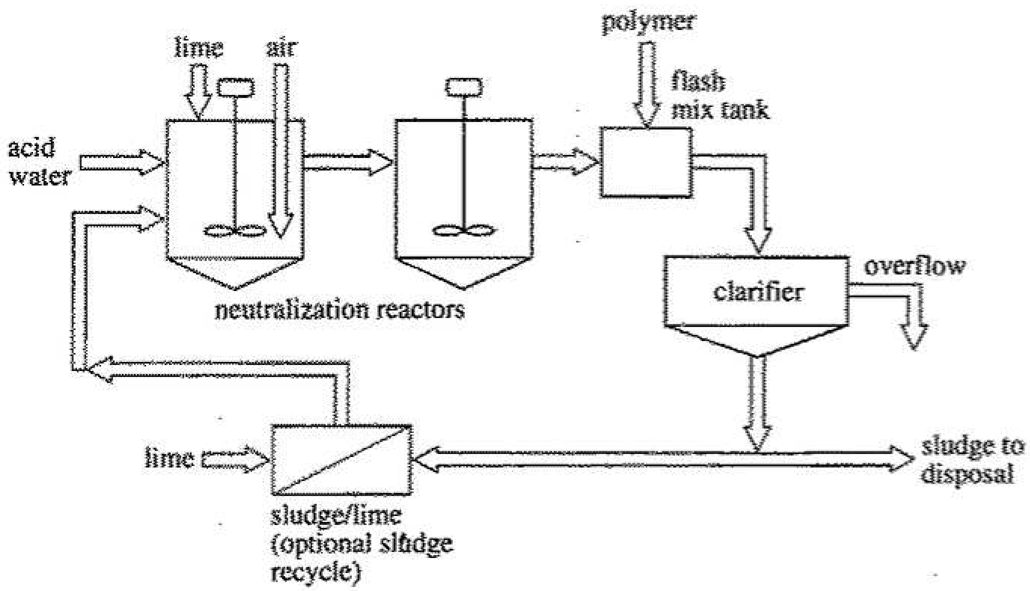

Figure 3 The high density sludge process.

2.3 *Enhanced lime neutralization process*

To be able to meet stringent final effluent limits for controlling dissolved metals and suspended solids, another reagent, such as sodium sulfide (Na_2S), and more sophisticated solid/liquid separation equipment, such as sand filters, are employed. Na_2S (e.g. 5 mg/l) is added in lime neutralization at pH 10.5 to lower Cd to less than 0.01 mg/l in the treated tailings water of the Samatosum treatment plant at Kamloops in British Columbia.[18] Dynasand filters were installed to separate solids and produce a final effluent of low turbidity and concentrations below 0.1 mg/l for each of the heavy metals (i.e. Zn, Pb, Mn, Cd).

