The Extractive Industries and Society 6 (2019) 241–249

Contents lists available at ScienceDirect

The Extractive Industries and Society

journal homepage: www.elsevier.com/locate/exis

Review article

# Acid mine drainage formation, control and treatment: Approaches and strategies

Jeffrey G. Skousen*, Paul F. Ziemkiewicz, Louis M. McDonald

*West Virginia University, Morgantown, WV 26506-6108, United States*

ARTICLE INFO

*Keywords:*
Acid-base accounting
Acid-producing material
Alkaline leach bed
Anoxic limestone drains
Chemical treatment
Open limestone channels
Passive treatment
Wetlands

ABSTRACT

Acid mine drainage (AMD) occurs after mining exposes metal sulfides to oxidizing conditions. Leaching of reaction products into surface waters pollute over 20,000 km of streams in the USA alone. The coal mine permitting process requires prediction of AMD potential via overburden analysis. Where a potential exists, AMD control measures including spoil handling plans, alkaline amendment, and oxygen barriers or water covers may be required to stop or hinder AMD generation. Other AMD control technologies include injection of alkaline materials (coal ashes and limestone products) into abandoned underground mines and into buried acid material in mine backfills, remining of abandoned areas, and installation of alkaline recharge trenches. Where AMD already exists, effluent treatment is required. Active treatment includes adding alkaline chemicals such as $Ca(OH)_2$, CaO, NaOH, $Na_2CO_3$, and $NH_3$, but chemical treatment is costly, requires dispensing equipment and facilities, and often extends for decades. Passive treatment systems may also be employed to treat problem drainages and are effective under certain flow and acidity conditions. Such systems include aerobic and anaerobic wetlands, anoxic limestone drains, vertical flow wetlands, open limestone channels, and alkaline leach beds. This article discusses the process of AMD formation, preventative and control measures, and describes treatment methods for existing AMD discharges.

## 1. Introduction: acid mine drainage formation

Acid mine drainage (AMD) forms when sulfide minerals deep in the earth are exposed during mining, highway construction, and other large-scale excavations. Upon exposure to water and oxygen, most sulfide minerals oxidize to form sulfuric acid, metal ions and sulfate, which, if untreated, enter surface or groundwater (Akcil and Koldas, 2006). The pH of AMD may range from 2 to 8. Group II and the transition metals, particularly iron (Fe) and aluminum (Al), dominate the dissolved cations in AMD while sulfate and bicarbonate are the most common anions (Skousen et al., 1999). While AMD can be either acidic or alkaline, we use the term AMD throughout to refer to drainage waters from coal and metal mines containing significant concentrations of contaminants.

The process of pyrite oxidation, the role of bacteria and other factors, and its effects on water resources have been known for centuries (Komnitasas et al., 1995; Nordstrom, 2011; Seal and Shanks, 2008; Stumm and Morgan, 1970) and AMD is a worldwide concern (Younger and Wolkersdorfer, 2004). Damaging effects of AMD have been describedby researchers in Asia (David, 2003; Wei et al., 2013; Yang et al., 2007), New Zealand (Trumm and Ball, 2014; Winterbourn et al., 2000), Europe (Antivachis et al., 2016; Balci and Demirel, 2018; Casiot et al., 2009; Gray and Delaney, 2008), South America (Strosnider et al., 2011a,b), Canada (Neculita et al., 2007; Sracek et al., 2004), and the USA (Cherry et al., 2001; Kleinmann, 1989; Soucek et al., 2000). In the eastern USA alone, > 10,000 km of streams and > 72,000 ha of lakes and reservoirs were adversely affected by AMD prior to 1990 (Herlihy et al., 1990; Kleinmann, 1989). When AMD enters surface water bodies, the effects include biotic impacts on stream and lake organisms through direct toxicity, habitat alteration by metal precipitates, visual changes from orange or yellow staining of stream sediments, nutrient cycle disruptions, or other mechanisms, and the water often becomes unsuitable for domestic, agricultural, and industrial uses (DeNicola and Stapleton, 2002; Evans et al., 2015; Hogsden and Harding, 2012; Soucek et al., 2001). Laws and regulations have been in place since the 1970s to reduce or eliminate the formation and release of AMD into the environment (Environmental Protection Agency, 2017), but problems still exist on abandoned and un-reclaimed sites and sparingly on newly mined sites.

The largest contribution of AMD into streams and lakes is from the numerous abandoned mined lands that were mined and left without reclamation (Skousen et al., 2000). The sulfide-bearing materials were

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* Corresponding author.
*E-mail address:* jskousen@wvu.edu (J.G. Skousen).

https://doi.org/10.1016/j.exis.2018.09.008
Received 10 August 2018; Received in revised form 24 September 2018; Accepted 24 September 2018

left exposed on the surface and, without reclamation, they continue to generate AMD, thereby releasing the metal ions and sulfuric acid into the environment. To deal with these problem discharges, reclamation and treatment have been employed, which over the past 40 years has greatly reduced the quantity of AMD into streams (Faulkner and Skousen, 1995; Skousen et al., 2017). Many AMD treatment methods are available and the costs for implementing the treatment systems are generally high. Plus, many factors determine the most efficient treatment method: AMD quality and quantity, formation and settling of metal hydroxides, receiving stream uses, and available resources.

