PROOF-OF-PRINCIPLE STUDIES FOR PASSIVE TREATMENT OF ACID ROCK DRAINAGE AND MILL TAILING SOLUTIONS FROM A GOLD OPERATION IN NEVADA[1]

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Abstract: Laboratory investigations were conducted to determine "in principle" whether passive treatment is a reasonable option for two water types produced by a surface gold operation in Nevada. One water was acid rock drainage (ARD) containing elevated arsenic and selenium. Arsenic was reduced first by allowing $Fe(OH)_3$ to form and then adsorbing anionic arsenic onto the positively charged precipitate. Then, all heavy metals, arsenic, and selenium were removed to within drinking water standards in bottle studies that promoted sulfate reduction at room temperature for 5 weeks. The second water type was underdrainage and seepage from a mill tailing pond containing elevated cyanide, nitrate, ammonia, copper, mercury, and selenium. The pH of the solutions was about 8, and the tailing underdrain solution contained greater concentrations of contaminants than the seepage. Both anaerobic and aerobic static tests were conducted at room temperature for 5 weeks. The aerobic tests were successful in reducing cyanide, ammonia, nitrate, copper, mercury, and selenium. Arsenic was reduced if soil as well as algae was present. In the anaerobic tests, cyanide, nitrate, copper, mercury, and selenium were reduced, whereas ammonia and arsenic were either unaffected or increased. The results of the laboratory tests were used to design a settling pond-anaerobic-aerobic passive system for the acid rock drainage and an aerobic passive system for the underdrainage and seepage. The pilot-scale ARD system and the aerobic algal pond for the tailing water were recently constructed to confirm the laboratory results.

Additional Key Words: Heavy metals, arsenic, selenium, cyanide, nitrate, ammonia, passive treatment, constructed wetlands

Introduction

Passive treatment methods that rely on aerobic reactions have been used to treat acid rock drainages (ARD) from coal mines. When the water has an initial pH greater than 5.5 and also has net alkalinity, aerobic constructed wetlands that precipitate iron hydroxides can be effective (Wildeman, Brodie, and Gusek 1993). Some ARD, especially that associated with metal mines, has a lower pH and higher concentrations of iron and other heavy metals. This type of water requires anaerobic passive treatment (Hedin and Nairn 1990). The anaerobic systems rely on bacterially mediated precipitation of metal sulfides.

Currently, little information exists on the ability of passive systems to treat (1) ARD that contains arsenic and/or selenium in concentrations above the Federal drinking water standards or (2) alkaline pH drainage containing low levels of cyanide. Both of these water types have been associated with gold mining operations in the arid regions of the Western United States. High arsenic and selenium concentrations in water are common in arid regions because these elements are more soluble in the natural alkaline and saline waters associated with this type of environment (Jacobs 1989). Additionally, alteration of rocks associated with gold mineralization often produces elevated concentrations of arsenic in ground water. Cyanide and alkaline waters are the products of mineral processing methods that are used to extract gold from ore (van Zyl 1984).

This study presents the results of two laboratory investigations conducted to determine "in principle" whether passive treatment is a reasonable option for these two water types. Both water types were collected from a surface gold mine operation in Nevada where closure is being considered within the decade. The laboratory investigations were the first stage in the investigation of passive systems suitable for closure.

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[1]Paper Presented at the International Land Reclamation and Mine Drainage Conference and the Third International Conference on the Abatement of Acidic Drainage, Pittsburgh, PA, April 24 - 29, 1994.

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Proceedings America Society of Mining and Reclamation, 1994 pp 387-394
DOI: 10.21000/JASMR94020387

https://doi.org/10.21000/JASMR94020387

Table 1. Substrates and waters used for the laboratory study.

