

CONTROL OF ACID MINE DRAINAGE USING ANIONIC SURFACTANTS

Robert L. P. Kleinmann and Patricia M. Erickson

U.S. Bureau of Mines
Pittsburgh Research Center
Post Office Box 18070
Pittsburgh, Pennsylvania 15236 U. S. A.

ABSTRACT

Acid drainage from abandoned coal mines pollutes thousands of kilometers of streams in the eastern United States and causes the coal mining industry of the area to spend millions of dollars annually to prevent further degradation of local waterways. To control the formation of acid drainage at its source, the U.S. Bureau of Mines has investigated the role of iron-oxidizing bacteria in the oxidation of pyrite. We have determined that inexpensive bactericides can be used to control the rate of pyrite oxidation and thereby reduce the acidity of drainage.

At concentrations of 25 mg/l or greater, certain biodegradable anionic surfactants, such as sodium lauryl sulfate, kill the acidophilic iron-oxidizing bacteria in the pyritic material. To prevent repopulation, we have incorporated the surfactant into rubber pellets, from which the surfactant is gradually released over time. After an initial application of surfactant solution, these pellets are placed so as to intercept and treat infiltrating rain water on a surface mine or refuse pile. The rubber formulation can be adjusted to modify the duration of release; results of laboratory studies are being used to optimize both phases of the treatment procedure.

The technique has been tested in small-scale laboratory and field experiments. Treatment resulted in titratable acidities which were 75 pct lower than those of controls in laboratory simulations, 67 pct lower when one of two 25 tonne coal refuse piles was treated and 95 pct lower when a test was conducted on isolated sections of an acid-producing surface mine.

Large-scale field tests have been initiated at surface mines and coal refuse piles in Ohio, West Virginia and Pennsylvania to provide information on adsorption of the surfactant by fine-grained particles and to determine if downstream concentrations of surfactant pose a potential environmental problem. Preliminary results indicate surfactant concentrations in the effluent of 0.1 mg/l or less, well within the World Health Organization's suggested stream concentration of less than 1.0 mg/l.

INTRODUCTION

Acid mine drainage is a pollution problem primarily associated with the mining of coal and sulfide minerals. Although it has long been recognized that controlling the cause of the acid mine water problem is preferable to continuous treatment of the resultant drainage [1], the few at-site control methods which exist share the disadvantage of high initial costs. Previous studies have demonstrated that an iron-oxidizing bacterium, Thiobacillus ferrooxidans, plays a major role in the oxidation of pyrite

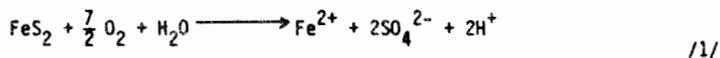
and the formation of acid mine drainage [2-5]. In this report we discuss a practical method of bacterial inhibition which has significantly reduced acid formation in both laboratory and field tests.

BACKGROUND

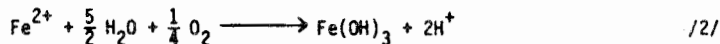
Almost since its isolation in 1947, *T. ferrooxidans* has been regarded as a possible agent in the problem of acid mine drainage [6]. In 1970, Singer and Stumm demonstrated that direct oxidation of pyrite by air was too slow to generate the amount of acidity observed in nature and suggested that *T. ferrooxidans* acts as a necessary catalyst for the relatively more rapid oxidation of pyrite by aqueous ferric ion [2, 7]. Lau, and others [8] argued that such a mechanism would be important only in near-surface environments. However, it has been shown in laboratory simulations that although *T. ferrooxidans* is unimportant in saturated environments, it significantly increases acid formation near the land surface, and is intermittently significant in the intermediate belt of the zone of aeration [5]. In the last case, *T. ferrooxidans* increased acid production for three to four days after each rainfall infiltration, after which acid production fell to the level of sterile controls.

Pyrite oxidation occurs in a multistage sequence dependent upon the activity of *T. ferrooxidans* and solution Eh and pH [9]. Within these stages, the mechanics of reaction are complex and only partially understood. For example, oxidation of the soluble sulfide species is accelerated by the action of *Thiobacillus thiooxidans*, and possibly *T. ferrooxidans* as well. In this discussion, only the general reaction system will be considered.