The acidity level, metal composition and concentrations of a given AMD source depend on the type and quantity of sulfide minerals present (acid-producing) and acid-neutralizing (carbonate) minerals contained in the exposed rock (Perry and Brady, 1995; Skousen, 2017). The carbonate content of overburden determines whether there is enough neutralization potential or base to counteract the acid produced from sulfide oxidation. Of the many types of acid-neutralizing compounds present in rocks, only carbonates (and some clays) occur in sufficient quantity to effectively neutralize acid-producing rocks. A balance between the acid-producing potential and acid-neutralizing capacity of the disturbed overburden will indicate the ultimate acidity or alkalinity that might be expected from water draining the disturbed material upon complete weathering. The reaction rates or kinetics of these materials are also important. For example, sulfide oxidation tends to occur quickly while limestone dissolution is slower and requires acid conditions for rapid reaction.

## 2. Overburden analyses, handling and placement

Control of AMD before land disturbance requires an understanding of three important factors: 1) overburden or mine waste geochemistry, 2) method and precision of overburden handling and placement in the backfill or waste pile during the operation and reclamation, and 3) the post-mining hydrology of the site. Pre-mining analysis of soils and overburden are required by law (Sobek et al., 2000). Identifying the acid-producing or acid-neutralizing status of rock layers before disturbance (Modis and Komnitsas, 2007) aids in developing overburden handling and placement plans. The most widely-used method to determine AMD potential is Acid-Base Accounting (Skousen et al., 1987). Acid-Base Accounting provides a simple, relatively inexpensive, and consistent procedure for evaluating overburden chemistry (Skousen, 2017). Rock samples from the overburden layers to be disturbed are ground and subjected to analysis to determine their chemistry. The procedure balances the maximum potential acidity or MPA (based on total or pyritic sulfur content) against the mass of neutralizing potential (NP). Samples containing more acid-producing than acid-neutralizing materials are “deficient” and can cause AMD, while those rock samples with the reverse situation have "excess" neutralizing materials and will not cause AMD. Because blending of alkaline and acid-forming rock during the mining and reclamation process is never perfect in field settings, a safety factor stipulating an excess of alkalinity is generally applied. An NP:MPA ratio of 2:1 is considered prudent (Skousen et al., 2002). Rock layers with equal proportions of each type of material should be subjected to leaching or weathering analyses to more accurately access the chemical production potential (Hornberger and Brady, 1998). Kinetic tests such as humidity cells and leach columns are important because they examine the rate of acid-producing and neutralization reactions (Geidel et al., 2000). This information from kinetic tests can supplement information given by Acid-Base Accounting and help regulators in permitting decisions. For example, the rate of sulfide oxidation and release of contaminants may occur quickly before neutralizing materials have time to react. Therefore, early pulses of pollutants may be released for a short time as the reactions are balanced.

In situations where a positive ratio of alkaline- to acid-forming materials cannot be achieved, alkaline amendments can be imported to achieve the desired NP:MPA ratio of 2:1. Otherwise, strategies such as caps and covers that would isolate pyritic materials from air and water are employed. Once overburden materials have been classified and their mass ratios have been determined, an overburden handling and placement plan for the site can be designed. Segregating and placing acid-producing materials above the water table in the backfill is generally recommended (Skousen, 2017; Ziemkiewicz and Skousen, 1998; Skousen et al., 1987). Where alkaline materials predominate and are effectively blended during spoil placement, no additional controls are generally necessary (Ziemkiewicz and Meek, 1994).

## 3. Post-mining hydrology

The hydrology of a backfill and the flow paths of water through unconsolidated rocks are complex. Generally, the porosity and hydraulic conductivity of the materials in a backfill are greater than those of the consolidated overburden that existed before mining, and changes in flow patterns and rates should be expected after mining (Caruccio and Geidel, 1989; Hawkins, 2005). Water does not develop a uniform wetting front in the backfill, but tends to follow channels and open voids. As water moves into coarse materials in the backfill, it follows the path of least resistance and continues downward through voids or conduits until it encounters a barrier or other compacted layer (generally the underlying intact rock). Therefore, the chemistry of the water from a backfill or waste pile will reflect only the materials encountered in the water flow path, and will not reflect the chemistry of all the materials in the overburden or waste pile (Ziemkiewicz and Skousen, 1992).

Diverting surface water above the site to decrease the amount of water entering the mined area is highly recommended. If it cannot be diverted, incoming water can flow through limestone to be pre-treated and increase the alkalinity of the water (Trumm et al., 2006). Under certain conditions, sulfide materials can be placed where it will be rapidly and permanently inundated, thereby preventing oxidation. Inundation is only suggested where a water table may be re-established such as below drainage deep mines (see Water Covers).

## 4. Control of AMD

Acid mine drainage control can be undertaken where AMD exists or is anticipated. Control methods treat the acid-producing rock directly and stop or retard the production of acidity. Treatment methods add chemicals directly to acidic water exiting the rock mass. Companies disturbing land in acid-producing areas must often treat AMD, and they face the prospect of long-term water treatment and its liabilities and expense. Therefore, cost-effective methods, which prevent the formation of AMD at its source, are preferable.

