EFFECTIVENESS OF THE ADDITION OF ALKALINE MATERIALS AT SURFACE COAL MINES IN PREVENTING OR ABATING ACID MINE DRAINAGE: PART 1. GEOCHEMICAL CONSIDERATIONS [1]

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Abstract. The addition of alkaline materials to supplement deficient "neutralization potential" (NP) of mine spoil, and thus to prevent or abate acid mine drainage, has riot been successful at most surface coal mines in Pennsylvania. A basic problem may have been improper accounting for acid-production potential and thus inadequate addition rates of calcium carbonate ($CaCO_3$), calcium oxide (CaO) , or calcium hydroxide [Ca $(OH)_2$] at many mines. The commonly used acid-base accounting method is based on the following overall reaction:

$$FeS_2 + 2\ CaCO_3 + 3.75\ O_2 + 1.5\ H_2O \Longrightarrow Fe(OH)_3 + 2\ SO_4^{-2} + 2\ Ca^{+2} + 2\ CO_2(g),$$

where the acidity from 1 mole of pyrite (FeS_2) is neutralized by 2 moles of $CaCO_3$. This method presumes that gaseous carbon dioxide (CO_2) will exsolve, and therefore may underestimate by up to a factor of 2 the quantity of $CaCO_3$ required to neutralize the "maximum potential acidity" (MPA) in the mine spoil. This paper reviews some geochemical reactions involving FeS_2 and various alkaline additives that support the argument that the acid-base accounting method for computing MPA from overburden analyses should be revised. Considering the stoichiometry of the following overall reaction:

$$FeS_2 + 4\ CaCO_3 + 3.75\ O_2 + 3.5\ H_2O \Longrightarrow Fe(OH)_3 + 2\ SO_4^{-2} + 4\ Ca^{+2} + 4\ HCO_3^{-},$$

4 moles of $CaCO_3$ are required to neutralize the maximum potential acidity produced by the oxidation of 1 mole of FeS_2. Therefore, the multiplication factor for computing MPA from the

overburden sulfur concentration, in weight percent, should be increased from 31.25 to 62.5.

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Introduction

Acid mine drainage (AMD), in which total mineral acidity exceeds alkalinity, is a persistent problem associated with many surface coal mines. AMD typically contains large concentrations of sulfate, iron, and other metals, and results mainly from the exposure and accelerated oxidation of pyrite (FeS_2) and additional iron-sulfide or -sulfate minerals in the coal and overburden. However, where substantial calcium- or magnesium-carbonate materials, such as limestone strata, overlie the coal, mine drainage is commonly alkaline. By corollary, where mined strata contain pyrite but lack naturally occurring calcareous material, the importation and addition of alkaline material to the mine spoil should offset the deficiency and prevent or abate AMD. However, where alkaline additives have been incorporated with mine spoil at surface coal mines in Pennsylvania, few sites have shown improvement in water quality or abatement of AMD (Brady et al. 1990).

This paper describes the chemical reactions that are the basis for computing the maximum potential acidity and net neutralization potential of mine spoil. Emphasis is placed on evaluating reactions with calcite (calcium carbonate, $CaCO_3$), "quick lime" (calcium oxide, CaO), and "hydrated lime" [calcium hydroxide, $Ca(OH)_2$], which are used as alkaline additives intended to produce near-neutral ($6 < pH < 8$) discharge water from surface coal mines.

Acid-Base

Acid-base accounting (ABA), which simplifies the complex hydrogeochemical system through use of a limited number of variables, has been used extensively in the past to estimate the quantity of alkaline material required to neutralize the potential acidity of mine spoil (diPretoro 1986; diPretoro and Rauch 1988; Skousen et al. 1987; Erickson and Hedin 1988; Ferguson and Erickson 1988; Brady and Hornberger 1989; Smith and Brady 1990; Brady et al. 1990). ABA was developed on the assumption that the stoichiometry of the following overall reaction of FeS2 and $CaCO_3$ applies (Sobek et al. 1978; Williams et al. 1982):

