



A REVIEW OF PASSIVE SYSTEMS FOR THE TREATMENT OF ACID MINE DRAINAGE

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ABSTRACT

This review presents the current state of development of the passive mine water treatment technologies. The background of passive treatment is reviewed and the chemical and biological processes involved in metals removal and acidity neutralisation are detailed. The types of currently existing passive treatment technologies and their applicability range as defined by the mine water chemistry are presented. Finally, the performance of passive systems constructed for the treatment of acid mine drainage from both coal and sulphide metal mines is summarised.

Keywords

Acid rock drainage; bacteria; environmental; pollution

INTRODUCTION

The principles of acid mine drainage (AMD) generation are fairly well understood [1, 2]; pyrite and other sulphide minerals on exposure to oxygen and water and in the presence of oxidising bacteria, such as *Thiobacillus ferrooxidans*, oxidise to produce dissolved metals, sulphate, and acidity. The resulting solution interacts with other mineralogical constituents in secondary reactions such as acid-induced metal dissolution, ion-exchange, and neutralisation.

Acidic drainage is a persistent environmental problem at many active and abandoned sulphide and coal mine sites. Untreated AMD is polluting receiving streams and aquifers and the overall effect on streams and waterways can be very dramatic. In the worst cases, all aquatic life virtually disappears, river bottoms become coated with a layer of rust-like particles, and the pH decreases. In Table 1 typical composition of water effluents resulting from sulphide and coal mine operations are presented along with the respective permissible levels for the mining effluents in USA.

In addition to pH and chemical composition, acidity and alkalinity, both of them expressed in mg CaCO₃/L, are also important parameters for the characterisation of mine waters. Acidity expresses the quantitative capacity of a water to react with a strong base to pH 8.3. The major components of mine waters acidity are proton acidity, associated with pH, and mineral acidity related to dissolved metals content. Alkalinity of a water presents a measure of its acid-neutralising capacity and it is defined as the sum of all the titratable contained bases. The alkalinity of waters is primarily a function of their carbonate (CO₃²⁻), bicarbonate (HCO₃⁻), and hydroxide content (OH⁻). Contribution from borates, phosphates, silicates or other bases present is also included in the measured values [6].

TABLE 1 Typical mine drainage composition and permissible levels for industrial effluents in USA

Composition metals in mg/L	Coal mines [3]	Cu-Pb-Zn mixed sulphide mines [4]	Limits for industrial effluents disposal in USA [4, 5]
pH	2.6 – 6.3	2.0 – 7.9	6 – 9
Fe	1 – 473	8.5 – 3,200	3.5
Zn		0.04 – 1,600	0.2 – 0.5
Al	1 – 58		
Mn	1 – 130	0.4	2
Cu		0.005 – 76	0.05
Pb		0.02 – 90	0.2

Several methods exist for the treatment of mine waters, depending upon the volume of the effluent, the type and concentration of contaminants present. An effective treatment is sought to generate water of neutral pH and low acidity, and to reduce the levels of the sulphates, iron and other metals present down to the environmental limits. To be attractive, this process must be of low-cost, easy to install and maintain, and producing limited quantities of solid by-products.

The conventional mine drainage treatment systems involve neutralisation by addition of alkaline chemicals such as limestone, lime, sodium hydroxide, sodium carbonate or magnesia to water; as a result pH is raised followed by the precipitation of metals. These active systems generally require the installation of a plant with agitated reactors, precipitators, clarifiers and thickeners with increased costs for reagents, operation, maintenance, and disposal of the resulting metal laden sludge. Given that the drainage on many sites will likely be contaminated for decades after the decommissioning of the mine, there is of financial incentive to develop alternative low-cost and low-maintenance treatment schemes, such as passive systems.

Passive treatment schemes take advantage of naturally occurring geochemical and biological processes in order to improve the quality of the influent waters with minimal operation and maintenance requirements. The pH of mine drainage is raised when the water mixes with alkaline water or through direct contact with carbonate rocks. The removal of metal contaminants is then effected through the precipitation of hydroxides, oxyhydroxides and sulphides; local conditions such as oxygen content, water and soil chemistry dictate whether these reactions will occur under oxidising (aerobic) or reducing (anaerobic) environments [3, 7].

During the last decade, passive systems have been developed from an experimental concept to full-scale field application. They have mainly been used to treat acidic waters with low metals content; in the Appalachia region alone more than 200 wetlands are operating for the treatment of coal mine drainage [8]. Passive treatment schemes remediate the acid drainage quality with minimal operation and maintenance. The costs of passive treatment schemes are generally measured in their land requirements rather than labour and consumables, since these systems employ slow processes for contaminants removal and thus require longer retention times than the conventional systems and large areas to achieve similar results [3].

In order to evaluate the potential application of constructed wetlands for the AMD treatment, it is essential to comprehend the movement, retention and transformation of metals in wetlands ecosystems. This information is vital for the selection of the optimal process route and the development of design criteria to be used in the wetlands construction. The aim of this review study is to describe the major processes encountered in passive systems for contaminants removal, the types of currently existing passive technologies and the design recommendations based on water chemistry.

BACKGROUND OF PASSIVE TREATMENT

Acid drainage remediation by passive systems was first documented in the early 1980's during studies conducted by Hunstman *et al.* and Wieder and Lang; natural *Sphagnum* mosses wetlands that received acid mine drainage resulted in the amelioration of water quality [9, 10]. These observations stimulated the idea that engineered wetland systems might be used for the intentional treatment of acid mine waters. During the last decade, many research and reclamation groups have experimented to evaluate the feasibility of these schemes. Most of these systems have been developed to treat acidic coal mine drainage, with limited applications reported on sulphide mine waters [11].

Since natural wetland treatment of mine drainage was found in *Sphagnum* dominated bogs, most of the early systems were attempts to construct such systems but with highly variable results [12, 13, 14, 15]. *Sphagnum* proved quite sensitive to transplanting, abrupt changes in water chemistry and increased accumulation of iron. At most sites the moss died within the first growing season [16].

Despite these initial negative results, the research on different types of constructed wetlands was continued. Eventually, a wetland design evolved that proved tolerant to contaminated mine drainage and was effective at reducing the levels of dissolved metals. Most of these treatment systems consist of a series of small wetlands that are vegetated with cattails, *Typha latifolia*, occasionally with the addition of organic substrate in which the cattails root [17, 18, 19].

For the development of the most recently applied passive treatment schemes anaerobic processes were found to be important in metals removal and ecosystems are not needed; the cells were effectively working without plants. Ponds, ditches, and rock-filled basins have been constructed that are not planted with emergent plants, and in some cases, contain no soil or organic substrate [20]. Pretreatment systems have been developed where acidic waters contact limestone in an anoxic environment before flowing into a settling pond or wetland system [21].

MECHANISMS FOR CONTAMINANTS REMOVAL

Several physical, chemical and biological processes are known to occur within wetlands to reduce the metal concentrations and neutralise the acidity of the influent water; however, limited quantitative data are presently available regarding the relative importance of the different mechanisms in the overall performance of the wetlands.