In March 1997, the U.S. Department of Interior, Office of Surface Mining Control and Reclamation (OSM) issued policy guidance to state programs on the correction, prevention and control of acid/toxic mine drainage. Known as the AMD Policy of 1997, it states “In no case should a permit be approved if the determination of probable hydrologic consequences or other reliable hydrologic analysis predicts the formation of a post-mining pollutional discharge that would require continuing long-term treatment without a defined endpoint.” The policy encouraged the development of AMD prevention and control methods. Some of these control methods are best suited for abandoned mines, while others are only practical on active operations. Other methods can be used in either setting.

### *4.1. Land reclamation*

Backfilling (regrading the land back to contour) and revegetation together can be effective methods of reducing acid loads from disturbed lands (Faulkner and Skousen, 1995). Water flow from seeps can be reduced by diversion and reclamation, and on some sites where flow may not be reduced, water quality can change from acid to alkaline by proper handling of overburden. Recharge to deep mines occurs when subsidence creates channels to underground workings. These can be mitigated by sealing the stream bed or cracks at the surface (Foreman, 1998). Surface diversion involves construction of drainage ditches to move surface water quickly off reclaimed surface mines by providing impervious channels to convey water across the disturbed area.

### *4.2. Alkaline amendment to active disturbances*

Certain alkaline amendments can control AMD from acid-producing materials (Brady et al., 1990; Perry and Brady, 1995; Rich and Hutchison, 1990; Rose et al., 1995). All alkaline amendment schemes rely on Acid-Base Accounting or kinetic tests to identify the required mass of alkalinity for neutralization of acidic materials. Special handling of overburden seeks to blend or co-mingling acid-producing and acid-neutralizing rocks in the disturbance/reclamation process to develop a neutral material. The pit floor or material under the coal is often rich in pyrite, so isolating it from groundwater may be necessary by building highwall drains (which move incoming groundwater away from the pit floor) or placing impermeable barriers on the pit floor (Skousen et al., 2012). Acid-forming material can also be compacted or capped within the spoil (Meek, 1994).

If insufficient alkalinity is available in the spoil, then external sources of alkalinity must be imported (Skousen and Larew, 1994; Wiram and Naumann, 1995). Limestone is often the least expensive and most readily available source of alkalinity. It has a neutralization potential of between 75 and 100%, and is safe and easy to handle. On the other hand, it has no cementing properties and cannot be used as a barrier. Fluidized Bed Combustion ashes generally have neutralizing amounts of between 20 and 40%, and they tend to harden into cement after wetting (Gray et al., 1998; Skousen et al., 1997). Other power-generation ashes, like flue gas desulfurization products and scrubber sludges, may also have significant neutralization potential, which make them suitable alkaline amendment materials (Stehouwer et al., 1995). However, scrubber sludges may contain sufficient selenium to affect discharge water quality. Other materials, like kiln dust, produced by lime and cement kilns, or lime muds, grit, and dregs from pulp and paper industries contain neutralization products (Rich and Hutchison, 1994). Steel slags, when fresh, have neutralizing amounts from 45 to 90%. Slags are produced by a number of steel-making processes, so care is needed to ensure that candidate slags are not prone to leaching metal ions like Cr, Mn, and Ni. Phosphate rock has been used in some studies to control AMD. It may react with Fe released during pyrite oxidation to form insoluble coatings (Evangelou, 1995), but phosphate usually costs much more than other calcium-based amendments and is needed in about the same amounts (Ziemkiewicz and Meek, 1994). When any of the waste materials are imported and co-mingled as sources of alkalinity, they could release elements of concern during weathering and should be tested before use (Tiwari et al., 2015). Costs of acquiring and transporting alkaline materials can be expensive, and therefore the economics must be favorable for this technique to be viable (Skousen and Larew, 1994).

### *4.3. Alkaline recharge trenches*

Alkaline recharge trenches (Caruccio et al., 1984) are surface ditches or cells filled with alkaline material, which can minimize or eliminate acid seeps through an alkaline-loading process with infiltrating water. They can be installed on backfills or waste piles during reclamation to add alkalinity to water before infiltration. Alkaline recharge trenches were constructed on top of an 8-ha, acid-producing coal refuse disposal site, and after three years the drainage water showed 50–90% acidity reductions with 70–95% reductions in Fe and sulfate (Nawrot et al., 1994). Pumping water into alkaline trenches greatly accelerates the movement of alkalinity into the backfill and can cause acid seeps to turn alkaline (Ziemkiewicz et al., 2000).

