Field Experiment on Iron and Aluminum Removal from Acid Mine Drainage Using an Apatite Drain System

Jung-Chan Choi* and Terry R. West**

ABSTRACT: An apatite drain was constructed on September 30, 1994 at the Green Valley Abandoned Coal Mine site near Terre Haute in west central Indiana. The primary objective of this experiment is to evaluate the long-term ability of the apatite drain to mitigate acid mine drainage (AMD) under field conditions. The drain 9 m long, 3.3 m wide, and 0.75 m deep, contain 95 mm to No. 30 mesh-size apatite ore (francolite) and receive AMD seepage from reclaimed gob piles, and designed according to the laboratory testing. The apatite drain was covered with limestone riprap and filter fabric to protect the drainage system from stormwater and siltation. The drain consists of about 50 metric tons of apatite ore obtained from a phosphate mine in Florida. A gabion structure was constructed downstream of the apatite drain to create a settling pond to collect precipitates. Apatite effectively removed iron up to 4,200 mg/l, aluminum up to 830 mg/l and sulfate up to 13,430 mg/l. The pH was nearly constant for the influent and effluent, ranging between 3.1 and 4.3. Flow rate measured at the gabion structure ranged from 3 to 4.5 l/m. Precipitates of iron and aluminum phosphate (yellow and white suspendid solids) continued to accumulate in the settling pond.

INTRODUCTION

Disturbance of coal strata during mining operations accelerates the natural deterioration of water quality by increasing the surface area of reactive minerals exposed to the atmosphere, hydrosphere, and biosphere.

Changes in the length of time that water spends in the groundwater, or changes in the physical state of the aquifer containing the water can results in a change in water quality. The most dramatic effect is the production of Acid Mine Drainage (AMD). The AMD is defined as water discharged from active, inactive, or abandoned mine, and from areas affected by surface mining, that has a pH less than 6.0 and a total acidity that exceeds the total alkalinity (Code of Federal Regulation, 1986).

AMD is resulted from oxidation of framboidal pyrite in the refuse piles. Pyrite oxidation occurs in the presence of water and oxygen, producing ferrous iron and sulfuric acid. The AMD problems are not unique to the United States. These are also known in Australia, Japan, Korea, Russia, and South Africa. Deterioration of streamwater quality can become so severe that, in some cases, the affected reaches of

STATEMENT OF PROBLEM

Green Valley, an Abandoned Mine Lands (AML) site, is located about 6.4 km northwest of West Terre Haute and about 0.6 km east of the Ina-Illinois State Line, in Vigo County, Indiana (See Figure 1). Coal mining activity occurred from 1948 to 1963 to mine Indiana Coal III and Coal V. Gob from underground mining operations and tailings from coal processing operations were abandoned without any site reclamation (See Figure 2). The gob piles are composed of shale, pyrite, coal, other associated clay and rock fragment which were separated from usable coal during coal processing, tailing ponds usually consist of water deposited accumulations of fine gravel, sand, silt and clay size fragments or particles of coal and other materials that are removed from coal in the wash or preparation plant.

A primary environmental concern at the Green Valley site is acid mine drainage resulting from oxidation of pyrite in the refuse piles. More than 76,

streams are totally devoid ofmicroscopic life. The length of a stream that is significantly affected by mine drainage depends upon the amount, concentration, and distribution of mine drainage inflows to the stream and upon the quantity of unaffected inflows from upstream areas and from tributaries downstream (Powell, 1988).

^{*} Water Resources Reearch Institute, KOWACO, 46-1 Jonmin-Dong, Yusung-Gu, Taejon 305-390, Korea

^{**} Department of Earth and Atmospheric Sciences, Purdue University, W. Lafayette, In 47907, U.S.A

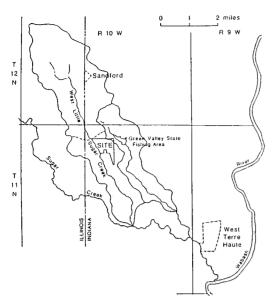


Fig. 1. Location map of the green Valley Mine site.

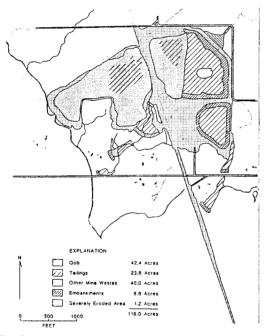


Fig. 2. Types of coal refuse materials at the Green Valley site.