Substrates used for waste rock drainage		Substrates used for tailings drainage	
Code	**Composition**	**Code**	**Composition**
MLI-B	3/4 M, 1/4 L, 10 g I-B	SI-T	1/2 S, 1/2 I-T
SMLI-B	1/2 S, 1/3 M, 1/6 L	SMI-T	1/2 S, 5/12 M, 1/12 I-T
L	100% L	I-T	100% I-T
H-M	100% H-M	A	100% A
H-J	100% H-J		C-M
100%	C-M	AS	1/3 A, 2/3 S

SUBSTRATE KEY

Substrate Code	**Inoculum Code**
M = Manure	I-B = Inoculum from Big Five Wetland
l = Limestone	H-M = Horse Manure
S = Mine site soil	H-J = Horse juice
A = Algal solution from seepage pond	C-M = Cow manure
	I-T = Anaerobic slime from seepage pond

WATER KEY

Waste Rock-Drainage		Tailings Water	
Code	Description	Code	Description
#1	Drainage from old waste rock dump issuing from Pipe 1, pH = 2.7	CP	Water from seepage pond taken from the pump outlet to insure a mixture of all sources
#2	Drainage from toe of waste rock pile issuing from Pipe 2, pH = 4.1	U	Water from tailings underdrain collected at the inlet pump into the storage pond
#3	Drainage from new waste rock pile issuing from Pipe 3, pH = 4.7	MX	A mix of 100 volumes of underdrain water to 15 volumes of seepage pond water to simulate water stored in the lined pond
1-2-3	A mix of waters from pipes 1, 2 and 3 in a ratio of 3-1: 3-2: 1-1 to simulate fresh pond water		

In the first case, water tested is ARD from a waste rock pile that contains water with concentrations of iron, arsenic, and selenium as high as 600, 10, and 1 mg/L, respectively. This drainage is presently being collected in a lined settling pond and is subsequently pumped to an alkaline milling process circuit. The drainage is intercepted and transported to the settling pond in three pipes, in volume ratios of three parts pipe 1, three parts pipe 2, and one part pipe 3. The ARD from pipe 1 had the lowest pH and a distinctive red color that was suspended $Fe(OH)_3$ that settled out upon aging for approximately 16 h.

The other water type is underdrainage and seepage from a drained mill tailing facility receiving cyanide effluents. The underdrainage contains concentrations of cyanide, nitrate, ammonia, mercury, arsenic, and selenium in excess of Federal drinking water standards. The seepage has lower concentrations of cyanide, associated nitrogen compounds, and metals than the underdrainage, which suggests that it has undergone some treatment by natural processes. The underdrainage and seepage are currently collected in ponds and reused in the gold extraction process.

The primary goals of the "proof-of-principle" experiments were (1) to determine the best types of microbial ecosystems to remove As and Se in both acidic and alkaline waters and (2) to determine whether aerobic, anaerobic, or a combination of processes is the most effective passive treatment method for the tailing drainages. Secondary objectives were to (1) determine local sources of sulfate-reducing bacteria (SRB) and bacteria and/or algae capable of

Table 2. Waste rock drainage test bottles - all bottles are anaerobic.

Bottle[1]	Substrate code	Substrate amount, g	Water Code	Water amount, mL	Inoculum	Inoculum amount, g
1	MLI-B	60	#1	60	I-B	10
2(1)	MLI-B	60	#1	60	I-B	10
3	MLI-B	40	#1	80	I-B	10
4	SMLI-B	60	#1	60	I-B	10
5	SMLI-B	40	#1	80	I-B	10
6	H-M	50	#1	60	H-M	50
7	H-J	60	#1	60	H-J	60
8	C-M	60	#1	60	C-M	60
9	L	30	#1	100	None used	NAP[2]
10	None used	NAP[2]	#1	100	None used	NAP
11	MLI-B	40	#3	80	I-B	10
12	SMLI-B	60	#3	60	I-B	10
13	SMLI-B	40	#3	80	I-B	10
14(13)	SMLI-B	40	#3	80	I-B	10
15	None used	NAP	#3	100	None used	NAP
16	L	30	#3	100	None used	NAP
17	MLI-B	40	1-2-3	80	I-B	10
18(17)	MLI-B	40	1-2-3	80	I-B	10
19	SMLI-B	60	1-2-3	60	I-B	10
20	SMLI-B	40	1-2-3	80	I-B	10
21	H-M	50	1-2-3	60	H-M	50
22	H-J	60	1-2-3	60	H-J	60
23	C-M	60	1-2-3	60	C-M	60
24	None used	NAP	1-2-3	100	None used	NAP

[1]Bottle Numbers in parentheses designate duplicates. [2]NAP means not applicable

degrading cyanide and associated nitrogen species and (2) develop a mix of organic and inorganic materials (labeled "substrates") that would promote the appropriate microbial activity. The anaerobic tests were patterned after those performed by Reynolds et al. (1991), and the aerobic tests were patterned after those of Duggan et al. (1992).