$$FeS_2 + 2\ CaCO_3 + 3.75\ O_2 + 1.5\ H_2O \Longrightarrow$$

$$Fe(OH)_3 + 2\ SO_4^{-2} + 2\ Ca^{+2} + 2\ CO_2(g). \quad (1)$$

The implication of reaction 1 is that acidity Produced from 1 mole (mol) of FeS_2 [64 grams (g) of sulfur (S)] is neutralized by 2 mol of $CaCO_3$ (200 g), or 1 g S to 3.125 g CaO_3. On this basis, 31.25 tons of calcium carbonate ($CaCO_3$) will neutralize the acidity from 1,000 tons of rock that contains 1. 0 weight percent (%) pyritic sulfur. In accordance with accepted ABA methods (Sobek et al. 1978), the total sulfur concentration in weight percent is multiplied by 31.25 to obtain a "maximum potential acidity" (MPA), which has units of tons of $CaCO_3$ per 1,000 tons of overburden (tons $CaCO_3$/1,000 tons) and which assumes that the sulfur is pyritic and acid producing. The 31.25 multiplication factor was intended to provide equivalent units for direct comparison with "neutralization potential" (NP), which has units of tons $CaCO_3$/1,000 tons. Subsequent workers computed the "net neutralization potential" (NNP) for coal-bearing strata by subtracting volume- or weight-weighted MPA from NP (NNP = NP - MPA) (Erickson and Hedlin 1988; diPretoro and Rauch 1988; Brady et al. 1990). A negative, or deficient, NNP Has been interpreted as the quantity of $CaCO_3$ that must be added to abate or prevent AMD.For example, if weight-weighted NP is 30 tons $CaCO_3$/1,000 tons and total sulfur concentration is 1.0%, then MPA = 31.25 tons $CaCO_3$/1,000 tons and NNP = -1.25 tons $CaCO_3$/1,000 tons. To create a net neutral mine spoil, 1.25 tons of $CaCO_3$ would need to be added to every 1,000 tons of overburden. However, the ABA method based on the stoichiometry of reaction 1 may underestimate MPA because of the presumption that CO_2 will exsolve, and thus may underestimate the $CaCO_3$ required to supplement deficient NNP.

Previous Work

Although not originally intended for the purpose, ABA following the method of Sobek et al. (1978) has been used in attempts to predict post-mining water quality. However, several researchers have arrived independently at the conclusion that equal quantities of NP and MPA (computed by multiplying the total sulfur in percent by a factor of 31.25) do not prevent AMD. Brady and Hornberger (1989) identified a given stratum as potentially acid or alkaline producing by using threshold concentrations for total sulfur or NP, respectively, of 0.5 % or 30 tons $CaCO_3$/1,000 tons (and which reacted with dilute hydrochloric acid). These threshold concentrations were corrorabated by laboratory experiments by Williams et al. (1982) and Morrison (1988) and also by Pennsylvania Department of Environmental Resources data on overburden and water quality at numerous surface coal mines. Note that 0.5 percent total sulfur, when multiplied by 31.25 to compute MPA, equals 15.6 tons $CaCO_3$/1,000 tons, roughly half the guideline number of 30 tons $CaCO_3$/1,000 tons for NP. Skousen et al. (1987, p.4) suggested that a stratum which contains values greater than 5 tons CaC03/1,000 tons as "Max Needed" (negative NNP) produced acid; conversely, values greater than 20 tons CaC03/1,000 tons as "Excess" (positive NNP) produced alkaline drainage.

diPretoro (1986) and diPretoro and Rauch (1988) showed that NP and MPA were not equivalent in using the ratio derived by dividing the cumulative volume-weighted NP by MPA for composite strata. diPretoro and Rauch (1988) found that sites having a NP/MPA ratio of less than about 2.4 produced acidic drainage, whereas most sites having a ratio greater than 2.4 produced alkaline drainage. Ferguson and Erickson (1988) showed that mine sites with a multiple-strata average NNP of 30 tons $CaCO_3$/1,000 tons or greater always produced alkaline drainage. They also found that 59 percent of mine sites with NNP of 7 to 30 tons $CaCO_3$/1,000 tons produced alkaline drainage, and only 11 percent of the sites with NNP less than 7 tons $CaCO_3$/1,000 tons produced alkaline drainage. Weighted NP in "equivalent" amounts as MPA was not sufficient to prevent AMD.

Geochemistry of Acid Mine Drainage and Alkaline Additives

The following discussion reviews some overall acid-forming and neutralizing reactions that are relevant to AMD, ABA, and the addition of alkaline materials at surface coal mines. No effort is made to account for hydrogeochemical variables such as surface- and ground-water flow paths, proximity and distribution of reacting minerals, solubilities and reaction rates of minerals, or the wide range of hydrochemical conditions in mine spoil.