000 cubic meters of this mine refuse material piled on the surface at the site, has produced acidic runoff, precipitates of metallic compounds (primarily iron and aluminum) and sedimentation such that no aquatic life exist for a distance of about 5.8 km downstream from the site in the west little sugar

Creek. Sugar Creek is also severely polluted for a stream distance of about 8.6 km, to its confluence with the Wabash River. The Green Valley Mine site was classified as a third place priority ranking for worst AMD conditions (Geoscience Research Associates, Inc., 1985).

Seepage from gob piles yields the worst water quality on the site. Groundwater emerges as seepage along the slope. During dry periods, salts precipitates at the ground surface. These surface salts (mostly iron sulfate hydrate) appear as powdery, white and platy yellow minerals, along with small green crystals. Because salts are highly soluble, surface runoff acidifies as water dissolves the salts and flow away across the ground. Ferrous iron forms the dominant species of the Fe ions. The Ca²⁺ category shows an almost constant concentration because Ca²⁺ in acid mine water, together with high sulfate concentrations from pyrite oxidation, results in a saturated condition with respect to gypsum.

OBJECTIVE OF STUDY

Conventional AMD treatment is simple process. The water is neutralized, typically to a pH of 8 to 9, and then aerated to oxidize the iron to the Fe³⁺ state, causing precipitation of Fe(OH)₃ (yellow boy sludge). The water then separated from the sludge in a series of settling basins or ponds and discharged. Alkaline reagents that may be used in conventional AMD treatment are lime (CaO), limestone (CaCO₃), sodium hydroxide (NaOH), and sodium carbonate (Na₂CO₃) (Ackman and Kleinmann, 1985). However, facilities cost and maintenance is high and continuous. It is obvious that conventional treatment of run-off and seepage is not a long-term solution, because formation of AMD can persist for many decades.

Staged, aerobic constructed wetlands offer a potential low-cost, natural, low-maintenance, and longterm alternative to conventional treatment of acid mine drainage. The Tennessee Valley Authority has a 75% sucess rate of aerobic wetlands consistently meeting compliance without chemical treatment. Some constructed wetlands for AMD control by the TVA have been enhanced by the addition of Anoxic Limestone Drains (ALD) at the initial stage of the treatment system. The ALD, which consists of a shallow, limestone-filled trench excavated into mine backfill and sealed from the atmosphere, passively introduces buffering capacity, as alkalinity, into the AMD (Brodie, 1991). Dissolution of limestone added 100~200 mg/l of alkalinity as CaCO₃ to the water (Kleimann et al., 1990). However, not all water is suitable for pretreatment with the ALD. The primary chemical factors believed to limit the utility of the ALD are the presence of ferric iron, aluminum and dissolved oxygen (DO). When acidic water containing any ferric iron or aluminum contact limestone, metal hydroxides will form. No oxygen is necessary. Ferric hydroxide can armor the limestone, limiting its further dissolution. Whether aluminum hydroxides armor limestone has not been determined. The buildup of both precipitates within the ALD can eventually decrease the drain permeability and cause plugging. The presence of dissolved oxygen in mine water will promote the oxidation of ferrous iron to ferric iron within the ALD, and thus potentially cause armoring and plugging.

Unfortunately, AMD from seeps of the Green Valley Mine site shows high concentrations of ferric iron and aluminum and very low pH because this gob pile has weathered for several decades. Therefore, the use of an ALD system is not appropriate because iron and aluminum hydroxides will coat the limestone surface and eliminate further dissolution of limestone, thus prevent neutralization.

For municipal wastewater treatment plants, aluminum and iron salts such as alum and ferric chloride, are used for phosphate removal. This concept can be applied to AMD. It seems that naturally occurring phosphate mineral can be used for iron and aluminum removal from AMD. An experimental laboratory test was performed to evaluate the effectiveness of apatite from Florida in the treatment of AMD under aerobic condition during August and September, 1991. Acid mine water was collected from a seep at the Friar Tuck Abandoned Mine site in southwestern Indiana. Using different flow rate, results showed that apatite effectively removed iron up to 1,200 mg/l, aluminum up to 800 mg/l and sulfate up to 8,600 mg/l in three weeks. In addition, the pH increased to values as high as 3.2 in the effluent water from a pH of the influent water ranging from 2.1 to 2.2. Removal of iron, aluminum, and sulfate as well as pH increase were inversely proportional to flow rates, ranging from 1.17×10^4 to 1.05×10^3 liter per minute per kg of apatite (Choi and West, 1995).