The simplest mechanism encountered in a wetland is dilution. In some systems, major inflows of uncontaminated water may cause changes in the water chemistry that might be mistakenly attributed to biological or chemical processes [3].

Even after dilution is accounted for, many constructed wetlands have considerable effect on the acidity of AMD and the concentrations of dissolved metals [22, 23]. Filtering of suspended material, metal uptake into live roots and leaves, adsorption and exchange by plants, soil and other biological materials, abiotic or microbially-catalysed metal oxidation and hydrolysis reactions in aerobic zones, and microbially-mediated reduction processes in anaerobic zones result in the amelioration of water quality. The above mechanisms as related to iron removal in a wetland are schematically presented in Figure 1 [16]. Given that wetlands have heterogeneous environmental conditions, these processes are intertwined.

In the following sections the main processes encountered in passive treatment systems for the removal of contaminants are summarised:

- Oxidation and hydrolysis
- Metal removal by plants, algae and organic substrates
- Reduction
- Limestone addition

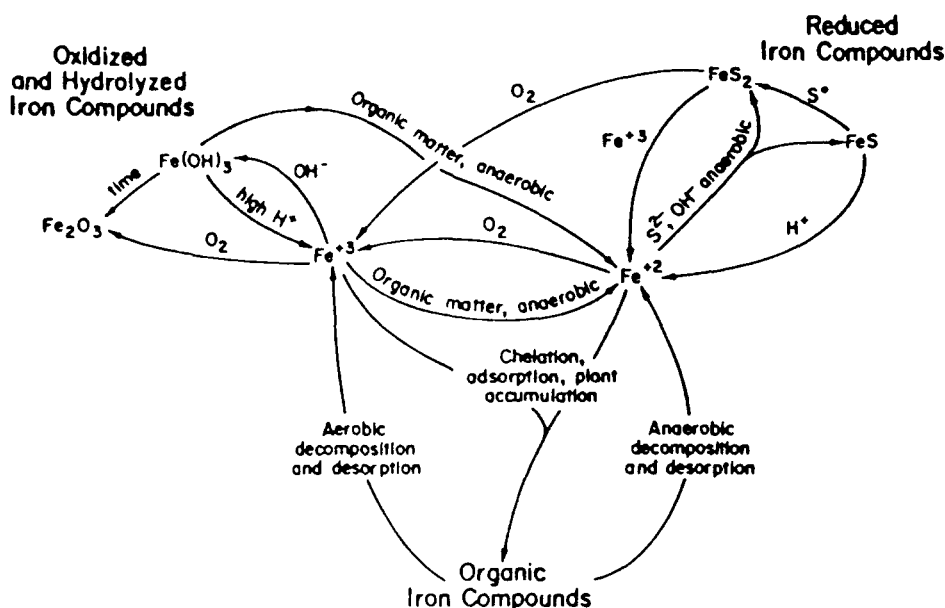


Fig.1 Forms of iron in a wetland receiving mine drainage and the respective environmental conditions [16].

Oxidation and hydrolysis processes

The most important metal removing mechanisms in aerobic wetland systems are associated with the oxidation and hydrolysis reactions that eventually result in the precipitation of dissolved metals. As most of the reported applications of passive systems refer to the treatment of coal mine drainage with Fe and Mn being the major contaminants, studies on removal mechanisms refer mainly to these elements.

Iron oxidation and hydrolysis

Iron which is the major contaminant in coal and sulphides mine drainage, is the metal most susceptible to oxidation in wetlands. It usually drains from spoils or underground mines in the reduced ferrous form which being unstable in oxygenated environments eventually oxidises to ferric iron:



Ferrous iron oxidation occurs both abiotically and is also catalysed by bacteria; the kinetics of both mechanisms are pH dependent.

The oxidation rate of Fe^{2+} under abiotic conditions as a function of pH is illustrated in Figure 2.

At $\text{pH} \geq 5$, the kinetics of oxidation can be described by the following equation [24]:

$$-\frac{d[\text{Fe}^{2+}]}{dt} = k_1[\text{Fe}^{2+}][\text{OH}^-]^2 p_{\text{O}_2} \quad (2)$$

where $k_1 = 8.0 \times 10^{13} \text{ min}^{-1} \text{ atm}^{-1} \text{ mol}^{-2} \text{ l}^2$.

Thus, a 100-fold increase in the reaction rate occurs for a unit increase in pH. Under these conditions, the most important role of the constructed wetland is to provide adequate retention time for the dissolved iron to oxidise and precipitate.

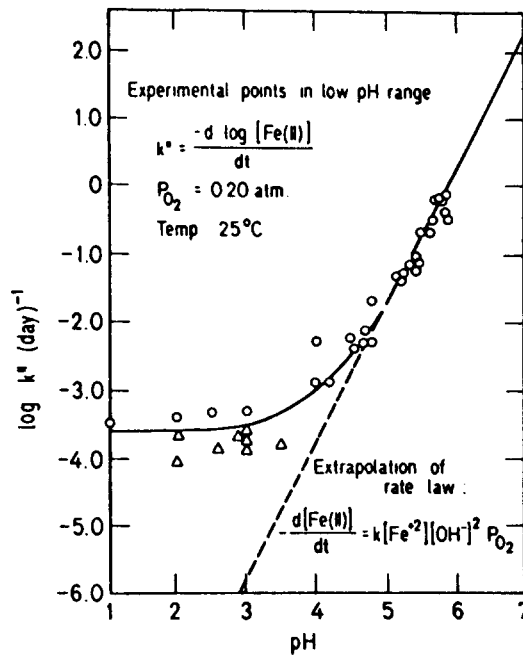


Fig.2 Oxidation rate of ferrous iron as a function of pH [24].

At $\text{pH} < 3.5$, the kinetic rate of the abiotic oxidation of Fe^{2+} is described by the equation:

$$-\frac{d[\text{Fe}^{2+}]}{dt} = k_2[\text{Fe}^{2+}]P_{\text{O}_2} \quad (3)$$

where $k_2 = 1.0 \times 10^{-7} \text{ min}^{-1} \text{ atm}^{-1}$

Under these conditions, the oxidation is slow and the contribution of iron-oxidising bacteria becomes important [25]. Bacteria, such as *Thiobacillus* and *Ferrobacillus ferrooxidans*, are acidophilic aerobic chemoautotrophic species that thrive in pH 1.5–3.5 and that act as electron acceptors during the Fe^{2+} oxidation, increasing thus the reaction rate by several orders of magnitude. In experimental wetland systems, the numbers of iron-oxidising bacteria have been positively correlated with iron oxidation and removal [26].

As ferrous iron is converted to ferric, it is subjected to hydrolysis that results in the precipitation of hydroxide that quickly blankets as an orange sludge the substrate surface and in the production of protons H^+ , according to the reaction:



The hydrolysis reaction occurs abiotically and depends on pH and on the total Fe^{3+} concentration. From the ferric hydroxide solubility diagram given in Figure 3 it is seen that under equilibrium conditions negligible concentrations of dissolved ferric iron exist at pH above 3 [24].