### *4.4. Oxygen barriers*

Oxygen barriers retard the movement of water or oxygen into areas containing acid-producing rock (Argunhan-Atalay and Yazicigil, 2018). These “water control” technologies (Ziemkiewicz and Skousen, 1998) include impervious membranes, dry seals, hydraulic mine seals, and grout curtains/walls. Many designs of oxygen barriers have been tested and applied (Aubertin et al., 2016; Nicholson et al., 1988). Surface barriers can achieve substantial reductions in water flow through piles, but generally do not control AMD completely (Sahoo et al., 2013). Grouts can be used to separate acid-producing rock and groundwater. Injection of grout barriers or curtains may significantly reduce the volume of groundwater moving through backfills. Gabr et al. (1994) found that a 1.5-m-thick grout wall (installed by pumping a mixture of Class F fly ash and Portland cement grout into vertical boreholes near the highwall) reduced groundwater inflow from the highwall to the backfill by 80%, resulting in some seeps drying up and others being substantially reduced in flow. A combination of pneumatically-placed barriers and cementitious fluidized-bed combustion ash reduced discharge from an underground mine from $1000\,\mathrm{L\,min^{-1}}$ (250 gpm) to $< 40\,\mathrm{L\,min^{-1}}$ (10 gpm) (Ziemkiewicz and Head, 1993). At the Heath Steele Metal Mine in New Brunswick, a soil cover was designed to exclude oxygen and water from a tailings pile (Bell et al., 1994). It consisted of a 10-cm gravel layer for erosion control, 30-cm gravel/sand layer as an evaporation barrier, 60-cm compacted till (conductivity of $10^{-6}$ cm/sec), 30-cm sand, and pyritic waste rock. This barrier excluded 98% of precipitation, and oxygen concentrations in the waste rock dropped from 20% initially to around 1%. At the Upshur Mining Complex in West Virginia, Meek (1994) reported covering a 20-ha spoil pile with a 39-mil PVC liner, and this treatment reduced acid loads by 70%.

### *4.5. Water covers*

Disposal of sulfide tailings under a water cover, such as in a lake or fjord, is another way to prevent acid generation by excluding oxygen from sulfides. Disposal of mine wastes into the ocean or lakes was done in the past, but most countries have banned this practice (Edinger, 2012), although still done sparingly in a few countries (Hughes et al., 2015). Water covers also include flooding of aboveground tailings in ponds (Anonymous, 2013). Fraser and Robertson (1994) studied four freshwater lakes used for subaqueous tailings disposal and found that the reactivity of tailings under water was low and that there were low concentrations of dissolved metals, thereby allowing biological communities to exist.

### *4.6. Alkaline amendment to abandoned mines*

Abandoned surface mines comprise huge volumes of spoil of unknown composition and hydrology. Abandoned underground mines are problematic because they are often partially caved and flooded, cannot be accessed, and have unreliable or nonexistent mine maps. Re-handling and mixing alkalinity into an already reclaimed backfill is generally prohibitively expensive.

Filling abandoned underground mine voids with non-permeable materials is one of the best methods to prevent AMD (Shen et al., 2017). Underground mine voids are extensive (a 60-ha mine with a coal bed height of 1.5 m and a recovery rate of 65% would contain about 600,000 $\mathrm{m^3}$ of voids), so fill material and the placement method must be cheap, readily-available, and have low transportation and pumping costs. Mixtures of Class F fly ash and 3–5% Portland cement are used to control subsidence in mined-under residential areas and these slurries are generally injected through vertical boreholes at between 8- and 16-m centers. Pneumatic (air pressure) and slurry injection for placing fly ash in abandoned underground mines can extend the borehole spacing to about 30 m (Burnett et al., 1995). On reclaimed surface mines still producing AMD, researchers in Pennsylvania saw small improvements in water quality after injecting coal combustion residues into buried pods of pyritic materials.

### 4.7. Remining and reclamation

Remining means returning to abandoned surface or underground mines for further coal removal. Where AMD occurs, remining reduces acid loads by 1) decreasing recharge, 2) covering acid-producing materials, and 3) removing the remaining coal, which is the source of most of the pyrite. Hawkins (1994) found contaminant loads of 57 discharges from remined sites in Pennsylvania to be reduced after remining and reclamation. Short-term loads sometimes increased during the first six months after remining and reclamation, but acid load reductions occurred after six months due to decreased flow rather than large changes in concentrations. Ten remining sites in Pennsylvania and West Virginia were reclaimed to current standards (which included eliminating highwalls, covering refuse, and revegetating the entire area), resulting in improved water quality in every case (Skousen et al., 1997).

## 5. Chemical treatment of AMD

If AMD problems develop during mining or after reclamation, a plan to treat the discharge must be developed. A simple water treatment system can consist of an inflow pipe or ditch, a storage tank or bin holding the treatment chemical, a valve to control its application rate, a settling pond to capture precipitated metal oxyhydroxides, and a discharge point. At the discharge point, water samples are analyzed to monitor whether specified parameters are being attained. More sophisticated systems add pumping stations, aeration equipment, electronic chemical dispensing systems, clarifiers, and continual automated discharge monitoring. Water discharge permits (as part of the US EPA's NPDES program in the Clean Water Act) on surface mines usually require monitoring of pH, total suspended solids, and Fe and Mn concentrations. The type and size of a chemical treatment system is based on flow rate, pH, water oxidation status, and concentrations of metals in the AMD. The receiving stream's designated use and seasonal fluctuations in flow rate are also important. After evaluating these variables over a year to monitor seasonal changes, the design, treatment capacity, and economics of the system can be evaluated using different chemicals and treatment options.

