RELATING SULFATE GENERATION TO A MODEL OF PHYSICAL FLOW THROUGH WASTE ROCK PILES: ATTEMPTS AND CONSEQUENCES[1]

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Abstract: In an effort to learn more about the causes of acid rock drainage, a study was conducted to see how physical flow through a waste rock pile can be linked to recently published sulfate generation rates. To model the amount of water passing through the waste rock pile, the EPA's Hydrologic Evaluation of Landfill Performance (HELP) program was used. The HELP model is an appropriate simulation for embankments utilizing waste rock because it evaluates the hydraulic characteristics of placed materials within unique layers based on physical properties such as permeability, soil moisture capacities, and drainage layer configurations. The first attempt at linkage of the physical and chemical parameters assumes that an average value for the sulfate generation rate integrated over time and space is reasonable, and thus the amount of sulfate generated will be primarily dependent upon physical flow through the pile. From recent published studies on waste rock piles, a sulfate release rate of between 2.1 and 10.5 millimole $SO_4^=$ / metric ton / day has been established. Using the mass of the pile, and the flow of water from it as predicted by the HELP model, the concentration of sulfate in the water can be readily calculated. This simple method of prediction of acid generation from a waste rock pile gives concentrations of sulfate that are comparable to published values. Attempts at adjusting physical parameters in the HELP model such as permeability, the slope of the drainage layer, the wilting point of the soil, the field capacities of the waste rock layers, and the thickness of the layers had little or no effect on the volume of drainage predicted and, therefore, the concentration of sulfate in the drainage. Predicted concentrations were most sensitive to the amount of precipitation. This was because the amount of flow from the pile was substantially changed. These preliminary conclusions suggest that purely physical considerations can explain some of the concentrations of sulfate generated by the oxidation of pyrite. Chemical changes that remove sulfate or lower the oxidation rate were ignored for this phase of the study. Attempts at doing this using chemical modelling programs are currently underway.

Additional Key Words: acid mine drainage, mine waste rock, HELP model

Introduction

In a broad analysis, the chemistry of drainage from sulfidic waste rock piles is a function of the sulfide oxidation rates, host rock mineral dissolution rates, the extent of the reaction product transport from the pile, and the volume of drainage. The purpose of this paper is to provide a simplistic approximation of sulfate concentrations in acid rock drainage. Published sulfate generation rates are used as representative of all waste rock. EPA's Hydrologic Evaluation of Landfill Performance (HELP) program is used to model the physical flow of water through the waste rock pile (Schroeder et al., 1992a, 1992b). Transport of the sulfate from the reaction sites to the outflow is assumed to be conservative. It is hoped that by assessing whether realistic sulfate concentration values result, a better knowledge can be obtained of the causes of acid rock drainage. This could lead to the development of treatment measures that eliminate generation of acid drainage at the source

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The authors realize that the approach applied in this paper is overly simplistic. Basing water chemistry only on an average groundwater flow and a single sulfate generation rate ignores many of the specific physical and chemical influences in waste rock piles. However, this approach can be applied quickly and inexpensively to obtain a "first cut" estimate of waste rock drainage quality. Furthermore, this approach may reveal the relative importance of specific physical and chemical processes on drainage quality, and thereby identify directions for future work.

The modelling concentrates on sulfate generation because it is a primary product in the first step of pyrite oxidation, which is the primary cause of acid mine drainage.

$$FeS_2 + 7/2\ O_2 + H_2O \text{ ------> } Fe^{2+} + 2\ SO_4^{=} + 2\ H^+$$

In addition, until the concentration of calcium is high enough for gypsum precipitation, sulfate will generally not be precipitated or adsorbed. Consequently, its concentration in the water is the best indication of the level of pyrite oxidation. Finally, knowing the sulfate concentration provides a good estimate of the total acidity generated through pyrite oxidation even though the water may have been subsequently neutralized. Two moles of hydrogen ions and one mole of Fe^{2+} are released for every two moles of sulfate released. Eventually, the Fe^{2+} will oxidize and precipitate.

$$Fe^{2+} + 1/4\ O_2 + 5/2\ H_O \text{ ------> } Fe(OH)_3 + 2\ H^+$$

This produces two more moles of hydrogen ions so that one mole of sulfate released from pyrite is associated with two moles of eventual acidity. The combination of these three facts makes knowing the amount of sulfate generated as an excellent way to measure the general chemical nature of an acid rock drainage.