Collection of Materials

Waste rock ARD and tailing drainages, materials for substrates, and inoculum candidates were collected in May 1992. Individual samples of ARD were collected from each of the three pipes draining into the settling pond. Because any future passive treatment system would treat the settling pond water, a 3-3-1 by volume composite of the ARD waters from the three pipes was made (labeled "1-2-3") and used in some of the bottle tests. Tailing underdrain water and seepage from the tailing dam were also sampled individually. Currently, the ratio of tailing underdrain water to seepage is about 100 to 15. One closure option is to treat this mixed water. Accordingly, a mix of 100 volumes of underdrain water to 15 volumes of seepage water was made (MX) and used in some of the bottle tests.

Table 1 provides a key to the various substrate and inoculum materials that were gathered. Inocula for SRB were collected at a cattle feedlot. Typically, the best sources for SRB bacteria are fresh, wet manure and the mucky places in a feedlot or farm. On this feedlot, all the manure was quite dry. The only mucky place was the area around the watering trough for the horses. In table 1, this muck is labeled "horse juice."

The tailing seepage pond contained abundant floating algae and a blackish anaerobic slime in the sediments. These materials were collected to serve as aerobic and anaerobic inocula, respectively, for the tailing drainage tests because they were expected to contain microbes already acclimated to cyanide and metals (Duggan, et al. 1992, Wildeman, et al. 1993). For most of the other bottles in the anaerobic study, SRB inoculum from the Big Five Pilot Wetlands (Wildeman, Brodie, and Gusek 1993) was used.

Table 3. Tailing water test bottles.

Bottle[1]	Substrate code	Substrate amount	Water code	Water amount, mL	Inoculum	Inoculum amount	Aerobic
25	SI-T	60 g	CP	60	I-T	30 g	No
26	SMI-T	50 g	CP	80	I-T	5 g	No
27(26)	SMI-T	50 g	CP	80	I-T	5 g	No
28	None used	NAP[2]	CP	100	None used	NAP	No
29	I-T	30 g	CP	100	I-T	30 g	No
30	SI-T	60 g	U	60	I-T	30 g	No
31(30)	SI-T	60 g	U	60	I-T	30 g	No
32	SMI-T	50 g	U	80	I-T	5 g	No
33	None used	NAP	U	100	None used	NAP	NAP
34	I-T	30 g	U	100	I-T	30 g	No
35	SI-T	60 g	MX	60	I-T	30 g	No
36	SMI-T	50 g	MX	80	I-T	5 g	No
37	None used	NAP	MX	100	None used	NAP	NAP
38	A	30 mL	CP	100	A	30 mL	Yes
39	AS	30 g	CP	100	A	15 mL	Yes
40	None used	NAP	CP	100	None used	NAP	NAP
41	A	30 mL	U	100	A	30 mL	Yes
42(41)	A	30 mL	U	100	A	30 mL	Yes
43	AS	30 g	U	100	A	15 mL	Yes
44	None used	NAP	U	100	None used	NAP	NAP
45	A	30 mL	MX	100	A	30 mL	Yes
46	AS	30 g	MX	100	A	15 mL	Yes
47	AS	30 g	MX	100	A	15 mL	Yes
48	None used	NAP	MX	100	None used	NAP	NAP

[1]Bottle numbers in parentheses designate duplicates. [2]NAP means not applicable

In both anaerobic and aerobic laboratory studies, a source of neutral to alkaline soil or limestone is needed to help raise pH, control hydraulic permeability, and serve as adsorption sites for bacteria and contaminants (Bolis et al. 1992, Duggan et al. 1992). Alkaline soil from the mine site and limestone from the nearest quarry were collected to serve these functions.

Experimental Design

The collected waters and materials were shipped overnight to the Colorado School of Mines, and the experiments were set up the following day. Splits of the water samples were sent to Core Laboratories for chemical analysis. Materials larger than 10 mesh were removed from the substrate candidates, and mixes of the material were made in the combinations given in table 1. The codes for the substrates and waters are given in table 1.