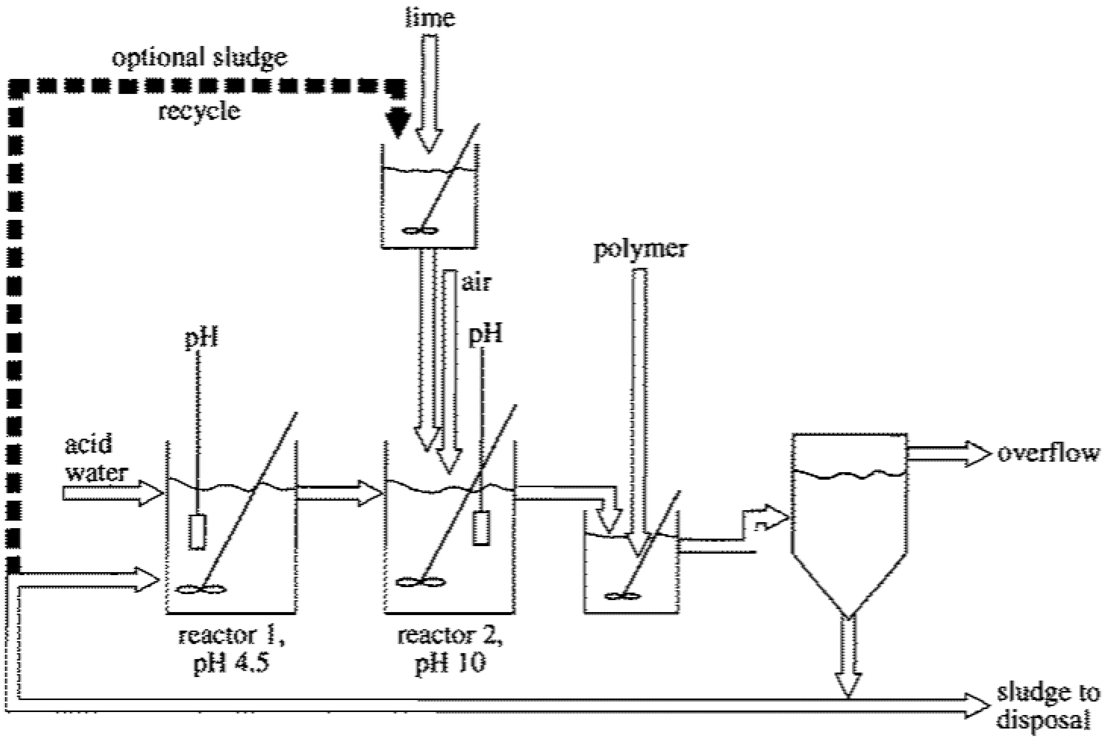

Figure 4 Two-step lime neutralization process.

2.4 *In-line lime treatment*

Neutralization and aeration were combined into single step by injecting a caustic reagent into the port of a jet pump.[22] The overall cost of the process, including capital, operating and maintenance, was also found to be much lower than that of conventional mechanical aerators. The in-line method was recommended to neutralize low-strength AMD containing Zn, Cu, Pb, Mg and Fe^{2+} (e.g. 38, 0.96, 0.4, 17 and <200 mg/l, respectively).

2.5 *Alternative neutralizers*

2.5.1 *Limestone neutralization*

Under controlled conditions, higher density sludges can be obtained by neutralizing AMD using $CaCO_3$ as opposed to lime. Limestone can remove acidity and precipitate iron.[23] Limestone in AMD dissociates and CO_2 gas evolves (Equations 3 and 4).

$$CaCO_3 + H_2SO_4 \rightarrow CaSO_4 + H_2O + CO_2 \quad (3)$$

$$3CaCO_3 + Fe_2(SO_4)_3 + 3H_2O \rightarrow 3CaSO_4 + 2Fe(OH)_3 + 3CO_2 \quad (4)$$

The CO_2 released forms carbonate ion, which acts as a buffer and sets an upper limit on the pH (max 6.5), and also affects the rate and amount of lime consumption.[24] The precipitates may settle very slowly because of their small particle size. Removal of a broad range of metals and ferrous iron cannot be achieved because they require higher pH levels than 6.5. A combined limestone–lime treatment process was suggested for removal of a wide range of metal ions.[23] The Dowa Mining Co. (Japan) uses limestone neutralization to remove iron following biological conversion of ferrous into ferric iron.[25]

2.5.2 *Magnesium hydroxide*

$Mg(OH)_2$ can result in a lower volume and more dense metal hydroxide sludge when it is properly applied in the neutralization system. $MgSO_4$, which is more soluble than $CaSO_4$, forms in the process, and $Mg(OH)_2$ can also remove metals through surface adsorption.[26] However, the rate of neutralization is slow and the buffering capability of $Mg(OH)_2$ prevents the pH from exceeding 9. Depending on the pH requirements, it can be used in conjunction with NaOH. $Mg(OH)_2$ is usually employed in treatment plants, such as Canadian Copper Refinery (CCR) in east Montreal, where the disposal cost of sludges generated is high, in order to reduce sludge disposal costs.