The solubilities of iron and some other common metal hydroxides as a function of pH are given in Figure 4 [27].

Since the net result of iron oxidation and hydrolysis is the production of hydrogen ions, H^+ , natural or constructed wetlands receiving neutral iron containing waters commonly result in an outflow with a final pH of 2.0 or 3.0 after most of the iron is hydrolysed.

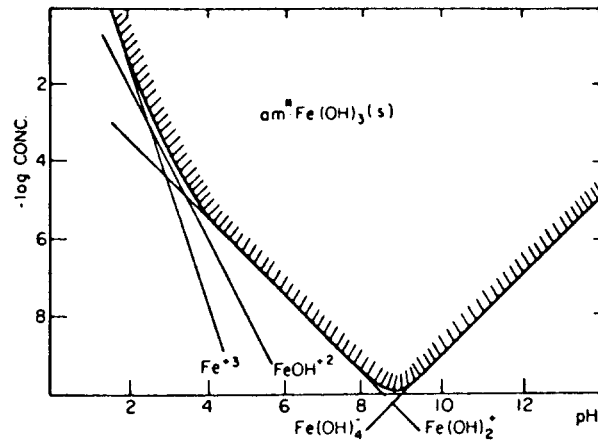


Fig.3 Solubility diagram of Fe(OH)₃ [24].

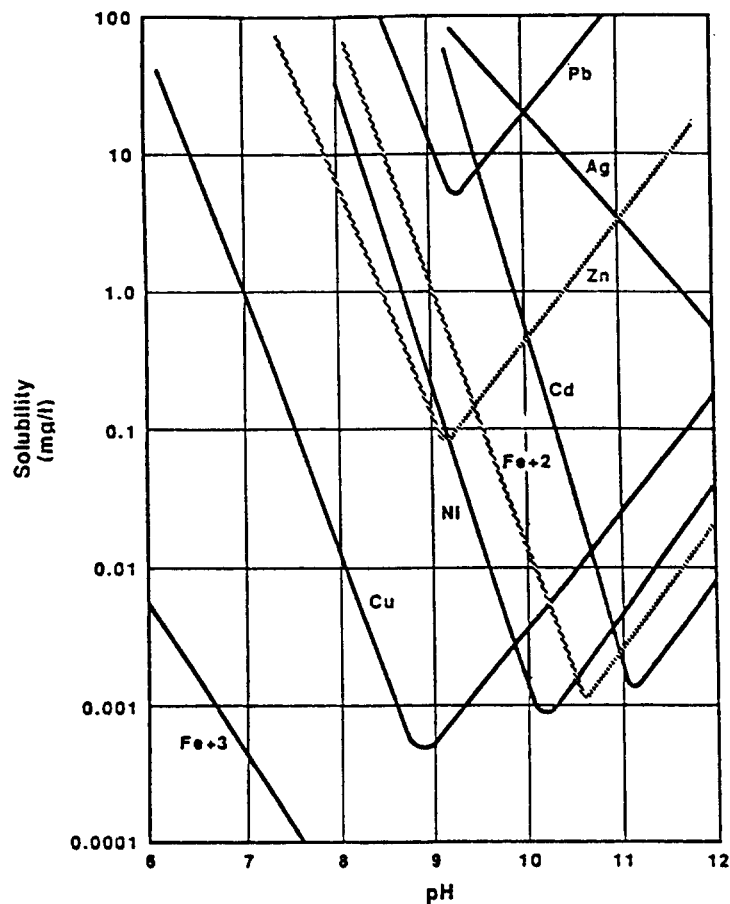


Fig.4 Solubilities of metal hydroxides as a function of pH [27].

Manganese oxidation and hydrolysis

Manganese is another common contaminant in mine drainage. Efficient Mn removal by hydroxide precipitation requires the oxidation of Mn²⁺ to the higher oxidation forms, however the specific mechanism(s) encountered in aerobic mine waters are not fully elucidated. Hedin *et al.* suggest that Mn²⁺ may be oxidised to either the trivalent or tetravalent form, which then precipitates as MnOOH [3]:



MnOOH over time is likely to oxidise to the more stable MnO_2 .

The kinetics of Mn^{2+} oxidation, are strongly pH dependent as seen in the following integrated form of the rate expression [24]:

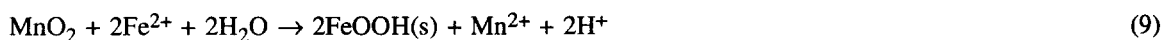
$$-\frac{d[\text{Mn}^{2+}]}{dt} = k_1[\text{Mn}^{2+}] + k_2[\text{OH}^-]^2 p_{\text{O}_2} [\text{Mn}^{2+}][\text{MnO}_2] \quad (6)$$

This reaction is described as “autocatalytic”, given that MnO_2 solids act as adsorption surfaces for Mn^{2+} catalysing thus the oxidation reaction [24]. Manganese abiotic oxidation is very slow at pH lower than 8, however, in many natural waters the oxidation of Mn^{2+} to MnOOH or manganate (Mn^{4+}) is enhanced by manganese-oxidising microorganisms which are most readily found at pHs above 7.0 [28].

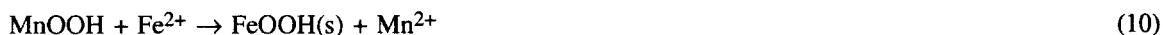
In alkaline environments, Mn^{2+} can also precipitate in the carbonate form, which in the presence of oxygen may also further oxidise to MnO_2 [29]:



Data collected from wetland applications indicated that as a mine water with increased concentrations of Mn and Fe flowed through the system, Mn removal was less effective than iron and occurred sequentially, not simultaneously [3]. Simultaneous precipitation of Fe and Mn from aerobic alkaline waters is possibly prohibited from the reduction of oxidised Mn by ferrous iron [3]:



or



In conclusion, Mn removal is a slow process even in the absence of ferrous iron. It is expected that conditions prevailing in wetlands will accelerate the above mechanisms, however field data suggest that inefficient manganese reduction is one of the “great frustrations in the use of wetlands for the treatment of acid mine drainage”.

Metal removal by plants, algae and organic substrates

Plants

Emergent vegetation is one of the components in wetland systems. However, accumulation of metals in plant biomass is reported as a minor fraction of the overall metals removal [30]. On the other hand, the ability of emergent plants to diffuse oxygen from the roots into the surrounding substrate and the water column has been proposed as an important indirect plant function in engineered wetlands [31]. The result is the formation of localised oxidising zones within the substrate, which conduces to metals removal by oxidation.

Another significant role that the plant material play in a wetland is to provide the substrate of the wetland with the biomass required for the other processes. Decayed plant material produces organic matter that is capable of removing metals by adsorption and exchange whereas plants cellulose provides the nutrients for the sulphate reducing bacteria.

Additional benefits in the wetlands performance associated with the emergent plants include alteration of the permeability and hence the flow dynamics in the upper part of the substrate, effects upon the water budget of the system through evapotranspiration, improvement of the aesthetics of the system and reduction of erosion by wind at periods when the water level drops below that of the substrate.