The anaerobic tests were conducted in 150 mL screwcap culture bottles. A small amount of head space was left in the bottles to accommodate gases evolved during the experiment because this minimizes solution loss. The bottles were incubated for 5 weeks in boxes at ambient temperatures in the laboratory. The aerobic tests were conducted in 500-mL wide-mouthed erlenmeyer flasks that are covered with 250-mL beakers to allow gas exchange. The bottles were incubated for 5 weeks on a sunny window ledge at ambient temperatures in the laboratory. The protocol for the ARD experiments is given in table 2. The protocol for the tailing drainage experiments is given in table 3.

All bottles except 9 and 16 were unsealed to allow access to the solutions during the test period. Color, odor, pH, and Eh were recorded for each bottle once per week. Bottles 9 and 16 were sealed to ensure that atmospheric oxygen did not invade the systems and cause armoring of the limestone. Spot colorimetric tests on a few drops of solution were conducted using HACH kit reagents. The spot tests were conducted once during the course of the experiment for Fe and Cu on both water types, and twice during the experiment for cyanide on the tailing drainages. At the end of the experiments, quantitative determinations were made on representative bottles.

Table 4. Chemistry of waste rock drainages and test bottles after 5 weeks of incubation. All concentrations are in mg/L for unfiltered (total) water.

Bottle	**Original**	**1**	**2**	**3**	**10**	**Original**	**20**	**24**	**Water Stds**
Water	Pipe 1	Pipe 1	Pipe 1	Pipe 1	Pipe 1	1+2+3	1+2+3	1+2+3	NAP[1]
Substrate	NAP	MLI-B	MLI-B	MLI-B	None	NAP	SMLI-B	None	NAP
Sulfate	3370	5360	5130	4350	3850	2160	2350	2070	NAP
Arsenic	12	0.4	0.4	0.4	3	2.7	0.3	0.3	0.05
Cadmium	0.17	0.003	<0.003	<0.003	0.06	0.066	<0.003	0.04	0.01
Chromium	0.50	<0.05	<0.05	<0.05	0.5	0.20	<0.05	0.20	0.05
Copper	18	0.7	1.1	0.55	32	7.4	0.3	15	1.0
Iron	620	4.8	3.6	4.3	422	200	2.8	131	0.3
Manganese	12	1.7	1.4	1.8	11	6.7	5.2	6.4	0.05
Selenium	1.0	<0.01	<0.01	<0.01	<0.01	0.3	<0.01	0.01	0.01
Zinc	2.9	0.2	0.15	0.05	5.4	1.8	<0.05	3.0	5.0
pH	2.5	7.6	7.5	7.4	2.1	2.4	7.1	1.9	6-9

[1]NAP means not applicable

Results and Discussion

Anaerobic ARD Experiments

All bottles that tested whether the local cow manure, horse manure, or "horse juice" materials (not shown) were suitable SRB inocula, had Eh values below -100 mV, black solids, and strong hydrogen sulfide odors, indicating that all three materials were good sources of SRB inoculum. The final pH of ARD test bottles that used soil from the site was somewhat lower than that of bottles with undiluted manure, but still above pH 7. In addition, the Eh in bottles with soil tended to decrease over time in a manner that was similar to that in bottles that did not contain soil. These results indicate that the soil can be used to provide hydraulic control within the substrate in the anaerobic passive treatment system.

Initial values for pipe 1 and the expected holding pond water and results for regulated constituents after 5 weeks of incubation are given in table 4 for selected tests. In all cases the analyses were performed on whole, unfiltered waters. Bottles 1 and 2 are duplicates and provide intra-experimental variability. Bottles 10 and 24, which are controls of pipe 1 and 1-2-3 water, respectively, were analyzed to determine whether 5 weeks of iron hydroxide settling causes significant decreases in the concentration of other metals. Bottle 20 simulates the expected holding pond (1-2-3) water to be treated with a mixture of sand, manure, and limestone, the probable substrate of choice.