The primary objective of this field experiment is to evaluate the long-term effectiveness of the apatite drain on AMD under natural condition.

APATITE MATERIAL

Apatite, $Ca_{10}(PO_4)6X_2(s)$, where X is typically OH or F, commonly shows a variable Ca/P ratio in most naturally occurring, poorly crystallized, forms. In

areas of highly organic productivity, such as shallow portions of the ocean, carbonate phosphate minerals are deposited (Stumm and Morgan, 1981). The apatite found in the phosphate deposits of Floida is francolite with an empirical end-member formula as follows; Ca_{10} -X-YNa_XMg_Y(PO4)_{6-Z}(CO₃)_ZF_{0.4}ZF₂

Most apatite pebbles occur in the Bone Valley Formation. This formation consists of phosphate gravels and sand mixed with variable percentage of quartz sand. The most distinctive lithologic feature of this formation is the occurrence of large quantities of phosphate gravel (Scott, 1985). The recoverable phosphate grains range in size from 0.1 mm to as much as 30 cm in diameter. The Bone Valley Formation ranges in thickness from a feather edge to a maximum of about 15 m (Cathcart, 1989).

APATITE DRAIN DESIGN

A preliminary question to consider is under what conditions an apatite drain system is effective. The effectiveness of an apatite drain depends on pH, flow rate, and grain size. If an AMD site is selected for treatment with a apatite drain system, the pH and flow rate are almost constant for design purpose. In the regard of grain size, sand size apatite, which has a larger surface area per unit volume than does the pebble size. However, there is a possibility that the drain eventually stop reacting because it becomes plugged by gypsum precipitates, and the life of the system will be decreased if very small size of apatite is used. The selection of stream near West little Sugar Creek was proposed as the location for an apatite drain system because a gabion structure had been constructed downstream to create a settling pond to collect sediment. According to the results of chemical analysis for the stream water, 6,600 mg/l of iron and 1,200 mg/l of aluminum are available for removal from AMD. But multiple stages of treatment system are considered because the concentrations of iron and aluminum were high. It is assumed that the total chemical loading is constant. In other words, the concentrations of Fe and Al multiplied by amount of discharge is a constant. Based on the laboratory experiment, it was assumed that the bulk density of apatite is about 2.0 g/cm and porosity of the apatite layer is about 50%. Therefore, 1 metric ton of apatite occupies 1 m³ volume. Preliminary design specifications are considered as follows: Inlet water Quality: Al 1,200 mg/l, total Fe 6,600 mg/l (ferric iron 200 mg/l)

Apatite quantity: 40 metric tons

Flow rate: $(2 \text{ gpm} \times 3.785 \text{ } l/\text{gal})/(40 \text{ tons} \times 1000 \text{ kg/l})=1.89 \times 10^{-4} \text{ } l/\text{min/kg}$

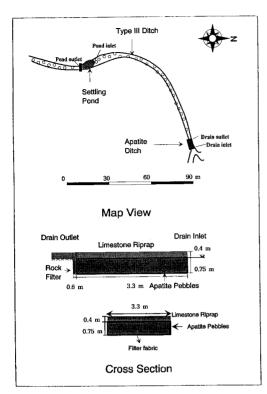


Fig. 3. Schematic of an apatite drain design.

Expected sulfate removal= $18890 + 6730 \log (1.89 \times 10^4) = 6170 \text{ mg/}l \text{ (See Choi and West, 1995)}$

Expected iron removal= $1460 + 620 \log (1.89 \times 10^{-4})$ =850 mg/l

Expected aluminum removal= $1240 + 460 \log (1.89 \times 10^4) = 470 \text{ mg/}l$

Retention time in apatite drain= $(40 \text{ m}^3 \times 0.5 \times 1000 \text{ l/m}^3)/(2 \text{ gpm} \times 3.785 \text{ l/gal} \times 60 \text{ min/hour})=44 \text{ hrs}$

The apatite drain was constructed on the 30th of September, 1994. The drain which is 9 m long, 3.3 m wide, and 0.75 m deep, contains 95 mm to No. 30 mesh size apatite which came from the Payne Creek Mine in central Florida. The apatite drain was covered with a 0.4 m depth limestone riprap and filter fabric to protect the drainage system from stormwater and siltation (See Figure 3). Limestone riprap was also used as a rock filter at the end of the apatite drain.