As already stated *Sphagnum* mosses used in many of the original constructed wetlands present low tolerance to acid mine drainage [16]. The most common emergent plants presently used in wetland systems are cattails with *Typha latifolia* as the predominant species and less commonly *Typha angustifolia*. *Typha* is readily available in most sites, transplants well, and has proven tolerant of a wide range of conditions [30, 32].

Algae

Certain algae have recently received attention from wetland researchers since the presence of algal blooms was occasionally associated with decreased manganese levels in the waters. Kepler [33] reported that a bloom of *Oscillatoria* sp. (a bluegreen alga) collected from a constructed wetland where manganese concentrations were effectively reduced, contained up to 56 g Mn/kg (dry weight) of algae. *Microspora* specimens sampled by the US Bureau of Mines contained 30–90 g/kg of Mn [16], whereas manganese encrustations were also observed in numerous samples of *Microspora* and *Oedogonium*, collected from constructed wetlands [34].

Algae utilise CO₂, sulphates, nitrates, phosphates, water, and sunlight to synthesise their own organic cellular material and give off free oxygen as a waste product [35]. The oxygen produced is available to bacteria and other microbes for their metabolic processes, which include respiration and degradation of organic material in the pond. Algae productivity may be enhanced with minor nutrient additions [7]. Thus, we have a complete cycle in which: (a) microorganisms use oxygen dissolved in water and (b) break down organic waste materials to produce (c) waste products such as CO₂, H₂O, nitrates, sulphates, and phosphates, which (d) algae use as raw materials in photosynthesis, thereby (e) replenishing the depleted oxygen supply and keeping conditions aerobic so that the microorganisms can function at top efficiency. However, algae are less effective in winter and when they die, they impose secondary organic loading on the pond. During winter months in cold climates ice and snow covers prevent sunlight from penetrating the pond, adversely affecting the size of the algae populations as well as the mixing and re-aeration of the pond. Algae are not necessarily killed in the absence of sunlight, but under these conditions their activity is significantly impaired.

Organic substrates

Exchange of dissolved metals with the humic and fulvic acids, the natural organic materials found in abundances in wetland substrates, especially peat, is another possible mechanism through which the metals are temporarily retained in a wetland [31]. Both groups of compounds have carboxyl and phenolic acid groups attached to a larger organic molecule and will readily dissolve in basic solutions. The exchange with metals is primarily controlled by these acid functional groups and can be described by the following equilibrium reactions:



The acid portion is represented by the carboxylic group, —COOH, that dissociates to the carboxyl, —COO—, and hydrogen ions (Eq. 11). The R— represents the inert, organic portion of humic or fulvic acid. Upon dissociation, the carboxyl ion can react with metal ion M²⁺ forming a complex.

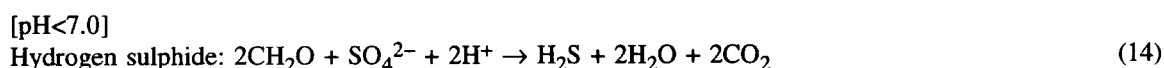
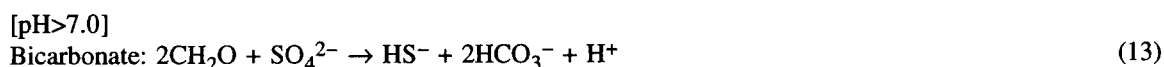
When the metal laden acidic drainage is brought in contact with humic materials, two are the possible consequences: if the acid groups of the organic substrate are sufficiently ionised, then the metal complexation equilibrium reaction will be strongly shifted to the right. On the other hand, if the substrate

already exhibits a high metal concentration then when contacted by an acidic solution, the above reaction may be reversed, resulting in the metals release. Consequently, the metal absorption capacity of humic materials is limited.

Depending on the specific metal ion, adsorption functions best at pHs ranging from 4.0 to 6.0. Thus, the pH of the mine water has to be raised for adsorption to take place. At pH 4.7, the following adsorption order was defined ranging from 100% to 10% metal removal: $\text{Hg} = \text{Fe} = \text{Pb} = \text{Cu} = \text{Al} = \text{Cr} > \text{Cd} > \text{Ni} = \text{Zn} > \text{Co} > \text{Mn}$ [36]. Therefore, metals as Mn, Zn, and Cd, which are often associated with mine drainage, are not easily removed by adsorption. Iron accumulations in peat samples collected from natural and constructed wetlands were as high as 2–5 g/kg dry weight [37, 38].

Reduction processes

Another significant mechanism encountered in wetlands is the anaerobic bacterial sulphate reduction taking place in the organic substrate. In an anaerobic wetland system, sulphate-reducing bacteria of the *Desulfovibrio* species [39], use sulphate to oxidise organic matter and release bicarbonate and hydrogen sulphide as waste products, according to the following reactions [7]:



where CH_2O represents the organic matter.

Bacterial sulphate reduction is limited to certain environmental conditions [39]. The bacteria appear to function best at pH>4, in the presence of a sulphate source, suitable concentrations of organic matter, and in the absence of oxidising agents such as O_2 , Fe^{3+} and Mn^{4+} , requirements which can be normally satisfied in the anoxic wetlands environments.

Under these conditions, the produced hydrogen sulphide reacts with dissolved metals to form insoluble metal sulphides that subsequently precipitate according to the reaction:



For dissolved zinc, amorphous zinc sulphide is formed whereas for iron the formation of pyrite is possible according to the reactions:



The removal of dissolved metals in the sulphide form depends on pH, the solubility of the specific metal sulphide, and the concentrations of the reactants. The solubilities of various common metal sulphides are presented in Figure 5; CuS is the first metal sulphide to precipitate whereas MnS presents the higher solubility, $K_{\text{sp}}=5.6 \times 10^{-16}$, and it is expected to precipitate after the quantitative removal of all other metals from solution.

It should be noted that an essential prerequisite for sulphides precipitation to occur is the presence of sufficient organic matter to maintain an anaerobic environment in the wetland. Thus, periodic addition of selected organic materials may be required.