Caruccio [10] has shown that the most reactive pyrite is the framboidal form due to the presence of pyrite granules less than 0.5 μm in diameter. During the first stage of acid formation, this fine-grained pyrite is oxidized either by *T. ferrooxidans* or by air with equal amounts of acidity produced by the oxidation of sulfide to sulfate (reaction 1) and by the hydrolysis of Fe^{3+} (included in reaction 2).



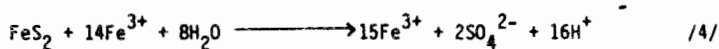
and direct oxidation of pyrite by *T. ferrooxidans*



The ability of *T. ferrooxidans* to accelerate the rate of pH decline is important, for each rainfall potentially interferes with the initial buildup of acid. During this stage, it is possible to forestall acidification by adding alkalinity to the reaction system; if alkalinity exceeds acidity, the only major downstream effect is an increase in sulfate concentration. Thus, adding crushed limestone or other sources of alkalinity to freshly exposed pyritic material can stop or postpone acidification. Once acidity significantly exceeds alkalinity, it becomes much more difficult to return the acid-producing system to stage 1, although the fall of pH is moderated as it approaches 4.5 by a pronounced decrease in the rate of reaction 2 [11].

As abiotic oxidation of Fe^{2+} slows, *T. ferrooxidans* takes on its primary role of oxidizing Fe^{2+} , thereby allowing reaction 2 to continue producing acidity and ferric hydroxide. This initiates stage 2 of the reaction process. Once again it is possible for the pH to stabilize in this region, though this will usually occur only when permeability or the amount of exposed pyrite surface area is low. Otherwise, the pH decline continues to the third stage, where acid production is most rapid.

At pH values less than 3, the increased solubility of iron and the decreased rate of $\text{Fe}(\text{OH})_3$ precipitation results in increased Fe^{3+} activity [11]. Stage 3 begins as Fe^{3+} activity becomes significant at a pH in the immediate vicinity of the pyrite of approximately 2.5; the associated drainage is typically a pH unit higher. A vicious cycle of bacterial oxidation of Fe^{2+} and oxidation of pyrite by Fe^{3+} results from the combined effects of reactions 3 and 4.



The rate of reaction 3 exerts primary control on the cycle by limiting the availability of Fe^{3+} which is the major oxidant of pyrite. The steady-state activity of Fe^{3+} is determined by the combined effects of bacterial oxidation of Fe^{2+} , reduction of Fe^{3+} by pyrite, and the formation of aqueous ferric complexes. Stage 3 includes the oxidation of both fram-boidal and coarser-grained pyrite.

The transition to stage 3 can be seen in the graph of total dissolved iron and acidity vs. pH (fig. 1) which has been compiled from a series of experiments conducted in previously described simulations of a coal refuse pile 5. Acidity was determined in the laboratory by titrating with NaOH to pH 8.3 after boiling with hydrogen peroxide and is expressed as mg/l CaCO_3 [12].

Figure 2 is an Eh-pH diagram with the three stages of acidification demarcated [9]. We have plotted points and field data obtained at actual refuse piles, abandoned surface mines, and at discharge points of abandoned deep mines. Also included in figure 2 are acid drainage Eh and pH data from the literature [13-17].

As can be seen in figure 2, pyrite and $\text{Fe}(\text{OH})_3$ theoretically cannot coexist at acid pH. The fact that they are found commonly together at sources of acid drainage demonstrates that pyrite oxidation is controlled by kinetic processes rather than equilibrium chemistry. Hence, data included in figure 2 do not necessarily represent solubilities of either pyrite or $\text{Fe}(\text{OH})_3$.

Generally, the Eh-pH values in stages 1 and 2 can be seen to approximate the solubility boundary for $\text{Fe}(\text{OH})_3$. The pH boundary between stages 1 and 2 does not appear as a change in slope because neither the mechanism nor the rate of pyrite oxidation are affected by the transition from inorganic to bacterial ferrous iron oxidation.