METHODOLOGY

Acid mine water was collected from the inlet of the apatite drain, the outlet of the apatite drain, inlet of the settling pond, and outlet of the settling pond. However, some water sampling opportunities were missed during first two months period of study. Each water sample was collected in a one liter plastic bottle and transported to the Indiana Geological Survey for analysis.

All measurements were made in the Geochemistry Laboratory of the Indiana Geological Survey. For pH measurements, an Orion model 91~56 electrode was used in conjunction with an Orion model 231 pH/mV/ATC portable meter. Eh measurements were made with a Corning combination platinum/calomel electrode (model 476064) in conjunction with an Orion model 231 pH/mV/ATC portable meter set for millivolt(mV) reading. Alkalinity was determined by manually titrating a known volume of each sample to a pH endpoint of 4.5, using standardized 0.02 N sulfuric acid(H₂SO₄). Water samples were prepared for analysis by filtering through a 0.2 μm filter.

The acidity measurements were coducted as follows: First the sample was titrated with standardized H₂SO₄ to below a pH of 4.00. Then, concentrated hydrogen peroxide was added, the sample heated to boiling, and allowed to cool to room temperature. Titrations were carried out using 0.02 N NaOH to a pH endpoint of 8.3. The anions were analyzed on a Wescan model 261, dual column, unsuppressed Ion Analyzer with conductivity detector. An acidified aliquot was analyzed for the cations by inductively coupled plasma atomic emission spectroscopy (ICP-AES) utilizing a Jarrell Ash model 975 Atomcom simultaneous instrument. In the case of ferrous iron, a potentiometric titration against a standard cerium ammonium nitrate solution was utilized and measurements were made using a Corning combination platinum-silver/silver chloride redox electrode (model 476080) and an Orion 960 Autotitration System programmed to utilize a second derivative method to detect the reaction endpoint (Branam and Harper, 1994).

RESULTS AND DISCUSSION

Water quality improvement has occurred during water collection periods using the apatite drain system except for storm water events. When the first water samples were collected from the apatite drain system on October 12, 1994, the limestone riprap covered stream channel was already coated.

According to the chemical analysis, 4,400 mg/l of iron, 1,190 mg/l of aluminum, and 13,900 mg/l of sulfate were removed in the section between drain inlet and the settling pond inlet. This suggest that the apatite fines reacted rapidly with AMD. When the

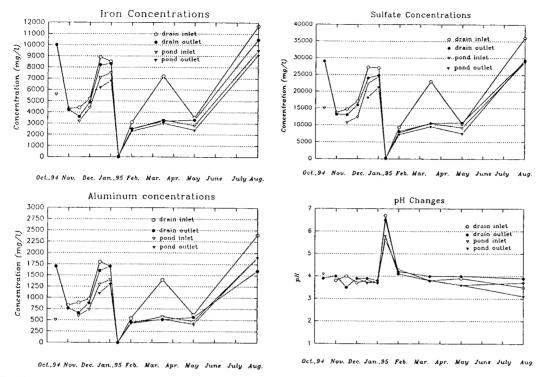


Fig. 4. Iron, aluminum and sulfate removals, and pH changes.

second water samples were taken from the system, the settling pond was filling with yellow and white precipitates and with olive gray clay.

The apatite drain system removed iron up to 4,200 mg/l, aluminum up to 830 mg/l, sulfate up to 13,430 mg/l, and acidity up to 8,900 mg/l during the nine month monitoring period (See Figure 4). The pH of untreated and treated water was almost constant, and ranged between 3.1 and 4.3. Results showed that other cations such as Ca²⁺, Mg²⁺, Mn_{tot}, Na⁺ had concentrations that were also almost constant (See Table 1).

The removal rates of iron ranged between 20.0 and 58.3 percent, and aluminum removal rates ranged between 20.8 and 58.6 percent. Sulfate between 18.3 and 58.4 percent (See showed similar removal rates ranging Figure 5). The variation of removal rates seems to results from the variation of flow rates. Flow rates measured at the gabion structure ranged 3.0 to 4.5 l/m.