From a regulatory point of view, a reasonably reliable prediction of the amount of acidity in the drainage from a waste rock pile would be a useful tool in designing a plan for operation and closure of a mine. How the chemistry of mine drainage is affected by changes in climate, particularly the amount of precipitation, is also an interesting question. Finally, if sulfate is one of the constituents measured in a humidity cell test, then an average sulfate generation rate can be determined from that test (Lapakko, 1988, 1994). Linking that laboratory sulfate generation rate with a physical flow model can give predictions on the chemistry of the drainage from a planned waste rock pile. When this is done, the question will be just how well do those predictions estimate the actual chemistry of the water.

Sulfate generation rates have been published by a few research groups who have extensively studied specific waste rock piles. Since 1978, Lapakko (1994) has been studying waste rock piles in northern Minnesota. Based on field data, he has estimated that the sulfate release rate in these piles is between 2.1 and 10.5 millimol $SO_4^{=}$ / metric ton / day. Bennett and others (1994) studied a waste rock dump in Australia and found the intrinsic oxidation rate to be about 10^{-8} kg O_2 / m^3 of waste rock / sec. However the values of sulfate generated did not correlate with the oxidation rate. They estimated 14 % of the dump would contribute pollution loads at this rate of oxidation and the remaining 86 % would contribute little to the total pollution load. Gibson, Pantelis, and Ritchie (1994) have used the Australian oxidation rate data to estimate the evolution of drainage chemistry from a pyritic waste rock dump. In their study, they suggest that the oxidation rate is independent of oxygen and sulfur concentrations as long as neither is zero. For microbiologically controlled oxidation, the result that pyrite oxidation is independent of oxygen, when the amount of oxygen in the atmosphere is above 1 %, has been verified in the laboratory by Hammack and Watzlaf (1990). If the oxidation rate is somewhat independent of oxygen and sulfur concentrations, then the generation of acid mine drainage would be somewhat independent of these variables.

Another parameter that is important to the outflow of drainage is transport of the water through the waste rock pile. Snyder and Caruccio (1988) and Wildeman (1991) have found evidence that suggests that, in the field, the release of acid rock drainage is kinetically controlled. It appears that the pyrite weathering products are retained in microfaults above the water table where the oxidation reaction is most favored. Then, this water is released from the faults when recharge of the ground water in the waste rock pile occurs. If the pile is large enough to maintain somewhat constant outflow, then the chemistry of the acid rock drainage will not change substantially with the seasons or precipitation events (Wildeman, 1991). If this evidence is correct, then the flow of recharge water to reaction sites is limited. In this study, it is also assumed that there is no production or consumption of sulfate during flow to the outlet.

If the above assumptions concerning the generation and release of sulfate are plausible, then linking a physical flow model with a gross oxidation rate should give a reasonable estimate of the amount of pollution. The HELP model used on landfills has been extensively applied to the flow of water through such situations (Schroeder, et al., 1992a, 1992b). The objective of our study has been to estimate flow from the waste rock pile using the HELP model. With this flow value and the sulfate generation rate, the concentration of sulfate in mg/L can be determined. Two additional assumptions made in the calculation are:

1. The concentration of calcium in the drainage is low enough that gypsum solubility does not control the concentration of sulfate.
2. From a chemical viewpoint, the pile is homogeneous and oxidation is equally likely at any place in the pile.

Correlation of this concentration with values from waste rock drainages provides the following insights:

1. If the modelled concentration values are far different from reported values, then generation of acid rock drainage may be much more complex. Further physical and chemical studies will be necessary to understand the process.
2. There may be certain parameters in the HELP model that drastically affect the amount of water flowing from the pile. Can these parameters be used to control the generation of acid mine drainage?
3. The HELP model can be adjusted for different climates and geographies. How does this affect the quality of drainage flowing from the waste rock pile?

