Supplementary Materials: Strategies for Reduced Acid and Metalliferous Drainage by Pyrite Surface Passivation

Gujie Qian, Russell C. Schumann, Jun Li, Michael D. Short, Rong Fan, Yubiao Li, Nobuyuki Kawashima, Yan Zhou, Roger St. C. Smart and Andrea R. Gerson

S1. Watering/Flushing Procedures for KLCs Using Synthetic Mine Waste

The detailed procedures for watering/flushing KLC tests using synthetic waste were as follows:

- The control KLC was only watered and flushed with Milli-Q water throughout the entire process ('Milli-Q water'; Figure 2).
- The second KLC ("Milli-Q with lime-saturated water"; Figure 2) was initially flushed with Milli-Q water (Flush 0 at week 0), then watered and flushed with lime-saturated water for the next monthly cycle (i.e., watering between weeks 1–3 and Flush 1 at week 4). The lime-saturated water was made by dissolving $Ca(OH)_2$ (>95% purity, Geo Discoveries) in Milli-Q water until saturation, after which the supernatant was used (alkalinity: ≈1,900 mg $CaCO_3$ L^{-1}). This was then followed by watering and flushing with Milli-Q water until the leachate pH dropped to <5.5. At this time, lime-saturated water was once again used for watering and the next flush. On increase of the leachate pH to greater than 5.5, watering and flushing with Milli-Q water was resumed. This cycle is intended to test the requirement for lime-saturated water used alone, without calcite-saturated water, for maintenance of circum-neutral leachate pH.
- For the third KLC ("Calcite-saturated and lime-saturated water"; Figure 2), the operational procedures are the same as those for the second KLC (i.e., cycling lime treatment at pH < 5.5) except that calcite-saturated water (made by dissolving a natural calcite in air-exposed Milli-Q water until saturation; alkalinity: ≈30 mg $CaCO_3$ L^{-1}) is used in lieu of Milli-Q water for all watering and flushing after Flush 1 when the leachate pH is >5.5 (which has remained the case to date). This cycle is intended to test the capacity of calcite-saturated water alone to maintain pyrite passivation after the initial establishment of surface passivation.
- For the fourth KLC ("Calcite-saturated water"; Figure 2), calcite-saturated water is used for watering/flushing throughout the leach process, except that Milli-Q water was used for Flush 0. There was no initial Flush 1 with lime-saturated water as per the third KLC. This KLC test is intended to test the effectiveness of watering/flushing with calcite-saturated water alone for establishment and maintenance of pyrite surface passivation.

S2. The Elemental Composition of Pyrite

The elemental composition of pyrite used for the KLCs with synthetic waste is given in Table S1.

Table S1. The composition (in ppm) of pyrite.

Sample	Al	As	Ba	Ca	Cl	Cu	F	Fe	K	Mg
Pyrite	100	30	<5	100	<60	80	<100	428000	60	<30
Detection limit	10	5	5	30	60	5	100	5	50	30
Sample	**Mn**	**Ni**	**Na**	**P**	**Pb**	**Si**	**Sr**	**Ti**	**V**	**Zn**
pyrite	<5	4	<50	40	40	<30	5	<5	<5	100
Detection limit	5	3	50	30	5	30	3	5	5	5

S3. Leachate Chemistry

The ICP-OES results and E_h/volumes of leachates for KLC tests using natural wastes are given in Table S2.

Table S2. E_h and elemental concentrations for the calculation of saturation index and ferric concentration using PHREEQC and the Nernst equation.