2.5.3 *Sulfides*

Sulfide precipitation (Na_2S) has been used to treat wastewaters from metal finishing industries; it is not routinely used to treat AMD. Solubilities of metal sulfides are usually several orders of magnitude less than those of metal hydroxides.[27] Na_2S, FeS, $(NH_4)_2S$, BaS and H_2S can be used as reagents. Sulfide precipitation results in better metal removal from effluents that contain phosphate, ammonia, organics, surfactants, chelators and Cr^{6+}.[28] However, H_2S evolution from the system, and fine and colloidal metal precipitation, which is difficult to settle and separate, have been found to be potential problems. The mine water occurring in the Laisvall mine of Boliden Mineral AB, Sweden, has been treated with Na_2S to remove low levels of Pb (0.5 mg/l) and Zn (0.7 mg/l). To facilitate solid/liquid separation, a cationic polymer and sand filters are employed.

2.5.4 *Sodium hydroxide*

This has high reactivity and results in less voluminous sludge. This is expensive and the resulting sludge does not settle well, and in most cases requires filtering.

2.5.5 *Ammonia*

Ammonia is preferred by coal mining industries owing to its high solubility and because of reduced sludge volume. It is usually injected near the bottom of ponds or inlet water as gaseous anhydrous ammonia.[29] Some hazards are associated with the handling of ammonia, as well as some uncertainty concerning potential biological reactions. However, ammonia has been suggested to be a feasible alternative to lime for neutralizing low flows containing low acidity.

2.5.6 *Others (waste/by-products of industries)*

Some waste or by-products of industries, such as fly ash from power plants, crude oil combustion and gasification processes, have potential as a lime substitute for the treatment of AMD. However, metal contaminants present in fly ash may raise some concerns, and it reacts more slowly than lime.

2.6 *Coagulation/flocculation for better solid/liquid separation*

Following neutralization, the fine particles (precipitates) in suspension need to be aggregated to improve the solid/liquid separation or sedimentation in clarifiers and dewatering of the sludge for further compaction in basins.[30,31] Coagulation is a specific type of aggregation, which leads to formation of aggregates, called flocs, that are compact.[32] The addition of a coagulant, such as inorganic Al^{3+} or Fe^{3+} salts or organic polymers, helps to discharge or destabilize the electronegative colloids and bridge the neutral particles. The type of polymer, the temperature of the system, the viscosity and the chemical characteristics of the pulp, and external stirring are important parameters in the flocculation process.

2.7 *pH adjustment for meeting final effluent quality requirements*

When it is required to lower the pH to between 6.5 and 8.5 in the final effluent following neutralization at a higher pH, the pH is adjusted to the desired level with either sulfuric acid or CO_2. If an increase in the water alkalinity is not required or an increase in SO_4 levels is not a concern, sulfuric acid is preferred.[18,33]

2.8 *Sludge dewatering options*

Lime treatment plants generally use one basin for the dual purpose of effluent clarification and sludge thickening. Dewatering via filters has not been widely practised in the mining industries and has been considered as a very site-specific application. However, sludges containing less moisture and high metal value can be sent to recovery operations (i.e. a smelter), or high sludge disposal costs can be reduced with an appropriate dewatering method.[8] It was reported that freeze-dewatering is an efficient alternative technique, and a single freeze–thaw cycle could reduce the volume of sludge by 90%. A feasible rate of freezing can be obtained by manipulating the depth of the sludge.[34]

2.9 *Sludge stability and fixation*

During storage of the sludge, heavy metals may become solubilized and released in the water in the sludge pond and into ground and surface waters. The possibility of metal mobilization is determined by a 'leaching test' (EPA's TCLP, Menviq, etc.). Depending on the sludge characteristics and site-specific requirements the sludge is stabilized by mixing it with cement and/or lime prior to its disposal.