The results indicated that the pH is raised from around 2 to above 7 and concentrations of cadmium, chromium, selenium, and zinc are reduced to within drinking water standards in all analyzed bottles. Iron and arsenic were reduced by over 95 %, but were not brought to within drinking water standards. Manganese was reduced by 50 to 80 %. Typically, manganese is the most difficult element to remove from ARD (Duggan et al. 1992). These results suggest that the anaerobic system will produce significant reduction of contaminants. However, to meet water quality standards, an aerobic polishing stage may be necessary after the anaerobic treatment.

Analysis of the aged water from pipe 1 (bottle 10) and the 1-2-3 mix (bottle 24), when compared with the original waters, shows that the concentration of Fe is reduced by 30 %. Also, the pH of these waters dropped upon storage, suggesting that Fe(III) hydrolyzed and precipitated from solution. Concomitantly, arsenic in aged pipe 1 water was reduced from 12 to 3 mg/L and in the simulated holding pond water from 2.7 to 0.2 mg/L. In addition, selenium was reduced to less that 0.01 mg/L in both waters. These elements occur in the water as the oxyanions arsenate and selenate. At the low pH of the ARD, the ferric hydroxide precipitate has a high positive surface charge and readily adsorbs the arsenate and selenate anions. Removal of selenium to below drinking water standards and the significant reduction in arsenic makes containment of the ARD in a holding pond a reasonable first step in passive water treatment.

Table 5. Chemistry of tailing waters and test bottles after five weeks of incubation. All concentrations are in mg/L for unfiltered (total) water.

Bottle	Original	30	31	32	41	43	Original	26	39	46	Water Stds.
Water	Underdrain	Underdrain	Underdrain	Underdrain	Underdrain	Underdrain	Seep	Seep	Seep	MX	NAP[1]
Aerobic	NAP	No	No	No	Yes	Yes	NAP	No	Yes	Yes	NAP
Substrate	NAP	SI-T	SI-T	SMI-T	A	AS	NAP	SMI-T	AS	AS	NAP
Cyanide-Total	<4.0	<0.2	<0.2	<0.2	<0.2	<0.2	0.10	<0.2	<0.2	<0.2	0.2
Cyanide, WAD	4	<0.2	<0.2	<0.2	<0.2	<0.2	0.06	<0.2	<0.2	<0.2	0.2
Ammonia (as N)	14	16	11	0.44	0.27	0.08	2.1	8.6	0.19	0.20	-
Nitrate and Nitrite (as N)	19	1.0	1.1	1.9	0.31	0.73	8.3	2.2	0.31	0.60	-
Sulfate	400	15	16	66	478	430	390	210	391	450	250
Thiocyanate	14	<2.5	<2.5	4.9	0.4	<0.25	<0.1	4.4	<0.25	<0.25	-
Arsenic	0.09	0.17	0.15	0.17	0.11	<0.05	<0.05	-	-	<0.05	0.05
Copper	3.3	<0.05	<0.05	0.30	<0.05	<0.05	0.06	-	-	<0.05	1.0
Iron	0.05	1.8	2.5	1.2	<0.1	<0.1	<0.03	-	-	<0.1	0.3
Mercury	0.27	<0.002	<0.002	<0.002	<0.002	<0.002	0.004	<0.002	<0.002	<0.002	0.002
Manganese	0.47	7.6	2.0	2.2	2.15	0.15	1.6	1.35	0.45	0.75	0.05
Selenium	0.2	<0.01	<0.01	<0.01	<0.01	<0.01	<0.1	<0.01	<0.01	<0.01	0.01
pH	8.0	6.6	6.5	6.5	7.5	7.0	7.3	7.1	7.5	7.5	6-9

[1]NAP means not applicable.

Tailing Drainage Experiments

Cyanide, related nitrogen species, and other regulated constituents are present in the underdrain in higher concentrations than in the seepage. Thus, the tailing drainage experiments concentrated on this water and the MX water. The results showed that cyanide was destroyed to below 0.2 mg/L in all bottles within 4 weeks. These results indicate that either an anaerobic or aerobic system will destroy cyanide and confirm previous results on anaerobic tests reported by Filas and Wildeman (1992).