In stage 3, the slope changes as steady-state cycling begins between reaction 3 and 4, causing the Eh to stabilize at a kinetically controlled Fe^{3+}/Fe^{2+} activity ratio. Once stage 3 is reached, acid production can only be reduced by slowing reaction 3 or 4. It is traditionally done by limiting the available oxygen, but the more direct route of inhibiting *T. ferrooxidans* is also possible.

BACTERIAL INHIBITION

We have evaluated inhibitors of *T. ferrooxidans* gleaned from the biological literature [18-21] and have determined that anionic detergents are the most economical. Normally considered as cleansers rather than bactericides [22], anionic detergents do have bactericidal properties at low pH [23], are readily available in biodegradable forms and are environmentally safe at low concentrations. Dugan [16] has demonstrated that in 9-K liquid culture media, anionic detergents can effectively stop iron oxidation by *T. ferrooxidans* at concentrations as low as 2 ppm. Somewhat higher concentrations (Figure 3) were necessary in laboratory simulations of a coal refuse pile [24]. For example, sodium lauryl sulfate (SLS) at concentrations of 10-20 ppm reduced acid formation by about 40 pct, and at concentrations of 25 ppm was bactericidal, with acid formation reduced by an additional 50 pct. Evidence indicates that the detergents alter the semipermeable properties of the cytoplasmic membrane [25], allowing seepage of H^+ into the cell [26]. The bacteria are thus appropriately attacked by the acid which they help to produce.

Control of bacteria in the field is complicated by several potential problems. For example, the high solubility of most anionic surfactants precludes long-term control of the bacteria from a single application; repopulation occurs if the concentration of SLS falls below 10 ppm. To provide long-term control, we have incorporated the surfactants into rubber pellets, which can be applied easily to a refuse pile or surface mine spoil or mixed with the pyritic material [27]. These pellets gradually release the surfactant into infiltrating rainwater.

Release rates of SLS from the rubber matrix were measured in the laboratory using periodic rinsing of the rubber samples to simulate detergent release during rainfall [28]. Ten of the eleven formulations tested exhibited two-stage release: an initially fast linear rate during which 4 to 21 pct of the detergent dissolved, followed by much slower release.

Figure 4 illustrates the relative rates of detergent release from identical matrices with different detergent loadings. The 41 pct SLS sample released a higher percentage of detergent than did the 26 pct SLS sample. The effect of matrix formulation is illustrated in Figure 5. Both samples were composed of 53 pct natural rubber, 37 pct SLS, and 5 pct vulcanizing additives. The remaining 5 pct was silicious filler in one case and carbon black in the other case. The nature of the filler significantly affected the SLS release curves. The presence of synthetic rubber and other additives were also found to influence detergent release rates. Figure 6 illustrates detergent release curves for two samples which differed only in pellet size. The 1.6 mm thick pellets displayed a markedly increased loss of detergent in comparison with the 6.4 mm thick pellets.

Adsorption also limits the amount of surfactant reaching the acid-producing environment. Coal and fine-grained particles such as clay retain the surfactant. To counteract this, before or during the application of the controlled-release rubber, we have to apply the surfactant in solution to saturate the adsorptive capacity of the site. Although field conditions cannot easily be duplicated, approximate values can be determined in the laboratory rapidly and at low cost using a detergent adsorption test [28].

To simulate field application, the material to be tested is packed in a Buchner funnel and treated with a volume of SLS solution to achieve the equivalent of 2 to 8 grams of dry detergent per cubic meter. The volume is insufficient to completely wet the refuse or overburden. A second Buchner funnel is used to drip a 25 ppm SLS rinse solution on to the material. The effluent is collected in 100 ml aliquots and analyzed by the methylene blue method [12]. Adsorbed detergent is the difference between applied and eluted SLS.

Adsorption values ranged from approximately 45 to 75 μg SLS per gram of refuse and from approximately 4 to 11 μg SLS per gram of overburden. The overburden samples appeared to be very fine-grained, yet higher adsorption rates were observed in the refuse samples. These values can be taken as upper limits of adsorption since the surface area of the material is decreased by compaction in the field. The variable adsorption values indicate that a similar test should precede each field application to ensure that an appropriate amount of detergent is applied. In recent field tests, the initial application rates have been increased 2- to 20-fold over original estimates that were based simply on grain size.