Six chemicals are commonly used to treat AMD (Table 1). Each is more or less appropriate for a specific condition (Trumm, 2010). The best choice depends on both technical (acidity levels, flow, types and concentrations of metals, receiving stream characteristics) and economic factors (chemical prices, labor, machinery and equipment, treatment duration, and interest rates) (Skousen et al., 1999). Enough alkalinity must be added to raise pH to between 6 and 9 so insoluble metal hydroxides will form and settle out in treatment ponds. Treatment of AMD with high Fe (ferric) concentrations often affords co-precipitation of other metals with the Fe hydroxide, thereby removing them from AMD at a lower pH.

### 5.1. Limestone

Limestone has been used for decades to raise pH and precipitate metals in AMD. It has the lowest material cost and is the safest and easiest to handle of the AMD chemicals. It's use in AMD treatment, however, is limited due to its low solubility and tendency to develop an external coating, or armor, of $Fe(OH)_3$ when added to AMD. Fine-ground limestone (sand-sized) may be dumped in streams directly (Menendez et al., 2000) or the limestone may be pulverized by water-powered rotating drums and metered into the stream (Zurbuch, 2011), both of which help to increase its dissolution and reaction with AMD. Limestone has also been used to treat AMD in anaerobic (anoxic limestone drains) and aerobic environments (open limestone channels) as will be discussed in passive treatment situations. Limestone is not used frequently in routine chemical treatment because it is slow to react and dissolve (Ziemkiewicz et al., 1997; Zurbuch, 2011), and other more reactive and effective liming materials are available.

### 5.2. Lime

Hydrated lime is commonly used for treating AMD (Othman et al., 2017). As a finely-ground powder, it tends to be hydrophobic, and extensive mechanical mixing is required for dissolution. Hydrated lime is particularly useful and cost effective in large-flow, high-acidity situations where a lime treatment plant with large storage bins and a mixer/aerator is employed to help dispense and mix the chemical with the water (Skousen and Ziemkiewicz, 1996). Hydrated lime has limited effectiveness if a very high pH ($>$ 10) is required to remove ions such as Mn. Increasing the lime rate to increase the pH has limited success and tends to only increase the volume of unreacted lime that enters the settling pond.

### 5.3. Quicklime

Pebble quicklime (CaO) is used with lime dosers and portable quicklime dispensing equipment machines, which may have controlled dispensers or utilize a water-wheel concept (Jenkins and Skousen, 2001). A water wheel is turned based on water flow, which causes a screw feeder to dispense the chemical. The water-wheel dispensing

**Table 1**
Six chemicals used in AMD treatment with associated conversion factors and neutralization efficiencies.

| Common Name | Chemical Name | Formula | Conversion Factor[a] | Neutralization Efficiency[b] | 2010 Cost[c] $ per Mg or L | |
|---|---|---|---|---|---|---|
| | | | | | Bulk | < Bulk |
| Limestone | Calcium carbonate | $CaCO_3$ | 1 | 30% | $11 | $16 |
| Hydrated Lime | Calcium hydroxide | $Ca(OH)_2$ | 0.74 | 90% | $66 | $110 |
| Pebble Quicklime | Calcium oxide | CaO | 0.56 | 90% | $88 | $264 |
| Soda Ash | Sodium carbonate | $Na_2CO_3$ | 1.06 | 60% | $220 | $350 |
| Caustic Soda | Sodium hydroxide | NaOH | 0.8 | 100% | $750 | $970 |
| 20% Liquid Caustic | Sodium hydroxide | NaOH | 784 | 100% | $0.06 | $0.16 |
| 50% Liquid Caustic | Sodium hydroxide | NaOH | 256 | 100% | $0.29 | $0.33 |
| Ammonia | Anhydrous ammonia | $NH_3$ | 0.34 | 100% | $330 | $750 |

[a] The conversion factor may be multiplied by the estimated Mg acid/yr to get Mg of chemical needed for neutralization per year. For liquid caustic, the conversion factor gives L needed for neutralization.

[b] Neutralization Efficiency estimates the relative effectiveness of the chemical in neutralizing AMD acidity. For example, if 100 Mg of acid/yr was the amount of acid to be neutralized, then it can be estimated that 82 Mg of hydrated lime would be needed to neutralize the acidity in the water (100(0.74)/0.90).

[c] Price of chemical depends on the quantity being delivered. Bulk means delivery of chemical in a large truck, whereas < Bulk means purchased in small quantities. Liquid caustic prices are for L. Others in Mg.

system was initially used for small and/or periodic flows of high acidity because CaO is very reactive, but more recently controlled automated dispensers have been attached to large silos for high-flow/high-acidity situations (Mills, 1996). Tests show an average of 75% cost savings using a portable water-wheel dispensing system over NaOH systems and about 20–40% savings over $NH_3$ systems.