The present study focuses on sulfate concentrations in drainage from test piles of Duluth Complex rock in northeastern Minnesota. The sulfate in drainage from these piles is primarily the result of pyrrhotite oxidation (Lapakko, 1994). The annual drainage volume predicted by the HELP model agrees well with reported values (Lapakko, 1994). Given this agreement, HELP model parameters were varied to investigate the influence of these variations on flow and, consequently, predicted sulfate concentrations in the drainage. Finally, the effect of climatic setting on sulfate concentrations was investigated.

HELP Model Considerations

The HELP model is an appropriate simulation for waste rock embankments because it accounts for the hydraulic characteristics of placed materials within unique layers based on physical properties such as permeability, soil moisture capacities, and drainage layer configurations. The model allows the pile to contain up to twelve different layers of materials each with its own characteristics. It also allows the user to select the location of the site, the only restriction being that must be located in the United States for the user to take advantage of the data base that comes with the program. From the data base in HELP, comes the precipitation, growing season, evaporative zone, and maximum leaf index data characteristic for that specific area. The data base includes major cities from all 50 states, however, modifications are possible for remote locations or another site around the world. Table 1 presents the HELP parameters that can be fixed by default and those that must be selected.

Once these data have been entered, the program uses this information to provide the amount of water in cubic feet per year that will percolate through the pile. The amount of percolation will primarily depend on the number of layers, the hydraulic conductivity of these layers, and the amount of rainfall. In addition, flow will also depend on other parameters included in the program. In this study, what flows from the pile is the acid rock drainage.

Table 1. Parameters capable of adjustment in the HELP Model

Parameters That Can be Set by Choosing a Default Site

- Yearly precipitation
- Growing season
- Evaporative zone depth
- Maximum leaf index data
- Mean solar insolation in langleys
- Mean monthly temperature

Parameters Set by Each Situation

- Area of the waste rock pile
- Number of soil (rock) layers and thickness of each layer
- Soil conservation System (SCS) curve number for the uppermost soil layer
- Type of layers (vertical percolation, lateral drainage, or barrier soil)
- Soil porosity - the ratio of the volume of voids to total volume occupied by the soil
- Soil field capacity - the ratio of volume of water that a soil retains after a prolonged period of gravity drainage to total volume occupied by a soil.
- Soil wilting point - the ratio of volume of water that a soil retains after plants can no longer extract water (thus the plants remain wilted) to total volume occupied by a soil.
- Saturated hydraulic conductivity (cm/sec) - the rate at which water moves through soil in response to gravitational forces.
- The slope of the bottom of drainage layers (in percent)
- Maximum horizontal drainage distance (in feet)

Modelling the Minnesota Waste Rock Pile

The waste rock pile is sited in northern Minnesota and the climate conditions for St. Cloud Minnesota were chosen. The pile has no topsoil layer. As shown in the diagram of the waste rock pile in figure 1, three layers were chosen:

1. A 380 cm (150 inch) waste rock layer, which is set as a vertical percolation layer. The hydraulic conductivity is 10 cm/s, porosity is 0.4, field capacity is 0.2, and wilting point is 0.1.
2. A 18 cm (7 inch) waste rock layer with the same properties as Layer 1 except that it is set as a lateral drainage layer. To do this, the layer was set upon a 5 ° slope. The actual pile slope is 1.4 °.
3. A 4.5 m (15 foot) native ground layer that was set to be a barrier soil layer. The hydraulic conductivity is 2×10^{-6} cm/sec, porosity is 0.4, field capacity is 0.31, and wilting point is 0.21. The actual piles are underlain by Hypalon® liners.

Layers 2 and 3 serve to cause the drainage to flow from the pile. The actual dimensions of 4.0 m (157 inches) by 13.7 m by 26.0 m (45 ft x 85 ft) for the pile and mass of 1100 metric tons were provided by Lapakko (1994). For the modelling calculations, the SCS runoff curve is set at 85.00, evaporation zone is 20.3 cm (8.00 inches), the total area of cover is 370 m^2 (4000 $ft^{2)}$.