According to the X-ray diffraction analysis, only gypsum was detected in the precipitates collected from the water in the settling pond. Therefore, it seems that the remaining precipitates consist of amorphous phosphates. The MINTEQA2 program was applied to define the mechanism of reaction

between AMD and apatite. MINTEQA2 is a geochemical equilibrium specification model capableof computing equilibrium among the dissolved, adsorbed, solid, and gas phases in an environmental setting (Allison, et al., 1991). The model can be used to calculate the equilibrium composition of dilute aqueous solutions in the laboratory or in natural aqueous system. It can be used to calculate the mass distribution between the dissolved, adsorbed, and multiple solid phases under a variety of conditions including a gas phase with constant pressure. According to the result of MINTEOA2,

The main conclusions are as follows:

- Ferric iron was removed as strengite, and hematite was precipitate from solution.
- The concentration of ferrous ion in treated AMD was controlled by precipitation of iron phosphate (vivianite). The oxidation of ferrous to ferric iron was apparently inhibited by the low pH.=Aluminum appeared to be controlled by the solubility of AIOHSO₄ at low pH. Diaspore and alunite were other potential solid phases at the low pH
- Calcium and sulfate concentrations were controlled by precipitation of gypsum.

(l/gm)

1700 1700 1400 1300 Al⁺³ (mg/l) 970 880 740 1800 1600 1300 1100 0068 (l/gm) 41.0 41.0 37.0 35.0 Mn⁺² (mg/l) 38.0 35.0 31.0 23.0 20.0 21.0 21.0 17.0 27.0 27.0 25.0 8300 8200 7600 7000 5300 4900 7700 6800 5900 Fe⁺² (mg/l) 8500 4500 8500 8300 7500 6800 4400 8900 8200 Fe (total) (mg/l) 10000 5600 4300 4200 4400 4600 3200 700 6200 5200 4900 Na⁺¹ (mg/l) 150 140 130 110 Mg⁺² (mg/l) 490 470 440 450 50 50 500 470 480 490 490 510 510 420 420 420 27000 24800 24400 21300 27200 24000 22400 18200 17100 13700 14700 13100 SO₄-2 (mg/l) 1240 <100 <100 (l/gm) <100 Ü Table 1. Analyses of water samples related to the apatite drain. Acidity Alkalinity (equ. CaCO₃) 24000 25000 22000 21000 25000 22000 20000 17000 11000 14000 12000 10000 15000 13000 Eh (vs. SHE) (mV) Hd 12/28/94 12/28/94 12/28/94 12/14/94 12/14/94 12/28/94 11/1/94 11/16/94 11/16/94 11/30/94 11/30/94 11/30/94 12/14/94 2/14/94 11/16/94 Date GVA-A GVA-B GVA-C GVA-D GVA-A GVA-B GVA-C GVA-D GVA-B GVA-A GVA-B GVA-C GVA-B GVA-A GVA-B GVA-A GVA-C

Table 1. Countinued.

57	(mg/l)	23	2.0	14.0	13.0	12.0	13.0	30.0	13.0	14.0	14.0	17.0	15.0	14.0	12.0	14.0	27.0	35.0	33
Al ⁺³	(mg/l)	0.4	0.2	540	460	430	420	1400	510	580	520	620	260	470	410	2400	1600	1900	1900
Zn ⁺²	(l/gm)	0.0	0.1	3.0	2.5	2.5	2.4	NA	NA	ΝA	Ϋ́	Ϋ́	Ϋ́	×	N V	Z	Į V	Y Z	N A
Mn ⁺²	(l/gm)	0.5	0.4	16.0	15.0	14,0	14.0	33.0	20.0	19.0	18.0	18.0	18.0	17.0	15.0	51.0	49.0	48.0	84
Fe ⁺²	(l/gm)	74	. 6	2900	2500	2400	2200	7200	3100	3200	3000	3300	3200	2600	2300	11100	0800	9200	8700
Fe	(total) (mg/l)	62	57	3100	2500	2400	2200	7200	3200	3300	3000	3500	3200	2800	2400	11700	10500	9500	9100
Na ⁺¹	(l/gm)	6	· 10	64	28	99	59	110	9/	08	9/	72	72	72	62	160	170	160	160
K ^{±1}	(l/gm)	5	4	25	23	22	70	59	56	56	25	27	72	72	62	52	51	46	43
Mg ⁺²	(mg/l)	14	14	190	180	170	160	330	320	250	240	250	270	260	230	573	208	681	662
Ca ⁺²	(l/gm)	09	57	360	370	350	360	480	440	470	470	480	470	470	450	260	260	610	290
SO ₄ -2	(l/gm)	295	278	9310	8140	7710	7220	2300	10600	10500	9570	10400	10700	9230	7530	36100	29200	28800	29500
්ර	(mg/l)	10.5	10.6	20	47	45	43	34	36	51	55	18	70	28	56	<250	<100	9	<50
Alkalinity	CaCO ₃)	4	9	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Acidity ,	(equ. Ca((mg/l)	151	133	8200	0069	00/9	6200	19000	8000	8200	8100	10400	9500	8100	7200	31600	23900	25700	25100
盟	(vs. SHE) = (mV)	383	349	458	452	451	470	439	438	452	463	498	501	527	529	505	496	520	535
	Hd	5.9	2.8	4.1	4.2	4.3	4.1	3.8	4.0	3.8	3.8	3.9	4.0	3.6	3.6	3.5	3.9	3.7	3.1
	Date	95-01-11		95-02-01			95-02-01	95-03-13	95-03-13	95-03-13	95-03-13	95-05-10	95-05-10	95-05-10	95-05-10	95-07-31	95-07-31	95-07-31	95-07-31
		GVA-C	GVA-D	GVA-A	GVA-B	GVA-C	GVA-D	GVA-A	GVA-B	GVA-C	GCA-D	GVA-A	GVA-B	GVA-C	GVA-D	GVA-A	GVA-B	GVA-C	GVA-D