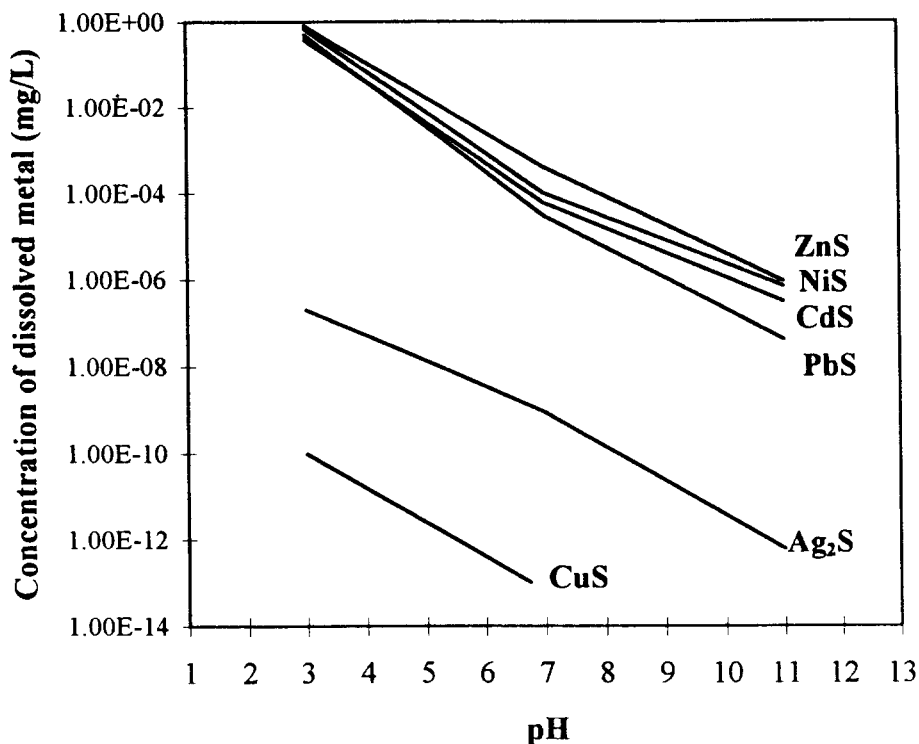


Fig.5 Solubilities of metal sulphides [40].

Although at low pH the growth and activity of sulphate-reducing bacteria is inhibited, their activity results in pH increments in their immediate environment, allowing their activity in the sediments beneath extremely acidic waters [41, 42]. Kleinmann and Hedin [43] reported that sulphate reducing bacteria are also tolerant of high heavy metal concentrations whereas the sulphate-reduction potential is not inhibited in the presence of nickel, lead or zinc at levels averaging 60 mg/L.

Bacterial sulphate reduction is potentially very important for the long term prospects of constructed wetlands. As opposed to the voluminous blanketing sludge that results from iron oxidation and hydrolysis, iron sulphide precipitates are more dense and form within the organic substrate. The processes involved are the reverse of pyrite oxidation; in fact, acidity is consumed by the sulphate reduction processes. Finally, unlike bacterial oxidation and hydrolysis processes which are not effective in removing metals such as Zn and Mn at pHs lower than 8.0, hydrogen sulphide produced by bacterial sulphate reduction readily reacts with heavy metals at pH \geq 3 to form virtually insoluble sulphide compounds [4].

Limestone addition

Another method for adding alkalinity to acidic water is through its direct contact with limestone; limestone dissolves and produces calcium and bicarbonate alkalinity, the latter neutralising acidity and buffering pH. The solubility of limestone depends on pH, temperature and CO₂.

More specifically, when acidic waters, pH<6.4, contact limestone, the limestone reacts according to the equation [44]:



Dissolved CO₂, conventionally noted as H₂CO₃*, is a weak acid and continues to react with limestone, producing calcium and bicarbonate alkalinity that is available for acid neutralisation reactions:



The effectiveness of this treatment is practically reduced in the cases that the influent contains appreciable amounts of ferrous iron and contacts limestone in an oxidising environment. In this case, the limestone is rapidly coated with ferric hydroxide precipitates, the dissolution rate decreases and the production of alkalinity virtually ceases [45]. However, in an anoxic environment, i.e. in the absence of oxygen, dissolved iron remains in the ferrous state, Fe^{2+} , and no precipitation occurs at $\text{pH} < 5.5$. Thus, limestone dissolution and subsequent production of alkalinity can proceed without inhibitory armouring. Furthermore, the closed nature of anaerobic environments allows the increased generation of alkalinity due to elevated CO_2 levels.

The idea of generating alkalinity with anaerobic limestone treatment systems has recently gained attention by the mining community [21, 46, 47, 48]. These passive systems designed for mine water pretreatment have become known as anoxic limestone drains and they introduce buffering capacity, in the form of alkalinity, into the acid drainage. The effluent of the anoxic drains may then be routed through a constructed wetland for the subsequent removal of the contained metals. pH changes in the wetlands due to acid produced from metals hydrolysis are buffered due to the high alkalinity already added to the influent.

Upon review of the wetland processes and on the question of the relative importance of each mechanism already presented, the answer is that all processes contribute in the overall performance of the system. The wetland design problem then becomes how to develop stages that will employ the group of available mechanisms so as to effectively remove the specific metal contaminants and control the pH to values complying with the environmental regulations.

TYPES OF PASSIVE TREATMENT SYSTEMS

Three are the principal types of passive technologies developed for acid mine drainage treatment: (1) aerobic wetland systems, (2) anaerobic organic substrate systems and (3) anoxic limestone drains. Aerobic wetland systems promote mixed oxidation and hydrolysis reactions, and are mainly applicable to the treatment of *net alkaline* waters. Organic substrate systems enhance anaerobic bacterial activity that results in the sulphate reduction followed by the precipitation of metal sulphides and the generation of alkalinity. Anoxic limestone drains add alkalinity to the waters and may be used as a pretreatment stage before the wetlands. The above systems are briefly presented in the following section.

Aerobic wetland systems

Aerobic wetland systems are effectively employed for the treatment of net alkaline waters. In these systems, oxidation reactions occur and metals precipitate primarily as hydroxides, oxyhydroxides and oxides. The central role of the aerobic wetland cells is to allow for the oxidation of ferrous iron in the aerated water and the precipitation of the resulting ferric hydroxide floc out of suspension. As a result of the oxidation process acidity is generated which in turn retards the oxidation rate. The acidity produced in the Fe hydrolysis reaction is neutralised if bicarbonate alkalinity is available and pH is maintained between 5.5 and 6.5, enhancing the precipitation of Fe and other contained metals such as Mn.

Aerobic wetland systems are similar to “natural” wetlands. In Figure 6 a diagram of a typical aerobic, free surface flow, wetland system is given. An aerobic wetland consists of basins or channels, with relatively impermeable bottom to prevent seepage, soil or another suitable medium to support the vegetation, and water at a relatively shallow depth (10–50 cm) flowing over the soil surface. The shallow water depth, low-flow velocity, and presence of the plant stalks and litter, regulate water flow mainly through long, narrow channels that minimise short circuiting. Typically, the length to width ratio in an aerobic cell is above 10. The presence of vegetation in the cells enhances bacterial activity, removal of iron flocs and reduces the erosion potential of the precipitates.