### 5.4. *Soda ash*

Soda ash ($Na_2CO_3$) is generally used to treat AMD in remote areas with low flow ($< 750\,L\,min^{-1}$ or 200 gpm) and low amounts of acidity and metals (Skousen et al., 1999). The choice to use soda ash is usually based on convenience rather than chemical cost. Soda ash comes as solid briquettes, and is gravity fed into water through storage bins. The number of briquette bags used per day is determined by the rate of flow and the quality of the water. One problem is that the briquettes absorb moisture, then expand and stick to the corners of the bin, which hinders their contact with water. For short-term treatment, some operators use a much simpler system employing a wooden box or barrel with holes that allows water inflow, contact with the briquettes, and then outflow. The operator simply fills the barrel with briquettes on a regular basis and places the barrel in the flowing water. This simple system offers less control of the amount of chemical used. Soda ash is not used in flow situations of more than $750\,L\,min^{-1}$ (200 gpm) because of its high cost.

### 5.5. *Caustic soda*

Caustic soda (i.e., lye, NaOH) is often used in remote low-flow ($< 100\,L\,min^{-1}$ or $< 30$ gpm), high-acidity situations, or if Mn concentrations in the AMD are high (Skousen, 2014). The system can be gravity-fed by dripping liquid NaOH directly into the AMD. Caustic is very soluble, disperses rapidly, and raises the pH quickly. Caustic should be applied at the surface of ponds because the chemical is denser than water and moves downward when applied to water in ponds. The major drawbacks of using liquid NaOH for AMD treatment are high cost and dangers in handling.

### 5.6. *Ammonia*

Ammonia compounds ($NH_3$ or $NH_4OH$) are used to treat acid mine drainage but anhydrous ammonia is extremely hazardous. $NH_3$ is compressed and stored as a liquid but returns to the gaseous state when released from the tank. Ammonia is extremely soluble, since it forms a gas upon release from the tank, reacts rapidly, and can raise the pH of receiving water to 9.2 (Faulkner, 1990). At pH 9.2, it buffers the solution to further pH increases, and therefore very high excess amounts of $NH_3$ must be added to go beyond 9.2. Injection of $NH_3$ into AMD is one of the quickest ways to raise water pH and it should be injected near the bottom of the pond or water inlet because $NH_3$ is less dense than water. $NH_3$ is quite cost effective and a cost reduction of 50%–70% is usually realized when $NH_3$ is substituted for NaOH (Skousen et al., 1990). Major disadvantages of using $NH_3$ include 1) the hazards of gas release and human exposure, 2) uncertainty concerning nitrification, denitrification, and acidification reactions downstream, and 3) consequences of excessive application rates, which cause toxic conditions to aquatic life.

### 5.7. *Costs of treating AMD*

Costs for treating AMD are site and case specific. To demonstrate the range of costs, five treatment chemicals under four sets of flow and acid concentration conditions were used to estimate costs (Table 1 from Skousen et al., 2000). Each chemical was found to be cost-effective under different scenarios based on labor requirements, equipment and dispensing facilities, and cost. In essence, for low flows of $< 750\,L\,min^{-1}$ (200 gpm), NaOH, $Na_2CO_3$ and ammonia had the lowest costs because no expensive mechanical equipment was needed. Hydrated lime treatment systems had the highest installation costs of the five chemicals because of the need to construct a lime treatment plant and install a pond aerator. However, the cost of $Ca(OH)_2$ was very low, and the combination of high installation costs and low reagent cost made $Ca(OH)_2$ systems particularly appropriate for long-term treatment of high-flow/high-acidity conditions because the installation cost could be amortized over a long period.

### 5.8. *Floc and special chemicals*

After chemical treatment, the treated water flows into settling ponds so metals in the water can precipitate (Skousen and Ziemkiewicz, 1996). All AMD treatment chemicals cause the formation of metal hydroxide sludge or floc. Sufficient residence time of the water (dictated by water inflow, pond size and depth) is important for adequate metal precipitation. The amount of metal floc generated depends on water quality and quantity, which in turn determines how often the ponds must be cleaned. Knowing the chemical and AMD being treated will provide an estimate of the stability of metal compounds in the floc (McDonald and Webb, 2006; Winland et al., 1991). Floc disposal options include: 1) leaving it submerged indefinitely in the pond, 2) pumping or hauling it to abandoned deep mines or to pits dug on surface mines, and 3) dumping it into refuse piles. Pumping flocs onto land and letting them age and dry is a good strategy for disposal, because they become crystalline and behave like soil material.

Sometimes special chemicals are needed to speed up the reaction or to improve floc settling efficiency (Skousen, 2014). Chemicals such as coagulants and flocculants aid in metal hydroxide precipitation and can reduce the residence time and pond size needed for effective treatment. Other chemicals like hydrogen peroxide or potassium permanganate can be added to accelerate aeration, hydroxide formation, and more rapid settling. These special chemicals are usually expensive and laboratory testing should be conducted to determine the cost-benefit ratio of using these chemicals for treatment.

### 5.9. *Summary of AMD treatment with chemicals*

Each AMD is unique, requiring site-specific treatment. Each AMD source should be tested with various chemicals by titration tests to evaluate the most effective chemical for precipitation of the metals. The costs of each AMD treatment system based on neutralization (in terms of the reagent cost, capital investment and maintenance of the dispensing system) and floc disposal should be evaluated to determine the most efficient and cost-effective system.