An intriguing possibility is that the coal refuse might act as a controlled release matrix if the adsorption is slowly reversible. H. Olem [29] of the Tennessee Valley Authority is currently investigating the effect of SLS on acidification of runoff from stored coal. After a single treatment, acidification was delayed 2 months, although no SLS was detected in the runoff. Acid production continues to remain lower than in the control sample after 4 months. This indicates that adsorbed SLS may be bacteriostatic or that slow desorption may be responsible for delayed acidification. Our laboratory tests also suggest that the adsorbed surfactant may be slowly released. Laboratory testing of these possibilities have been initiated; results will be presented at the meeting.

FIELD TESTS

Our first field test had two purposes: (1) to test the concept of controlled release of anionic detergents as a method of slowing or stopping acidification and (2) to determine the amount of rubber necessary to produce the desired detergent concentrations.

Two 25 tonne coal refuse piles were established for experimentation near Carolltown, Pennsylvania, by the Barnes and Tucker Coal Co. The ground beneath the pile had been graded so as to drain in one direction. Plastic sheeting beneath the piles and a perforated pipe running through the piles directed drainage to a collecting bucket, which was emptied after each sampling. One of the piles was treated with seven slabs of SLS-rubber (totalling 27 kg) which were distributed beneath the surface of the pile

to minimize interference with natural infiltration; the other pile was left untreated as a control.

Detergent concentrations in the drainage ranged from 2-12 mg/l, slightly less than half of the desired concentration range. Acidity and iron concentrations of the drainage were nonetheless consistently lower in the treated pile (figure 7). Acidification was delayed by the treatment to the extent that at 100 days, acidity and iron concentrations were one-eighth of those in the treated pile. After about five months, acidity and iron values stabilized at a level one-third of those in the control pile. These reductions occurred despite lower than desired detergent concentrations in the pile and were probably due to bacteriostatic or bactericidal detergent concentrations in the immediate vicinity of the rubber slabs.

It was apparent that higher detergent concentrations were required and that therefore either the amount of impregnated rubber or the detergent release rate had to be increased. We have accomplished the latter by decreasing the thickness to less than 1 cm, thereby increasing the surface area [9].

In our second field experiment, we tested the controlled release of anionic detergents as a means of reducing acid formation in an abandoned and unreclaimed surface mine near Bruceton Mills, West Virginia. The SLS-rubber formulation was applied to the more acid-producing of two small sections of pyritic shale overburden, the drainage from which had been artificially isolated by F. Caruccio. Sufficient rubber was applied to generate, at the release rate observed in the first field test, an anticipated average detergent concentration of 25 ppm. Measured detergent concentrations ranged from 20-60 ppm, the latter observed at the end of the winter freeze. Significant reduction in drainage acidity was observed after two months of treatment; after six months of treatment acidity was reduced to approximately 5 pct of original levels (figure 8). Nine months after treatment, pH of the drainage from the treated section was 6.86 with only 25 mg/l acidity as compared with pH in the control section of 2.0 and acidity of 25,250 mg/l. It is to be expected that somewhat longer lag periods can be anticipated in the treatment of larger acid-producing areas because of the higher volumes of "stored" acidity and ferric iron which must first be flushed from the system. Lag time can, of course, be reduced by application of an alkaline solution or by adding a soluble source of alkalinity.

The concentration of detergent in the drainage decreased to less than 10 ppm after approximately 11 months of treatment. Acidity gradually increased as bacterial inhibition lessened, so that after approximately 18 months, acidity was once again greater in the treated section than in the control (figure 8).

We have recently initiated full-scale field tests at sites where suitable experimental control exists, and anticipate treating 5 to 10 active and abandoned surface mines and refuse piles by the end of 1981. We have selected sites having active operations which can be divided into at least two sections, only one of which will be treated, thus providing experimental control. At inactive sources of acid drainage, we are using sites that will not be otherwise disturbed or treated and that have a consistent chemical history that can be used for comparison.