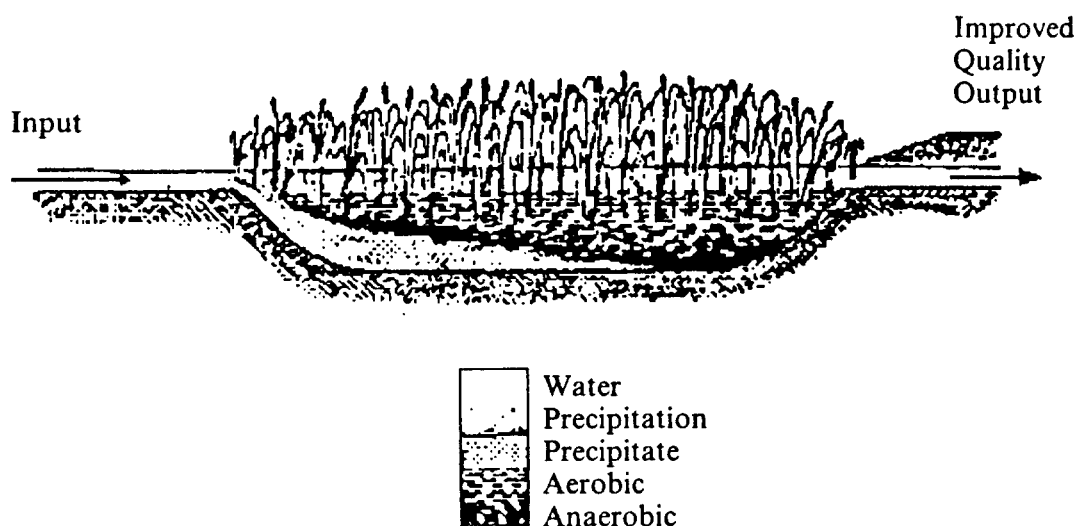


Fig.6 Diagram of a typical aerobic wetland [31].

Anaerobic organic substrate systems (or Compost wetlands)

Compost wetlands is one of the systems currently available for the treatment of net acidic mine waters. In these systems alkalinity is generated which in turn neutralises the acidity contained in the influent waters. Anaerobic wetlands are applicable to the treatment of mine waters containing high levels of dissolved oxygen (DO), Fe^{3+} or Al, and acidity higher than 300 CaCO_3 mg/L [3, 49]. Compost wetlands generate alkalinity through a combination of bacterial activity and limestone dissolution. The sulphate-reducing bacteria require a rich organic substrate in which anoxic conditions will develop; limestone dissolution also occurs within this anoxic environment.

Typical materials that are used as substrates in these wetlands include low-cost natural products and wastes such as spent mushroom compost, horse and cow manure, haybales, peat, wood chips and saw dust. Spent mushroom is a commonly used substrate also containing limestone (10% dry weight). Compost substrates with low CaCO_3 content may be supplemented with limestone. For wetland construction, substrate is generally placed in a non compacted layer, 30–45 cm thick [3]. The propagation of plants on the surface of anaerobic cells should be discouraged so as to avoid the penetration of the cell surface by the plant roots and the subsequent injection of excess oxygen into the substrate adversely affecting the performance of the sulphate reducing bacteria.

The water flow should be primarily directed within the substrate. A diagram of an anaerobic subsurface flow wetland system, where the water flows laterally through the substrate is given in Figure 7.

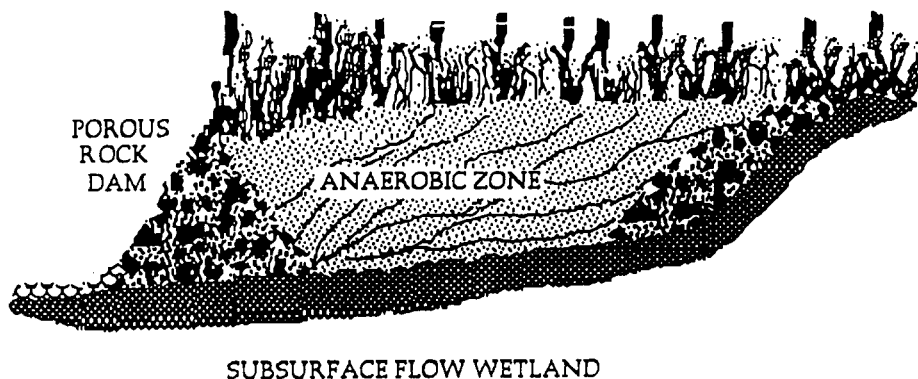


Fig.7 A diagram of a typical anaerobic system [31].

Anoxic limestone drains (ALDs)

Another method for adding alkalinity in an acidic mine water is the pretreatment with an anoxic limestone drain. ALDs produce alkalinity at a lower cost than do compost wetlands. However, according to recent studies this technique is not applicable to all waters since the mine waters considered suitable for ALD should have $\text{Fe}^{3+} < 2 \text{ mg/L}$, net acidity lower than $300 \text{ CaCO}_3 \text{ mg/L}$ and dissolved oxygen less than 1 mg/L [3, 49, 50].

The primary chemical factors that limit the performance of ALDs are the presence of ferric iron (Fe^{3+}), aluminium (Al) and dissolved oxygen (DO). When acidic waters containing $\text{Fe}^{3+} (> 2 \text{ mg/L})$ or Al ($> 2 \text{ mg/L}$) contact limestone, $\text{Fe}(\text{OH})_3$ or $\text{Al}(\text{OH})_3$ will form. Ferric hydroxide armour the limestone, limiting its further dissolution whereas gelatinous aluminium hydroxides plug the drain flow paths and eventually decrease the permeability of the limestone bed. The presence of dissolved oxygen ($\text{DO} > 1 \text{ mg/L}$) promotes the ferrous/ferric iron oxidation with the adverse consequence already mentioned.

Mine waters before exposure to the atmosphere are often very low in oxygen and dominated by ferrous rather than ferric iron. Diverting such a stream through an anoxic drain filled with limestone can add up to 400 mg/L of alkalinity to the water. ALDs are buried under several meters of clay; plastic is commonly placed between the limestone and clay as an additional gas barrier. Thus, ALD is sealed so that atmospheric oxygen inputs are minimised and the accumulation of CO_2 is enhanced. The ALD concept was first proposed by the Tennessee Department of Health and Environment for the treatment of coal mine waters, and is now being tested for the treatment of both coal and sulphide mine waters at sites throughout United States [7, 21, 47, 51].

The dimensions of existing ALD vary considerably. Initially, ALDs were constructed as long narrow ditches, approximately 1 m wide and sections of such an ALD are shown in Figure 8 [47]. At sites where the construction of linear ALD is not possible, anoxic limestone beds 10 to 20 m wide have been constructed and the alkalinities produced in these beds were similar to those of the more conventional drain systems [3].