## 6. Passive treatment of AMD

Active chemical treatment of AMD is often an expensive, long-term proposition. Passive treatment systems (Gazea et al., 1996; Hedin et al., 1994; Skousen et al., 2017) have been developed that do not require continuous chemical inputs and take advantage of natural chemical and biological processes to treat contaminated mine waters. Passive technologies include constructed wetlands, anoxic limestone drains, vertical flow wetlands, open limestone channels, and alkaline leach beds (Fig. 1 from Skousen et al., 2017). In low-flow and low-acidity situations, passive systems can be reliably implemented as a single permanent solution for many AMD problems. When either flow or acidity concentrations are moderate, passive systems may still be appropriate if properly designed and maintained (Zipper and Skousen, 2010).

### 6.1. *Constructed wetlands*

Constructed wetlands for AMD treatment can be either primarily aerobic or primarily anaerobic. Aerobic wetlands are shallow ponds ($< 30$ cm), with or without wetland plants, used primarily to slow

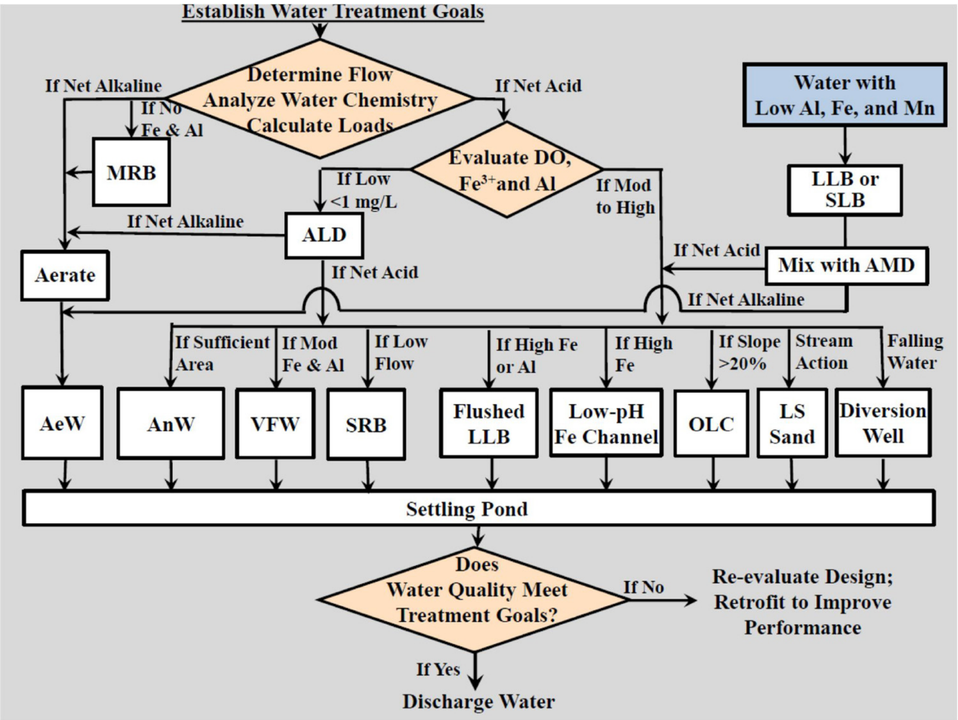

**Fig. 1.** Flow chart for selecting a passive AMD treatment system based on water flow and water chemistry (from Skousen et al., 2017).

down influent water to allow metal oxidation, hydrolysis, and particle settling. Flow-diverting baffles are often used to help reduce flow rates. Limestone can be used to facilitate neutralization but aerobic wetlands are typically used as the final 'polishing' treatment step before discharge to receiving streams (Mays and Edwards, 2001).

In contrast, anaerobic wetlands, sometimes called 'bioreactors' are relatively deep ( > 30 cm) with substrates of soil, peat moss, spent mushroom compost, sawdust, straw/manure, hay bales, or other organic carbon-based materials, often underlain or mixed with limestone. Water passes through the thick permeable organic subsurface layer and becomes anaerobic due to the high biochemical oxygen demand. The lack of oxygen promotes bacterial sulfate reduction to sulfides which then form insoluble metal precipitates (e.g. $FeS_2$). Microbial sulfate reduction also produces alkalinity leading to metal precipitation as oxyhydroxides (Neculita et al., 2007). Another possible metal removal mechanism from AMD is sorption onto organic materials. Metal removal by sorption is limited by the availability of appropriate functional groups in the substrate whereas metal removal by precipitation as sulfides or hydroxides is limited only by alkalinity generated (limestone dissolution or sulfate reduction), sulfate availability and a community of sulfate reducing bacteria. The predominant removal mechanism is hydroxide precipitation (Sheoran and Sheoran, 2006; Wieder, 1993), although this aspect of the system is less-well studied than for other aspects of AMD passive treatment systems. While typically best suited for low AMD flows, some treatment success has been achieved in > 750 L $min^{-1}$ ( > 200 gpm) flow situations (Canty, 2000; Gusek et al., 2000).

Sulfate-reducing bioreactors are modifications of anaerobic wetlands. They can still be constructed as ponds, but are often formed in large containers or tanks. Bioreactors generally are inoculated with specialized microbial populations that were designed for a particular water or may include specific substrates (Lefticariu et al., 2015; Neculita et al., 2007; Trumm and Ball, 2014). The mechanisms for treatment are similar to those of anaerobic wetlands but tend to rely more on microbial treatment for acid neutralization and metal removal (Gusek, 2002).