Using these parameters in the HELP model produced a flow from the pile is 112 m3 / yr (3970 ft^3 / yr) or 3.6×10^{-3} L / sec. The average sulfate release is 2.1 to 10.5 millimole $SO_4^=$ / metric ton / day or 2.6 to 13 x 10-6 kg $SO_4^=$ / sec. Division of the sulfate release by the flow gives a concentration of sulfate in the drainage of between 720 and 3600 mg / L.

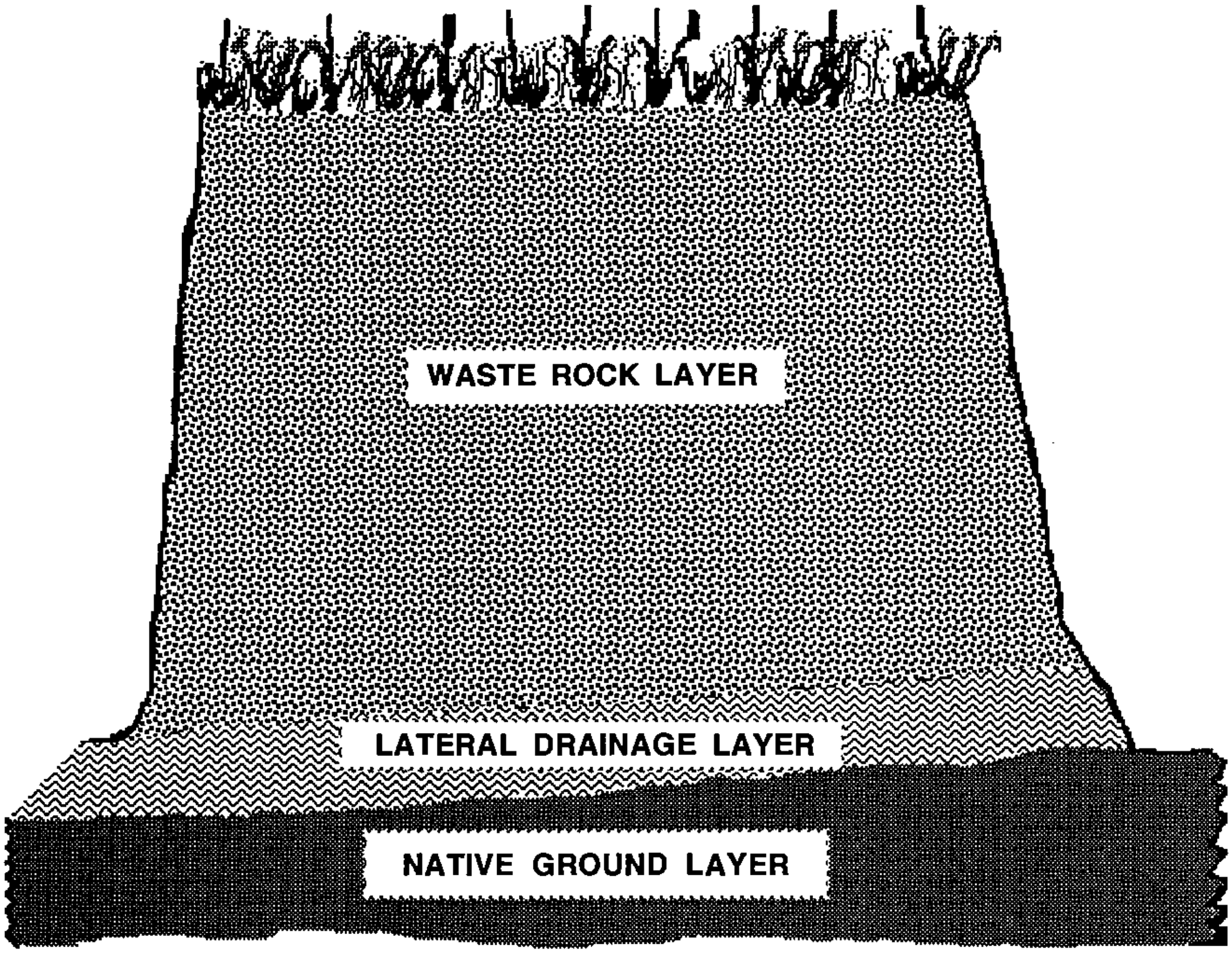

Figure 1. A schematic diagram of the northeastern Minnesota waste rock pile showing the layers used in the HELP model.