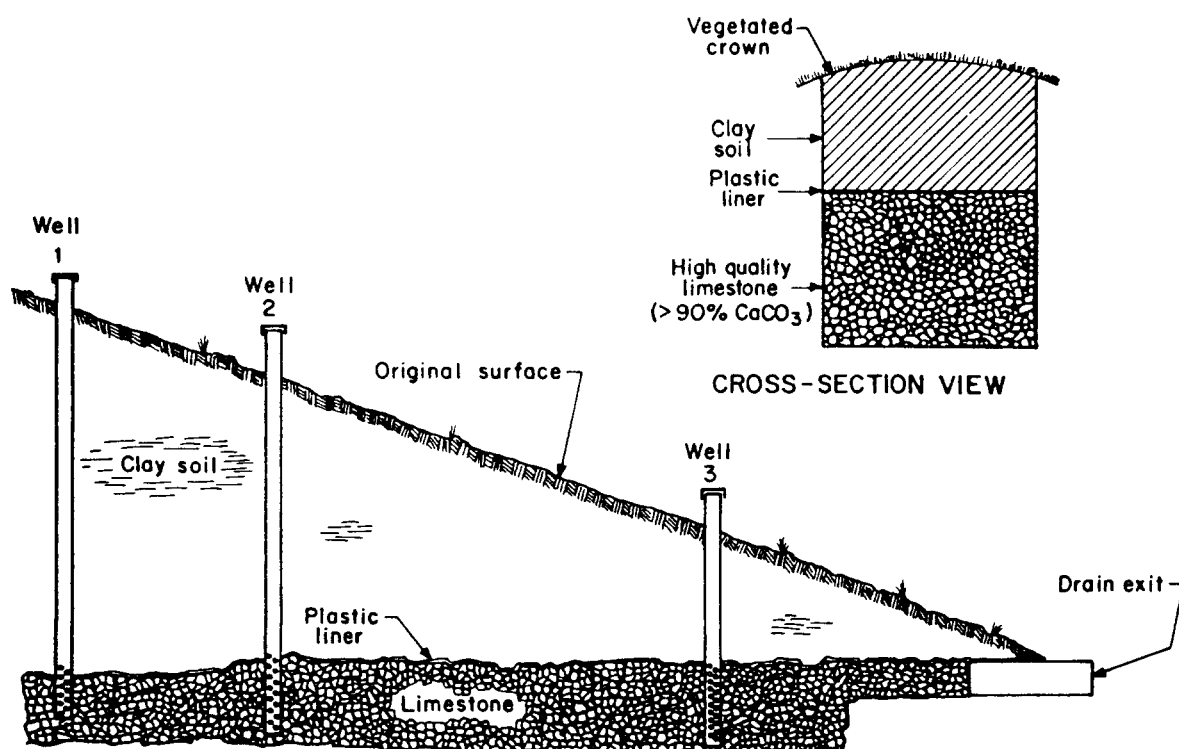


Fig.8 Longitudinal and cross section of the Morrison ALD [47].

The ALD is one component of the passive treatment system. When the ALD operates ideally, it results in raising the pH to near neutral levels and increase concentrations of calcium and alkalinity. Dissolved Fe^{2+} and Mn should be unaffected by flow through the ALD. Thus, in order to effectively remove the metal contaminants the ALD must be followed by an aerobic (settling basin or aerobic wetland) or anaerobic system, and the selection of the next stage depends on the net alkalinity of the ALD effluent, as well as the type and concentrations of the metals present.

In conclusion, each of these three passive technologies is applicable to a specific type of mine water treatment scheme and they are often most effectively employed in combination with each other. A model for the design of passive treatment systems based on the mine drainage chemistry is presented in Figure 9; this model was developed by the USBM for the treatment of acidic mine waters where the iron was the major contaminant. The design of the system becomes more complicated if other heavy metals such as manganese, aluminium, copper, lead, zinc, arsenic and mercury are present [7].

Currently a new type of passive treatment technology is under development called rock filters. It consists of cells that are aerobic and contain granite rocks which provide a large surface area on which algae grow. These systems are used downstream of compost wetlands and they generate a suitable environment with a high pH which allows for the quantitative removal of heavy metals such as Mn, not effectively stripped from solutions in the preceding stages.

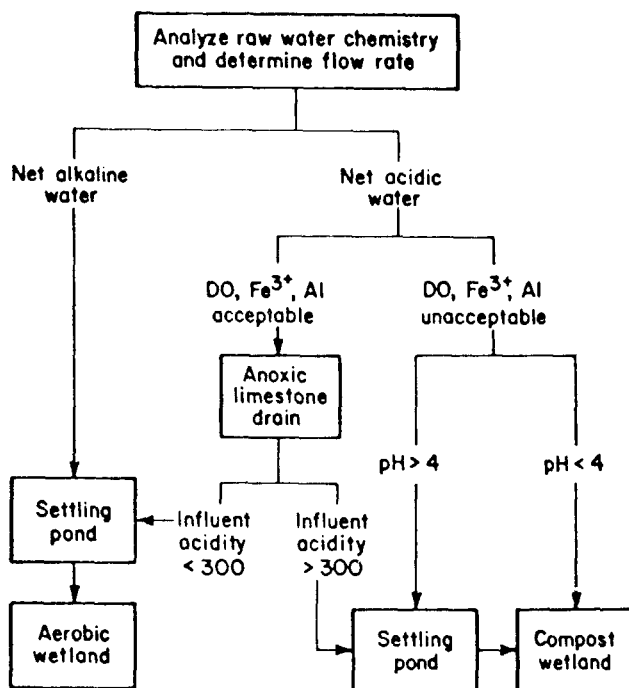


Fig.9 Flow chart showing chemical determinations necessary for the design of passive treatment systems [7].

Another new component of the passive treatment schemes is the anoxic ponds. These are anaerobic cells with organic substrate, similar to the compost wetlands, which are used upstream of ALDs aiming at decreasing dissolved oxygen and/or reducing Fe^{3+} to Fe^{2+} and removing Al.

PASSIVE TREATMENT SYSTEMS PERFORMANCE

To illustrate the potential applications and effectiveness of passive systems, characteristic examples regarding their performance for the treatment of coal and sulphide metal mine drainage are presented in Tables 2 and 3. In these Tables the design characteristics, the mean influent flow rates and the average composition of the influent and effluent streams are summarised.

TABLE 2 Characteristic applications of passive treatment systems on coal mine drainage [3,20,22,23,52]

Site, Constructed year	Design	Substrate	Vegetation	Surface area m ²	Flow rate m ³ /day	Composition					
						pH		Fe, ppm		Mn, ppm	
						In*	Ef*	In	Ef	In	Ef
<i>Donegal, 1987</i>	Pond, 8 Cells	Limestone, Spent mushroom compost	Typha	8,100	720	6.4	7.4	34	<1	9	2
<i>Cedar, 1989</i>	5 Cells	Clay, Limestone	Typha	1,360	225	6.3	6.4	92	41	2	2
<i>Keystone, 1989</i>	Ditch	Topsoil	None	4,200	12,400	6.3	6.4	37	32	<1	<1
<i>Blair, 1989</i>	Ditch	Manure, Straw	Mixed	1,080	16	6.2	7.0	52	<1	30	5
<i>Shade, 1989</i>	ALD	Limestone	None	880	15	6.0	6.8	2	<1	23	10
<i>Morrison, 1990</i>	ALD, 3 Cells	Clay, Manure	Typha	1,075	10	6.3	6.6	151	<1	42	11
<i>Emlenton, 1987</i>	9 Cells	Limestone, Manure	Typha	643	80	4.7	3.2	89	15	77	73
<i>Somerset, 1984</i>	2 Cells	Haybales, Limestone, Spent mushroom compost	Typha	1,005	70	4.4	5.5	162	18	50	33
<i>Latrobe, 1987</i>	3 Cells	Haybales, Limestone, Spent mushroom compost	Typha	2,800	124	3.5	3.7	125	56	32	29
<i>REM, 1992</i>	2 ALDs, 9 Cells	Spent mushroom compost	Typha	4,849	300	3.5	2.9	246	115	92	88
<i>Montana, 1985</i>	1 Cell, Limest. channel	Limestone-soil mix, Peat	Typha, Sedges	111	43	3.1	2.8	149	94	1	0
<i>Small Tracy Wetland</i>	1 Cell, Limest. channel	Limestone-soil mix, Peat	Typha, Sedges	418	82	2.7	2.6	284	271	2	2
<i>Large Tracy Wetland</i>											
<i>Ohio, Tuscarawas, 1986</i>	Ditch, Cell	Limestone, Peat, Compost, Sandy, Soil	Typha	200	55	5.5	-	31	8	34	31
<i>Ohio, Sinco #4, 1985</i>	3 Cells	Limestone, Compost	Typha	3,000	654	6.4	6.2	100	19	2	2