Production of Acidity

AMD results from the interactions of oxygen, water, bacteria, and sulfide minerals (Singer and Stumm 1970a, 1970b; Nordstrom et al. 1979; Kleinmann et al. 1980; Cathles 1982). Pyrite (FeS_2), and less commonly, marcasite (FeS_2) are the principal sulfur-bearing minerals in bituminous coal (Davis 1981; Hawkins 1984), and because of its wide distribution, pyrite is recognized as the major source of AMD in the eastern United States (Stumm and Morgan 1981, p. 469-471). The following overall stoichiometric reactions may characterize the oxidation of pyrite and other FeS_2 minerals:

$$FeS_2 + 3.5\ O_2 + H_2O \Longrightarrow Fe^{+2} + 2\ SO_4^{-2} + 2\ H^+, \qquad (2)$$

$$Fe^{+2} + 0.25\ O_2 + H^+ \Longrightarrow Fe^{+3} + 0.5\ H_2O, \qquad (3)$$

$$Fe^{+3} + 3\ H_2O \Longrightarrow Fe(OH)_3 + 3\ H^+. \qquad (4)$$

The oxidation of sulfide in pyrite to sulfate (reaction 2) releases dissolved ferrous iron (Fe^{+2}) and "acidity" (H^+) into the water. Subsequently ferrous iron is oxidized to ferric iron (Fe^{+3}) (reaction 3), which if separated from the pyrite surface, hydrolyzes and forms insoluble ferrihydrite [$Fe(OH)_3$)] (reaction 4), and releases more acidity. The overall combination of reactions 2 through 4 may be written as follows:

$$\begin{aligned} FeS_2 + 3.75\ O_2 + 3.5\ H_2O &\Longrightarrow \\ &Fe(OH)_3 + 2\ SO_4^{-2} + 4\ H^+. \qquad (5) \end{aligned}$$

In reaction 5, 3.75 mol of oxygen are consumed to oxidize 1 mol of pyrite, and 2 mol of sulfate, 4 mol of acidity, and 1 mol of ferrihydrite are produced.

Neutralization of Acidity

Acidity produced by the aqueous oxidation of pyrite may react with carbonate, silicate, and hydroxide minerals composing the sedimentary rocks in the coal-bearing sequence. Dissolution of these acid-soluble minerals neutralizes acidity and produces the other major ions in AMD in addition to sulfate and iron, such as manganese, aluminum, calcium, magnesium, sodium, potassium, and silica.

The most acid-reactive minerals are the carbonates: calcite ($CaCO_3$), dolomite [$CaMg(CO_3)_2$], and siderite ($FeCO_3$). Carbonates are present in variable quantities as individual mineral grains and as cementing agents in limestone, dolostone, sandstone, and shale. Limestone and dolostone are composed predominantly of calcite and dolomite, respectively; shale and sandstone are composed predominantly of silicate minerals, but may contain some carbonate as cement or matrix. Dissolution of calcite, dolomite, and other calcium, or magnesium-bearing carbonate minerals tends to reduce acidity, increase alkalinity, and raise pH; however, dissolution of siderite and the subsequent hydrolysis of iron may increase acidity, and reduce pH. The dissolution of calcite by the following sequence of reactions serves as an example:

$$CaCO_3 + 2\ H^+ \Longrightarrow Ca^{2+} + H_2CO_3^*, \qquad (6)$$

$$CaCO_3 + H_2CO_3^* \Longrightarrow Ca^{2+} + 2\ HCO3^-. \qquad (7)$$

Reaction 6 represents acidic conditions (pH < 6.4) where calcite is not abundant and is totally dissolved by reacting 1 mol of calcite and 2 mol of free acidity and producing 1 mol of dissolved calcium and 1 mol of dissolved carbon dioxide {$[H_2CO_3^*] = [CO_2(aq) + [H_2CO_3^0]]$}, which is a weak acid (Krauskopf 1979, p. 40-42; Stumm and Morgan 1981, p. 171-214; Hem 1985, p. 92, 105-111). Note that gaseous CO_2 is not indicated as the product in reaction 6. In practice, during laboratory determination of overburden NP, CO_2 may be exsolved when calcite and other carbonate minerals are reacted with dilute acid. However, CO_2 may concentrate in both the gaseous and aqueous phases in mine spoil. Elevated partial pressure of CO_2 in the unsaturated zone of mine spoil is common (Lusardi and Erickson 1985), especially during the growing season, and will cause a concomitant increase in the concentration of H_2CO_3 * and other aqueous carbon-dioxide species (Langmuir 1971; Shuster and White 1972; Harmon et al. 1975).