Lapakko (1994) reports a flow from the piles of between 0.27 and 0.38 L / mt of rock / day. Converting 112 m^3 / yr (3970 ft^3 / yr) to this scale gives a value of 0.28 L / mt of rock / day. Because the annual flow rate from the HELP model reasonably approximates the actual annual flow, the sulfate concentration values of between 720 and 3600 mg / L correlate well with the concentrations of 680 - 3800 mg / L over 1989 - 91 from the FL1 waste rock pile (Lapakko, 1994). These concentrations are at least a factor of 2 higher than the sulfate concentration values that Lapakko (1988) reported on humidity cell tests on similar waste rock samples. In more recent laboratory studies, Lapakko and Antonson (1993) found that laboratory release rates of sulfate were 3 to 10 times higher than the field release rates. Lapakko (1994) hypothesized a number of reasons for the differences between laboratory and field release rates, however there is not enough information to suggest a primary cause for the difference.

Effect of HELP Model Variables on Flow

Sensitivity of the flow rate of 112 m^3 / yr (3970 ft^3 / yr) to changes in many of the parameters shown in Table 1 were slight. Adding a top layer of plant growth medium of 5 inches thickness to act as a vertical percolation layer with a conductivity of 6×10^{-4} cm / sec had no effect on the amount of flow. Doubling the thickness of this layer also had no effect. Changing the slope of the lateral drainage layer did not affect the flow. Changing the field capacities, wilting point, and thicknesses of the other drainage layers likewise had little effect on the flow. Of course, changing the surface area of the pile while maintaining the same volume, will change the amount of flow. This is the reason why the model has to be calibrated using an actual situation.

The one parameter that did appreciably change the flow is the amount of precipitation that falls upon the pile and this is a function of climate. This can be tested in the HELP model by "moving" the pile to different sites. To test the effect of climate, all the variables that were not default variables dependent upon the site were kept constant and the HELP model was run in a variety of cities to test the effect of climate. In this exercise, the SCS runoff curve of 85.00 was kept constant but the evaporative zone depth changed with the climate of the site. The predicted drainage flow in cubic feet per year and the ranges and average concentration of sulfate at different cities is given in Table 2. Cities in Table 2 are chosen because they:

1. Provide a diversity of climates,
2. Are near areas of coal or metals mining,
3. Have default historical precipitation data in HELP
4. Have default temperature and solar radiation in HELP.

The sulfate concentration values are comparable to some situations found in metal mining areas. Wildeman, Filipek, and Gusek (1994) report on an acid rock drainage from near Ely, NV. The water had a pH = 2.5 and a sulfate concentration of 3370 mg / L. This is within the range for the Ely site. Wildeman et al. (1994) also report on drainage from a closed gold mine in northern California near Sacramento. It had an average pH of 3.8 and an average sulfate concentration of 2800 mg / L. On the other hand, sulfate concentrations can be quite a bit higher than the Table 2 ranges. In eastern U.S., Gusek, Gormley, and Scheetz (1994) report on the acid rock drainage chemistry from a gold mine near Jefferson, South Carolina that is undergoing closure. The climate is similar to that in Knoxville, TN. For waters that range from pH 2.3 to 2.5, the sulfate concentrations range from 2500 to 3300 mg/L. On the highest side in western US, Alpers and Nordstrom (1991) report that the drainage from the Richmond portal at Iron Mountain, CA near Sacramento has a pH of 0.5 and sulfate concentration of 120,000 mg / L. All the sites mentioned are areas where the oxidation of sulfidic minerals is vigorous enough to lower the pH of the drainage to values below 5, the situation that prevails for the Minnesota waste rock piles (Lapakko, 1994).