* In: Inflow / Ef: Effluent

TABLE 3 Applications of passive treatment systems on sulphide metal mine drainage [7,53]

Site, Constructed year	Design	Substrate	Flow rate m ³ /day	pH		Fe, ppm		Zn, ppm		Cu, ppm		Other components	
				In*	Ef*	In	Ef	In	Ef	In	Ef	In	Ef
<i>Leviathan mine, California, USA, 1993</i>	3 cells: ALD, aerobic, anaerobic	Horse manure	5.5	4.7	6.5	310	31					Al: 48 As: 0.4 Ni: 1.8	Al: <BDL As: <BDL Ni: <BDL
<i>Seeps, Gold mine, Nevada, USA, 1993</i>	14 chambered aerobic system with algae	River rock with algae	55	6-8	6-9	0.05	0.03	1.0	0.08	1.8	0.63	As: 0.12 Hg: 0.0008 CN: 31	As: 0.054 Hg: <0.0002 CN: 1.2-6.7
<i>ARD, Gold mine, Nevada, USA, 1993</i>	3 cells: aerobic pond, anaerobic, rock filter	Cow manure, Sandy soil	33	3.2	6.5-7.1	216	0.9	1.8	<BDL	7.4	<BDL	Mn: 7.1 As: 2.7 Se: 0.3	Mn: 2.9 As: 0.032 Se: 0.005
<i>Pit Cell, Brewer Gold mine, South California, USA, 1993</i>	1 anaerobic cell	Turkey litter, Sawdust Cow manure, Limestone	5.5	2.3	5.5	735	215			76	16	Al: 113	Al: 30
<i>Heap Leach Pad Cell, Brewer Gold mine, South California, USA, 1993</i>	1 anaerobic cell	Turkey litter, Sawdust Cow manure, Limestone	4.2	2.4-4.7	6.2	25-380	44			2-30	0.25	Al: 31	Al: 12.4
<i>Burleigh Tunnel, Colorado, USA, 1993</i>	2 anaerobic cells: 1 Upflow & 1 Downflow	Composted manure, Hay	55	6.8		5	<BDL	40	Upf: 0.35 Downf: 10				
<i>Asarco West Fork Unit, Lead mine, Missouri, USA, 1994</i>	1 anaerobic cell	Cow manure, Sawdust, Hay, Dolomitic tailings, Coarse mine waste	267	7.9	7.0	0		0.18	<0.008			Pb: 0.4	Pb: <0.02

* In: Inflow / Ef: Effluent

Regarding the treatment of coal mine drainage it is noted that the substrates used vary between minerals, such as clay and limestone rocks, to organic substances such as spent mushroom compost, manure, and hay bales. Cattails, *Typha latifolia* and less commonly *T. angustifolia*, are the emergent plants commonly growing in the cells. The systems range in size from 111 to 8,100 m² and most of them consist of a number of cells or ponds connected in series. The mean influent flow rates of mine drainage at the sites range from 10 to 12,400 m³/day; significant variations are also observed in the influent chemistry. In all systems examined iron levels are effectively reduced; discharges often contained less than 1 mg/L. The effectiveness of Mn removal is less pronounced than that of Fe.

Regarding the treatment of sulphide mine drainage it is noted that all the applications listed in Table 3 are pilot scale systems. Preliminary treatment results presented in Table 3 show that in addition to Fe removal, significant reductions in the concentrations of other contaminants such as As, Ni, Al, Cu, Zn Pb and CN can be also achieved. The first large scale passive system for the treatment of metal mine drainage is planned to be constructed by Asarco within 1995 for its West Fork lead sulphide mine in Missouri, USA. This full scale system includes: settling basin, anaerobic cell, rock filter and aeration basin and is intended to treat 8,200 m³/day [7].

Recently, a pilot passive system has been developed and constructed in Cornwall, England by the National Rivers Authority (NRA) for the treatment of the highly contaminated Wheal Jane tin mine water. This water with an average flow rate of 12,000 m³/day, is highly acidic, pH 2.5–3, and loaded with significant quantities of dissolved heavy metals such as Cd, Zn, As and Fe, with a total metal concentration in excess of 5,000 ppm. This treatment system involves a series of shallow lined cells providing a combination of pretreatment cells, aerobic and anaerobic environments. Three alternative treatment schemes will be tested the difference between them being the pre-treatment methods used [54].

SUMMARY AND CONCLUSIONS

Passive treatment of acid mine drainage represents an alternative to conventional neutralisation techniques with alkaline reagents such as lime, limestone, magnesia and others. Passive systems typically require less input for energy, reagents and manpower and present lower operation and maintenance costs than conventional treatment schemes. Thus, they may be employed during the mine operation as well as in the post-closure period.

Three principal types of passive technologies have been developed for the treatment of AMD: the aerobic wetlands, the compost wetlands, and the anoxic limestone drains.

- In aerobic wetlands, oxidation reactions occur and metals precipitate as hydroxides and oxyhydroxides.
- Compost wetlands promote anaerobic bacterial activity resulting in sulphate reduction and the subsequent precipitation of metal sulphides and the generation of alkalinity.
- Anoxic limestone drains generate alkalinity and may be used for the pretreatment of the acidic mine waters.

The applicability of each of the three passive technologies mainly depends on the composition of the mine waters under treatment. Often, they are most effectively used in combination especially when low pH, metal laden acidic mine drainage solutions have to be treated. Based on field observations, it is recommended that anoxic limestone drains should be staged first as a pretreatment stage aiming to add alkalinity to the waters, followed by aerobic wetland to remove as much iron as possible through oxidation. Directing the partially treated flow through a series of anaerobic cells with composted organic substrate will result in raising the pH to near neutral and in reducing the other contained metal levels to values close to environmental permissible limits.

These design recommendations represent current methodologies that will be potentially replaced with more efficient techniques presently under development. Research is underway testing successive alkalinity-producing systems (SAPS). These experimental systems combine ALD technology with sulphate reduction mechanisms; they promote vertical flow through rich organic substrates into limestone beds beneath the organic compost. The limitations that the presence of dissolved oxygen and ferric iron exert on ALD application is eliminated in SAPS; ferric iron is reduced to the ferrous form and dissolved oxygen is removed from the water within the organic substrate prior to its treatment through the limestone bed. Applications of these systems have recently been reported with encouraging results [55].

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