If calcite is abundant, the dissolved carbon dioxide will continue to react with calcite (reaction 7) producing bicarbonate ions and raising pH. The overall combination of reactions 6 and 7 represents the condition where dissolution of calcite produces "alkalinity" in excess of "acidity" and raises the pH above 6.4 where bicarbonate (HCO_3-) is the dominant dissolved carbon-dioxide species:

$$CaCO_3 + H^+ \Longrightarrow Ca^{2+} + HCO3^-. \qquad (8)$$

Reaction 8 shows that 1 mol of calcite will neutralize 1 mol of free acidity and produce 1 mol each of dissolved calcium and bicarbonate. Reactions 5 and 8 may be combined to indicate a "maximum" neutralization potential of calcite, where no CO_2 is exsolved:

$$FeS_2 + 4\ CaCO_3 + 3.75\ O_2 + 3.5\ H_2O \Longrightarrow$$

$$Fe(OH)_3 + 2\ SO_4^{-2} + 4\ Ca^{+2} + 4\ HCO3^{-}. \quad (9)$$

Reaction 9 shows that the acidity produced from the oxidation of 1 mol of FeS_2 (64 g S) may be neutralized by 4 mol of $CaCO_3$ (400 g), which is a mass ratio of 6.25 g of calcite to 1 g of pyritic sulfur. The calcite-to-sulfur mass ratio of 6.25 is twice the ratio of 3.12 which would be derived considering the unlimited exsolution of CO_2 (Williams et al. 1982).

On the basis of the calcite-to-sulfur mass ratio of 6.25, a multiplication factor of 62.5 to compute MPA from total sulfur is appropriate for ABA if all sulfur is from pyrite. Therefore, considering the earlier example for overburden, where NP = 30 tons $CaCO_3$/1,000 tons and total sulfur = 1 % (only now using the 62.5 factor), then MPA = 62.5 tons $CaCO_3$/1,000 tons and NNP = -32.5 tons $CaCO_3$/1,000 tons. Instead of the previously computed 1.25 tons, now 32.5 tons of $CaCO_3$ per 1,000 tons overburden would be required to supplement the deficient NNP.

"Quick lime" (calcium oxide, CaO) and "hydrated lime" [calcium hydroxide, $Ca(OH)_2$] (Rochow 1977, p. 129), which compose lime-kiln flue dust, have twice the neutralization potential as calcite. Because the lime compounds have lower unit mass than $CaCO_3$, they are required in equivalent ratios less than 3.12 according to the following reactions:

$$Ca(OH)_2 + 2\ H^+ \Longrightarrow Ca^{+2} + 2\ H_2O, \quad (10)$$

$$CaO + 2\ H^+ \Longrightarrow Ca^{+2} + H_2O. \quad (11)$$

Reactions 10 and 11 show that 1 mol of hydrated lime (74 g) or 1 mol of quick lime (56 g) may neutralize 2 mol of free acidity. Combining reactions 10 and 5:

$$FeS_2 + 2\ Ca(OH)_2 + 3.75\ O_2 \Longrightarrow$$

$$Fe(OH)_3 + 2\ SO_4^{-2} + 2\ Ca^{+2} + 0.5\ H_2O. \quad (12)$$

Reaction 12 shows that the acidity produced from the oxidation of 1 mol of pyrite (64 g S) may be neutralized by 2 mol of calcium hydroxide (148 g), which is a mass ratio of 2.31 g of calcium hydroxide to 1 g of pyritic sulfur. Analogously, from combining reactions 5 and 11, a mass ratio of 1.75 g of calcium oxide to 1 g of pyritic sulfur is required to attain neutralization. Thus on a weight basis, 1 ton of $Ca(OH)_2$ has the neutralization equivalent of 2.7 tons of $CaCO_3$.