Our first full-scale field test was initiated at a 2 hm² (5 acre) revegetated surface mine near Buckhannon, WV. Approximately 180 kg of rubber pellets and 90 l of SLS solution were applied in November 1980, as a conservative initial treatment. No surfactant was detected in the acid spring drainage, so a second treatment was planned. However, a percolation test was conducted after we observed rainwater ponding on the surface; the results indicated virtually no percolation. Apparently, oxygenated water is being supplied through the highwall. No decision has been made yet on whether or not to excavate a trench at the highwall and apply the bactericide there.

A second field test has been initiated at a 4 hm² (10 acre) coal refuse pile near Beckley, WV. SLS solution has been applied at 180 mg/l by hydroseeder; we are currently waiting to see if any surfactant appears in the drainage. We plan to adjust the initial treatment until surfactant is detected or until acid production drops. Rubber pellets will be applied subsequently.

A third field test was initiated in early July 1981 in eastern Ohio on a well-studied inactive surface mine. The area was mined between 1968 and 1970, exposing a pyritiferous sandstone in a series of ridges and valleys. The area has been divided into seven hydrologic basins; tracers indicate flow-through times of 6 months to 2 years. Using a hydroseeder, one of the basins has been treated at a cost of about \$1,200/hm² (\$500/acre), with 30 pct SLS solution at a rate of approximately 170 l/hm² (180 gal/acre) and with rubber pellets at approximately 170 kg/hm² (150 lbs/acre). If further laboratory study indicates that this treatment is insufficient, additional rubber will be added. The other six basins will remain untreated for control.

Several other field sites will be treated during the summer and fall of 1981. These include an 36 hm² (88 acre) coal refuse pile near Wheeling, WV, a barren 40 hm² (100 acre) surface-mined area in Somerset County, PA, and an active 4 hm² (10 acre) coal refuse pile in Preston County, WV. Other potential sites for 1981 include two active contour mines in Preston County, WV, where drainage divides provide potential experimental control; two surface mines that are soon to be reclaimed and that have dividing rock drains; and one of two large neighboring refuse piles that receive material from the same preparation plant.

CONCLUSIONS

Controlled release of anionic surfactants can prove useful in augmenting revegetation of mined lands and in abating acid at refuse piles and operating surface mine spoil piles. The recently-initiated field tests will allow us to evaluate bacterial inhibition over a wide range of site conditions and to determine cost effectiveness and optimal application rates. Completion of the laboratory tests described in this paper will allow optimizing field methodology to achieve bacterial population control economically for 3 to 10 years. If acidity can be successfully reduced for that time interval, revegetation and re-establishment of a stable soil environment will be more successful and will, in turn, limit acid production in the future.

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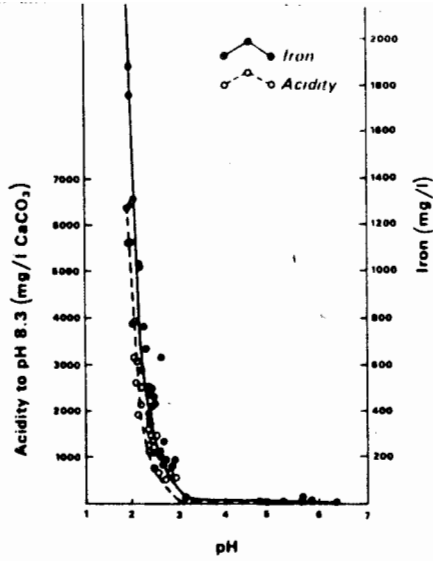


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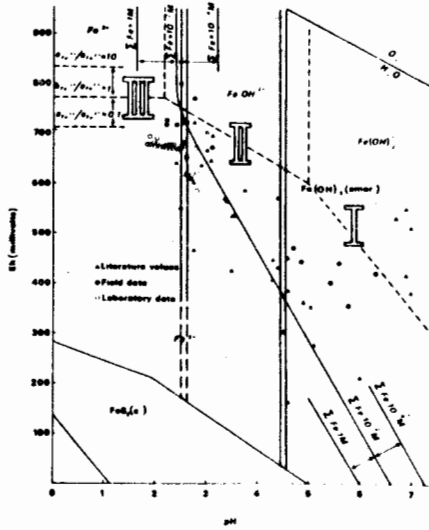