GVA-A; the water sample collected from the inlet of the apatite drain. GVA-B; the water sample collected from the outlet of the apatite drain. GVA-C; the water sample collected from the inlet of the pond. GVA-D; the water sample collected from the outlet of the pond.

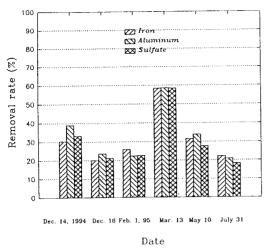


Fig. 5. Iron, aluminum and sulfate removal rates.

• The maximum amount of apatite dissolvable in the solution was 0.082 mol/l at pH 4.09, beyond which the pH was constant.

The results of MINTEQA2 runs appeared to support the conclusion of the experimental work. Apatite is effective in removing iron, sulfate, and aluminum from acid mine water and the pH was inversely proportional to flow rates.

CONCLUSION

The apatite drain system developed by author is a new method for AMD treatment. A naturally occurred material was sought which will remove metals from AMD, produce insoluble and powdery precipitates, have buffering capacity or at the least maintain the pH after reaction, and dissolves slowly so that it last several years or several decades. Apatite from Florida satisfied these conditions. Laboratory tests and the field experiment showed that this technique is quite effective for removal of high concentrations of iron and aluminum in the AMD with pH less than 4.0. In addition this system removes both ferric iron and ferrous iron whereas constructed wetlands systems remove only ferric iron when it is converted from ferrous iron in the wetlands.

However, future research on the reaction between AMD and apatite is recommended to define the mechanism of precipitation and constituents of the precipitates in order to optimize the design of the AMD treatment system.

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인회석 배수시스템을 이용한 산성수의 철 및 알루미늄 제거에 대한 현장경험

최정찬

요 약: 인회석 배수로가 1994년 9월 30일 미국 인디아나주 중서부에 위치한 Green Valley 폐탄광에 건설되었다. 현장설치의 일차적인 목적은 현장조건에서 산성수에 대한 인회석 배수로의 장기적인 처리능력을 평가하는 데 있다. 이 배수로는 실내실험결과에 따라 설계되었는 데 연장 9 m, 폭 3.3 m, 심도 0.75 m이며, 95 mm~30번 채 크기의 인회석광 (francolite)으로 채워져 있고 복구된 폐석더미로 부터 흘러나오는 산성수가 유입되게 되어 있다. 본 배수로는 폭우 및 실트의 퇴적으로 인한 손상을 막기 위해 석회석 자갈과 filter fabric으로 덮여져 있다. 본 배수로는 플로리다주의 인광석광산에서 채광된 50톤의 인회석으로 채워져 있다. 인회석은 산성수내의 철 최대 4,200 mg/l, 알루미늄 최대 830 mg/l, 황산염 최대 13,430 mg/l를 제거하였다. 산도는 거의 일정하였으며 3.1에서 4.3까지 변화를 보였다. 배수로 하류에서 측정한 유출량은 3~4.5 l/m였다. 철 및 알루미늄 인산염 침전물은 노란색 및 흰색 부유성 입자인데 침전못에 계속적으로 퇴적되었다.