Siderite ($FeCO_3$) is common in coal-bearing strata and is frequently cited as having no net effect on acid-production or neutralization where CO_2 gas is exsolved (Stumm and Morgan, 1981; Williams et al. 1982). However, considering the argument for conditions with elevated partial pressure of CO_2, oxidation of siderite may produce net acidity in the form of dissolved carbon dioxide:

$$FeCO_3 + 0.25\ O_2 + 2.5\ H_2O \Longrightarrow Fe(OH)_3 + H_2CO_3^{*}. \quad (13)$$

The H_2CO_3 * generated in reaction 13 may react with additional carbonate, silicate, or hydroxide minerals. The effect of siderite as a potential acid-forming mineral is apparent by combining reactions 7 and 13 as follows:

$$\begin{aligned} &FeCO_3 + CaCO_3 + 0.25\ O_2 + 2.5\ H_2O \Longrightarrow \\ &\qquad\qquad Fe(OH)_3 + Ca^{+2} + 2\ HCO3^{-}. \qquad (14) \end{aligned}$$

In reaction 14, the acidity produced from 1 mol of siderite is neutralized by 1 mol of calcite. Thus, if siderite is present, additional alkaline material beyond that required to neutralize the acidity from pyrite may be necessary.' Impurities such as Mn, Mg, and to a lesser extent Ca, may substitute for Fe in siderite (Morrison et al. 1990). The Fe and Mn may hydrolyze and produce acid; however, the Mg and Ca may have neutralizing ability similar to dolomite and calcite.

Discussion

The presentation of acid-forming and acid-neutralizing reactions was simplified by writing and then combining independent equations as "neutral overall" reactions that eliminated H^+ as a reactant or product. Thus, the overall stoichiometries in reactions 1 and 9 equate quantities of acid-producing and -neutralizing materials and are useful for acid-base accounting application. However, reactions 1 and 9 are "end-member" reactions; the hydrogeochemical relations in mine-spoil ground water or discharge probably lie somewhere between the two end members because some CO_2 will exsolve and some will dissolve.

No attempt has been made in the above review to discuss the combined effects of variable purities, degrees of crystallinity, and particle sizes of minerals; microbiological catalysis of reactions; or relative reaction rates. For example, the presumption that 4 mol of $CaCO_3$ are required to neutralize the acidity from 1 mol of FeS_2 (reaction 9) implies that the production of acidity is rate limiting, or slow relative to neutralization, and that neutralization is instantaneous. Furthermore, the computation of maximum potential acidity (MPA) as 62.5 times the total sulfur concentration, in weight percent, should yield a conservative estimate, because not all CO_2 will dissolve nor will all sulfur be pyritic and acid producing. To determine quantities of alkaline additives required at surface coal mines, site-specific characteristics such as mining method, pre- and post-mining overburden composition, post-mining reclamation and hydrogeology, and alkaline additives used and placement technique also must be evaluated. The companion paper by Brady et al. (1990) reviews some of the site-specific factors and compares post-mining water quality and ABA computations Of MPA using the conventional and newly proposed Multiplication factors of 31.25 and 62.5, respectively, for selected surface Coal mines in Pennsylvania.

Summary and Conclusion

In summary, the ABA Method currently in use, which presumes 2 mol of H+ may be neutralized by 1 Mol of $CaCO_3$, may underestimate by up to a factor of 2 the $CaCO_3$ required to neutralize the maximum potential acidity from the oxidation of pyrite and the hydrolysis and precipitation of iron, because of the presumption that all CO_2 will exsolve. However, some CO_2 will dissolve forming a weak acid that reacts with carbonate minerals. Assuming no exsolution of CO_2, 1 mol of FeS_2 will produce 4 mol of H^+, which may be neutralized by 4 mol of $CaCO_3$ (reaction 9), 2 mol of $Ca(OH)_2$ (reaction 12), or 2 mol of CaO. on a weight basis then, 1 g of pyritic sulfur may be neutralized by 6.25 g of $CaCO_3$, 2.31 g of $Ca(OH)_2$, or 1.75 g of CaO. Considering these equivalent weights, MPA as tons of $CaCO_3$ deficiency per 1,000 tons of overburden should be computed by multiplying total sulfur, in weight percent, by 62.5. The above discussion is based only on the stoichiometry of the overall reactions (9 and 12) and assumes that the rate of acid production will not exceed the rate of acid neutralization. The actual acidity may be less than the computed MPA because not all CO_2 dissolves and not all sulfur generates acidity. Finally, dissolution of siderite will produce net acidity when the partial pressure of CO_2 becomes elevated and the iron is hydrolyzed and precipitated.

In conclusion, for conservative estimates of overburden net neutralization potential (NNP), a revised multiplication factor of 62.5 should be used to compute maximum potential acidity (MPA) from the total sulfur concentration, in weight percent.

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