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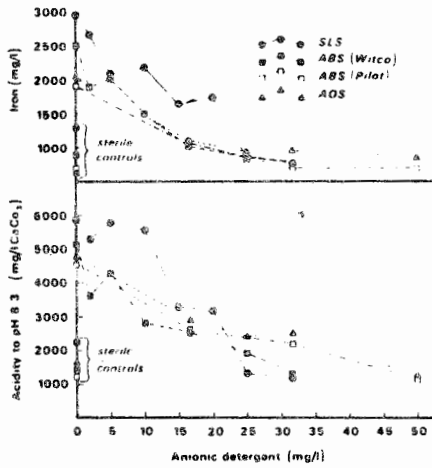


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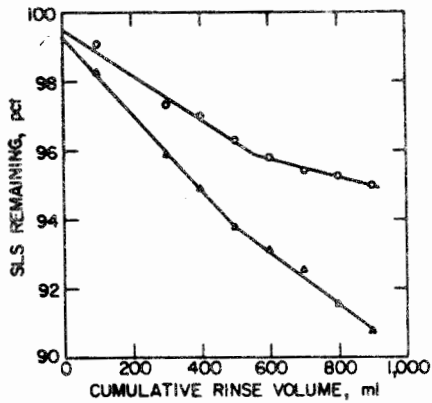


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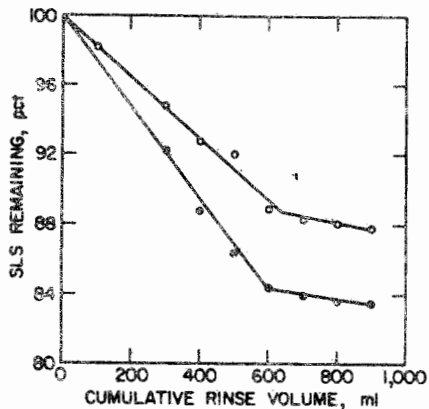


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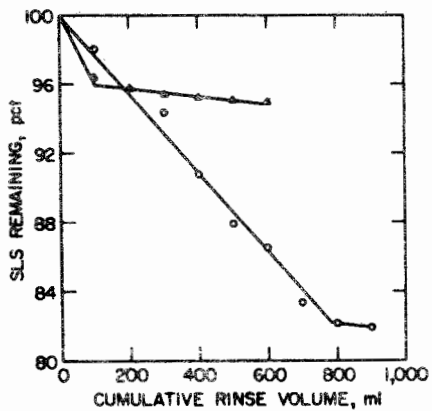


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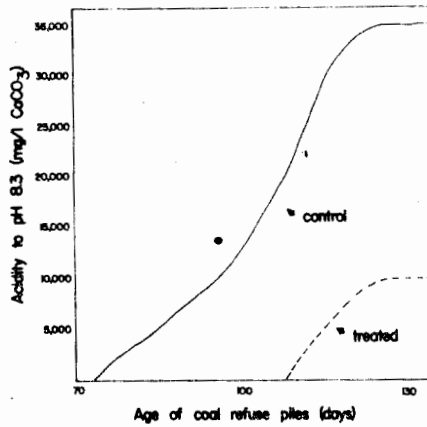


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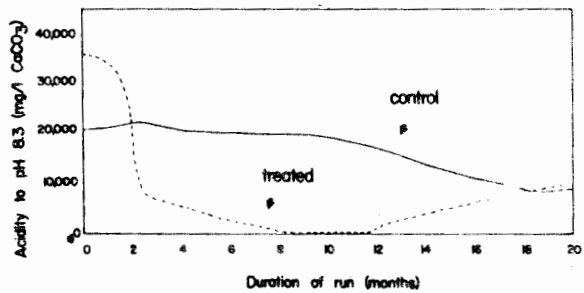


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