### 6.2. *Vertical flow wetlands*

In contrast to constructed anaerobic wetlands where AMD moves horizontally through the organic substrate, in vertical flow wetlands (VFW), a hydraulic head of AMD is forced down vertically through the organic substrate into a limestone bed with pipes to convey treated water to the next treatment stage. Otherwise, the mechanisms of treatment are identical. However, an advantage of VFWs is that when iron and aluminum precipitates clog the limestone bed and/or piping, the system can be flushed (Kepler and McCleary, 1997). VFWs are an active area of AMD treatment research, especially in the areas of accurate sizing, and flushing frequency and procedure (Rose, 2006; Skousen et al., 2017). Other common terms for VFWs are Successive Alkalinity Producing Systems (SAPS, Kepler and McCleary, 1994), Reducing and Alkalinity Producing Systems (RAPS, Watzlaf et al., 2000a) or vertical flow ponds.

### 6.3. *Anoxic limestone drains*

Anoxic limestone drains (ALD) generate alkalinity when water passes through a limestone bed in a semi-closed system, with as little dissolved oxygen as is possible (Turner and McCoy, 1990). When limestone is buried, covered and dissolves, the increase in $CO_2$ partial pressure leads to additional limestone dissolution and alkalinity. Treatment success depends on Fe being in the ferrous ($Fe^{2+}$) oxidation state because ferrous iron does not form a hydroxide precipitate in near neutral solutions. Any introduced or produced ferric iron ($Fe^{3+}$) will precipitate on the limestone surfaces and clog the system. Aluminum hydroxide and gypsum ($CaSO_4$) precipitates are also a concern. Although Hedin et al. (2010, 2013) evaluated an ALD that operated effectively for 18 years without maintenance, because of the difficulty in excluding oxygen and aluminum in most situations, eventual clogging is very likely (Nairn et al., 1991; Watzlaf et al., 2000b), and the need for treatment system replacement should be recognized.

### 6.4. Open limestone channels

Open limestone channels (OLC) generate alkalinity when AMD passes over large ($>$ 30 cm) pieces of limestone, usually in relatively steep channels (Ziemkiewicz et al., 1994). Concerns about the iron oxide coatings on limestone surfaces were alleviated when it was reported that ‘armored’ limestone was still effective in treating AMD (from 20 to 50% as effective as unarmored limestone, Ziemkiewicz et al., 1997). Treatment effectiveness has been reported to be between 4 to 62%, a reasonably good response from a low-tech, low-maintenance, inexpensive system. Thus, OLCs are a viable technology for AMD treatment in watershed restoration projects and AML reclamation projects where there can only be a one-time installation cost, little to no maintenance, and water exiting the system does not have to meet water quality standards. If constructed correctly, OLCs should be maintenance free and provide AMD treatment for decades (Skousen et al., 2017).

### 6.5. Alkaline leach beds

Alkaline leach beds are aerobic treatment systems and essentially the opposite of an open limestone channel. These systems use ponds instead of channels, have no slope, and are constructed with smaller pieces of limestone (2–10 cm). Limestone will dissolve slowly, continually add alkalinity until the limestone gets coated with metal hydroxides and becomes less effective (Ziemkiewicz et al., 1997). Because of the Fe-armoring problem, alkaline leach beds work best in low pH, metal-free water, to add alkalinity to streams before encountering AMD downstream (Ziemkiewicz et al., 2001). A novel improvement to the alkaline leach bed technology was to add steel slag (a byproduct of steel making) to the leach bed. Composed of hydrated amorphous silica and calcium compounds, steel slags can generate exceptionally high levels of alkalinity in water (pH $>$ 10 and alkalinity in the thousands of mg/L), and do not armor (Ziemkiewicz and Skousen, 1998). Thus, slag-based leach beds can receive AMD directly or the effluent from “fresh water” beds can be combined with AMD downstream for treatment (Ziemkiewicz et al., 2003).

## 7. Summary

Acid mine drainage occurs when metal sulfides are oxidized and the reaction products are leached into streams or lakes, thereby polluting over 20,000 km of streams in the USA alone. Laws require mining companies to predict AMD before mining by using overburden analyses. On sites where a potential exists, special handling of overburden materials and quick coverage of acid-producing materials in the backfill should be practiced. Alkaline addition to a pre-determined NP:MPA ratio with materials such as kiln dust and FBC ash is required to eliminate AMD problems. Other control techniques include oxygen barriers, water covers, injection of alkaline materials into underground mines, remining of abandoned areas, and alkaline recharge trenches. If AMD exists, six chemicals are typically used to treat AMD and each has characteristics that make it suitable for specific applications. Companies must choose the chemical(s) that treats the water adequately and cost-effectively, and specialized systems are sometimes needed to remove Mn, Se, and total dissolved solids. Passive systems are low maintenance systems that are implemented on abandoned mine land and stream restoration projects. Certain systems are better suited to specific water quality and, when sized and constructed properly, can provide good treatment success. Maintenance of passive systems is necessary over the long-term, but is not needed as frequently as active treatment systems.

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