3 Emerging treatment processes and biological passive methods

3.1 *Wetlands*

Organic and inorganic compounds and suspended solids can be removed. A wetland is usually composed of two zones: an oxidation zone, which is vegetated with aquatic plants, and a reduction zone, which is a sedimentation zone rich in sulfate-reducing bacteria (SRB), denitrifying and Mn-reducing bacteria. Plants play a filter role, take up metals and help the oxidation processes to occur, and bacteria catalyse chemical reactions.

3.2 *Sulfate reduction*

SRB convert SO_4 in AMD into H_2S, and carbonic nutrients into bicarbonates.[10] Thus, the reaction products play an important role in removing metals as sulfides and increasing alkalinity (pH). Organic materials, such as straw, sawdust, wood shavings, and manure, are added to the system to provide a slow release of nutrients to bacteria. Sometimes, limestone and soil are also added to the nutrient mixture to increase the alkalinity and, subsequently, enhance the activity of SRB. As the use of SRB in engineered lagoons, open pits and flooded mines is under development, their use in controlled reactors has been implemented by Budelco (the Netherlands) to remove metals and SO_4 from underground mine water.[35] Parameters critical to the success of the process are temperature, nutrients, alkalinity, retention time, bacterial population, contaminants present in AMD and loading rates (flow, acidity and metals).

3.3 Anoxic limestone drains (ALD)

An ALD system generally consists of an excavated seepage interception trench backfilled with crushed limestone and covered with plastic and clay-soil to keep air out.[14] ALD basically provides an increase in alkalinity. Oxidation of Fe^{2+} and formation of ferric oxyhydroxides, which armour the limestone and prevent an increase in alkalinity, are thus avoided. Designs of ALDs are reported to be site-specific.[36] Usually, an ALD system is followed by a wetland for oxidation and precipitation of iron and other contaminants.

3.4 Biosorbents

Biological materials such as sawdust, sphagnum moss or algae can be used as adsorbents (or biosorbents) in treating AMD. A bed of biosorbents can be placed where the seepage occurs. When it is saturated by metals, its proponents suggest it can either be disposed of (with tailings or recycled to a smelter), or washed with an appropriate eluate for the recovery of metals.[9]

4 Cyanide destruction and removal of nitrogen compounds

The widely used method for removal of cyanides and ammonia is natural degradation in holding ponds. Cyanides are biologically or chemically oxidized and are then converted into ammonia and carbon dioxide. Natural degradation of ammonia involves the evaporation of dissolved ammonia gas from the wastewater.[2,36] Removal is enhanced by increasing the pond area, increasing the pH and allowing for more contact with air. A biological process unique to Homestake Mining in South Dakota decomposes metal cyanide complexes and efficiently oxidizes cyanides to ammonia, which is further oxidized by bacteria (nitrification) to nitrate. Base metal cyanide complexes are selectively oxidized to cyanate by a mixture of SO_2 and air, in the presence of copper as a catalyst, in a controlled pH range. This a proven process and is known as the Inco Method. A number of operations use hydrogen peroxide to oxidize cyanides to cyanates. Hemlo Gold (Noranda) adds the premixed $CuSO_4/FeSO_4$ reagent to the mill solution, in which the pH is controlled at 9.5 and cyanide is removed with cuprous ions. Other methods, such as air stripping, steam stripping, alkaline chlorination with hypochlorite at pH 10–11, engineered wetlands, acidification/volatilization, adsorbents and ion exchange resins for the removal of cyanides and ammonia, have limited use. The potential processes for removal of nitrates and nitrites include biological denitrification in which nitrates/nitrites are reduced to nitrogen gas, ion exchange, and reverse osmosis in which nitrates/nitrites are removed from the water and are obtained in a very concentrated form requiring further disposal methods. Wetland filtration has limited use.

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