Table 2. Yearly flow and ranges and averages of sulfate concentrations from the northeastern Minnesota waste rock pile placed in different sites in the United States.

Geographic Site	Yearly drainage ft3 / year	Sulfate Conc. Range mg / L	Average Sulfate Conc. mg / L
St. Cloud, MN	3970	720 - 3600	2200
Cheyenne, WY	1470	1940 - 9600	5800
Des Moines, IA	5400	530 - 2600	1600
East St. Louis, IL	6200	460 - 2300	1370
Ely, NV	980	2900 - 14,400	8700
Great Falls, MT	2700	1010 - 5300	3200
Knoxville, TN	8800	330 - 1610	970
Miami, FL	7570	380 - 1860	1120
New Orleans, LA	12,430	230 - 1130	680
Phoenix, AZ	1259	2300 - 11,200	6800
Pittsburgh, PA	6500	440 - 2200	1300
Providence, RI	9430	300 - 1500	900
Sacramento, CA	3000	940 - 4700	2800
San Diego, CA	2365	1200 - 6000	3600
Seattle, WA	7900	360 - 1800	1080

Effect of Other Oxidation Rates on Sulfate Concentrations

Attempts were made to use the published oxidation rate of Bennett and others (1994) of 10^{-8} kg O_2 / m3 of waste rock / sec. In this case, the rate of oxidation is based on oxygen consumption and the amount of sulfate generated is assumed to be based on the pyrite oxidation reaction. Also in this case, oxidation on a volume basis has to be transformed to a mass basis by assuming that the in-place density of rock in the pile is 1.4 gm / cm^3. Using these values for a waste rock pile of 1100 mt, gives a sulfate generation rate of 14 x 10^{-6} kg $SO_4^{=}$ / sec. This rate would give sulfate concentrations at the high end of the ranges shown in Table 2. In their study, Bennett and others (1994) found that an intrinsic oxidation rate of 10^{-8} kg O_2 / m^3 of waste rock / sec was high and made the suggestion that only 14 % of the pile would contribute to the pollution load and the remaining 86 % would contribute little. Considering the high concentration of sulfate that this oxidation rate produces, this suggestion appears reasonable.

Conclusions

In this study, our expectations were to be within the range of sulfate concentrations usually associated with waste rock drainages. In this respect, we met with some success. To the extent that this approach is reasonable, it implies that in considering the causes of waste rock drainage, the primary factors are the amount of water flowing through the pile and the access of oxygen to the inside of the pile. Factors such as mineralogy, the abundance of sulfur, and the flow of water through the pile have a secondary impact. In considering the pyrite oxidation reaction, there are three reactants: pyrite, oxygen, and water. In most instances, oxygen is the limiting reactant and controls the kinetics of the reaction.

With respect to treatment activities, design of the waste rock pile to only minimize flow through the interior will decrease the flow but not decrease the loading. Consequently, the flow will be lower, and the sulfate concentration correspondingly higher. To decrease loading, oxygen access to the interior must be reduced. Gibson, Pantelis, and Bennett (1994) and Hammack and Watzlaf (1990) both suggest that until the atmospheric oxygen concentration falls below 1 %, the rate of oxidation is constant. Elimination of oxygen in the interior of the pile can be accomplished by sealing off the pile or by reacting the oxygen with some other material. If a top-cover is added to the pile that not only eliminates water but also cuts oxygen delivery to the interior of the pile, then addition of a cover may help to eliminate acidic drainage. James Gusek (personal communication) has long advocated the addition of organic material to the top-cover of a waste rock pile. If such a layer consumed the oxygen through oxidation of the organic material then this would be an example of having the oxygen react with another material. Both the addition of a cover to retain water and block oxygen diffusion and the addition of organic material to consume the remaining oxygen have been included in the design of a cover for the closure of a tailings pile in a mountainous region in western United States. This cover is described by Wildeman and others (1994) in another article in this proceedings.

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