Bottles 30, 31, and 32 tested the various anaerobic substrates, and bottles 41 and 43 tested aerobic substrates with underdrainage water. Bottles 26 and 39 tested anaerobic and aerobic substrates, respectively, with the tailing seepage (CP). Bottles 43 and 46 tested the same substrates with the underdrainage water and the mix of seepage pond and underdrainage water (MX). The results for constituents of interest are presented in table 5. For comparison, the analytical results for the original waters and solution mixes are given. In all cases the analyses were performed on whole, unfiltered waters.

Anaerobic Experiments. The black sludge sampled from the seepage pond proved to be an excellent source of SRB for anaerobic passive treatment of tailing drainages. After 1 week, some of the anaerobic bottles that contained the tailing sludge inoculum had negative Eh's and smelled of hydrogen sulfide. The concentration of sulfate was also reduced, confirming that bacterial sulfate reduction was operating. At the cyanide concentrations in these waters, the SRB were not killed. From the results of experiments that studied a tailings solution containing much higher concentrations of total cyanide, Filas and Wildeman (1992) suggested that, below a cyanide concentration of about 10 mg/L, sulfate reduction was possible. Only a small supply of this black, pond sludge exists. If a large anaerobic system were built using this inoculum, the material would have to be mixed with other sources and incubated for a period of time to produce an adequate supply.

The results for nitrogen species were mixed. As discussed previously, cyanide was removed. Thiocyanate was also removed. The ammonia concentration remained high, which was expected in an anaerobic treatment system. The nitrate plus nitrite concentrations were significantly reduced. They were probably converted to nitrogen gas because the concentration of ammonia was not appreciably changed.

Copper, mercury, and selenium were reduced to below drinking water standards. Iron, manganese, and arsenic, which had not been completely removed in the anaerobic ARD tests, were also not reduced to below drinking water standards in the tailing drainages. In fact, these elements were at higher concentrations in the final solutions than in the original underdrain water. It appears that the anaerobic conditions caused dissolution of these elements from the soil, manure, and/or inoculum that were used in the substrate mixes. Apparently, an equilibrium between solid phases and the solutions has been established in these bottles.

These anaerobic test results indicate that an anaerobic passive system will not remove all the contaminants to Federal water quality standards.

Aerobic Experiments. Both the algae alone and the soil-algae mix reduced ammonia, nitrate plus nitrite, and thiocyanate to concentrations that were below the results for the anaerobic experiments. The qualitative tests conducted during the experiment indicated that the concentration of cyanide decreased faster in the bottles with soil and algae (AS) than in the bottles with algae alone (A). At the end of the experiment, the Eh's in those bottles containing only algae (41, 42, 45) had fallen significantly below those of the other aerobic bottles. This decrease in Eh may be the result of death and decay of the algae and suggests that, if an algal pond were not designed properly, the water in the pond could turn anoxic. This phenomenon exists in the current seepage pond at the site, where buried dead algae provided the anaerobic inoculum.

The results of the qualitative tests suggested that the soil-algae mixture functioned better than the algae alone. As reported in table 5, the concentrations of constituents confirm this result. The soil-algae mixture reduced all contaminants except manganese in the underdrain water and in the underdrain-seepage mixture to below drinking water standard concentrations. As seen from bottle 41, algae alone did not reduce arsenic concentrations. Based on the laboratory studies, an algal system containing soil would be the best passive system for removal of all the constituents in the tailing water.

Design Conclusions

For ARD waters containing high concentrations of arsenic and/or selenium, anaerobic passive treatment is possible "in-principle". A holding pond prior to the anaerobic cell would remove substantial quantities of both anions before the water entered the sulfate-reducing treatment system. The anaerobic cell may not remove all constituents to drinking water quality standards. If necessary, an aerobic polishing system can be installed after the sulfate-reducing system to remove these contaminants.

For tailing waters that contain moderate concentrations of cyanide and other metals, the laboratory studies suggest that the best passive system would be an algal pond containing sandy soil. This system removes iron, manganese, and arsenic, as well as all cyanide and its degradation products. Also, such an aerobic system would appear to be a good candidate for the polishing of iron, manganese, and arsenic from the waters exiting the sulfate-reducing anaerobic cell. A possible design for such a system is given by Wildeman et al. (1993).

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