KLC test	Flushing Time (week)	Volume collected (mL)	E_h (mV, SHE)	mg/L													
				Na	Mg	Al	S	K	Ca	Fe	Zn	Pb	Si	Ni	Co	Cu	Mn
Natural PAF Only	0	269	725	1.5	62.5	137.3	2080.0	1.6	36.3	2060.0	3.1	0.0	1.2	4.5	3.2	15.9	3.8
	4	432	695	0.2	168.1	330.8	3579.0	BD	49.9	3926.0	11.3	BD	6.4	10.2	5.7	41.3	10.6
	8	412	665	BD	113.4	249.6	3431.0	BD	26.6	3559.0	7.3	BD	6.4	8.1	3.6	25.3	6.0
	12	427	660	BD	146.2	337.9	5353.0	BD	22.6	5542.0	10.3	BD	6.3	11.4	4.4	27.5	8.3
	16	427	650	10.3	197.7	465.0	5710.7	0.6	29.5	7690.8	12.2	0.4	17.4	8.9	3.4	23.6	6.2
	20	420	667	1.1	118.9	290.0	4399.3	BD	23.5	5752.3	7.1	0.3	12.5	6.5	2.4	14.2	4.4
	24	338	670	BD	96.6	255.5	6590.1	BD	19.4	7164.2	5.7	0.2	6.3	7.5	2.8	13.4	5.2
	28	485	691	BD	47.2	129.5	2887.5	0.1	15.6	3145.8	2.9	0.2	3.2	3.9	1.4	6.3	2.5
	32	475	680	0.6	48.4	139.3	5790.3	BD	23.6	5835.7	3.0	0.1	3.3	4.0	1.6	6.7	2.8
	36	377	698	BD	18.4	52.7	4787.3	BD	7.9	5188.1	1.2	0.1	1.3	2.1	0.8	2.6	1.3
Natural PAF + lime	0	231	360	1.4	BD	BD	505.5	2.3	799.3	BD	BD	BD	0.3	BD	BD	BD	BD
	4	273	390	BD	31.5	BD	418.9	2.1	475.3	2.1	0.0	BD	1.4	0.1	0.1	0.3	0.3
	8	253	334	BD	54.2	11.0	460.8	BD	430.6	36.5	0.5	BD	5.2	0.8	0.8	1.8	1.2
	12	247	590	56.9	218.1	5.3	680.2	24.9	414.2	6.2	0.3	BD	8.3	0.9	0.8	3.3	1.3
	16	277	646	2.2	91.8	50.4	682.1	1.7	467.8	118.7	1.3	0.1	7.6	1.7	1.1	4.9	1.8
	20	268	675	1.7	109.5	91.8	1258.9	0.4	430.9	903.5	2.0	0.1	9.1	2.9	1.8	8.1	2.9
	24	292	683	0.8	83.2	73.4	1679.4	0.3	332.0	1739.7	1.6	0.0	1.7	3.2	2.0	7.1	3.3
	28	468	710	1.8	67.4	153.8	4007.0	BD	206.4	3646.0	2.8	0.2	3.0	5.4	2.9	14.5	4.9
	32	538	680	0.5	56.0	144.2	4971.7	BD	202.6	4770.2	2.5	0.2	2.8	4.5	2.7	13.0	4.7
	36	409	723	1.8	15.4	40.1	3258.9	BD	118.6	2940.5	0.8	0.0	0.8	1.7	0.9	3.7	1.5

Table S2. ***Cont.***

KLC test	Flushing Time (week)	Volume collected (mL)	E_h (mV, SHE)	mg/L														
				Na	Mg	Al	S	K	Ca	Fe	Zn	Pb	Si	Ni	Co	Cu	Mn
Natural PAF + lime + NAF1	0	246	319	1.7	BD	BD	532.7	2.8	881.2	BD	BD	BD	0.5	BD	BD	BD	BD
	4	390	375	1.8	102.4	BD	554.5	4.2	458.7	BD	BD	BD	1.9	BD	0.0	0.0	0.1
	8	374	392	1.4	109.9	BD	466.2	4.9	414.6	BD	BD	BD	2.0	BD	0.1	0.0	0.1
	12	403	530	1.4	151.0	BD	464.5	4.7	348.8	BD	BD	BD	4.5	BD	0.0	0.0	0.1
	16	414	560	3.7	229.7	BD	558.0	7.7	414.1	0.2	0.0	0.0	2.7	0.0	0.1	0.0	0.2
	20	497	630	3.0	164.3	BD	496.5	6.4	384.6	0.4	0.1	0.1	2.9	0.0	0.1	0.0	0.2
	24	472	530	1.8	122.0	BD	373.5	5.9	298.3	0.6	BD	BD	BD	0.0	0.1	0.0	0.3
	28	635	540	7.2	212.5	0.7	465.3	12.1	298.8	3.1	0.2	BD	0.2	0.2	0.4	0.2	1.0
	32	602	525	5.8	161.7	4.3	413.5	13.4	292.8	5.8	0.4	0.0	0.4	0.1	0.4	0.7	0.9
	36	533	540	0.6	69.9	5.5	280.7	6.9	265.9	6.7	0.2	BD	0.2	BD	0.2	0.8	0.4
Natural PAF + lime + NAF2	0	231	376	24.8	BD	BD	561.2	6.7	1128.0	BD	BD	0.0	0.3	BD	BD	BD	BD
	4	323	380	91.1	154.8	BD	784.8	18.5	504.9	BD	0.0	BD	3.1	BD	0.1	0.1	1.5
	8	303	350	83.6	226.2	BD	733.3	26.7	436.4	1.2	0.1	BD	5.7	0.1	0.3	0.3	4.3
	12	340	540	11.3	64.5	21.7	471.2	2.5	385.6	60.9	0.7	BD	2.0	0.5	0.7	1.2	5.2
	16	349	550	58.1	242.7	18.5	814.6	34.8	527.6	1.0	0.9	0.1	14.1	1.3	1.3	2.3	8.8
	20	419	570	33.9	197.0	24.2	683.2	24.9	555.8	2.4	1.6	0.1	17.1	1.1	1.3	3.2	8.0
	24	389	590	60.3	138.0	19.1	613.2	22.5	412.8	4.1	0.9	BD	1.0	1.1	1.3	2.9	8.6
	28	607	608	23.8	104.3	51.9	568.0	14.9	409.0	59.3	1.3	0.0	1.4	0.5	1.2	6.7	8.7
	32	625	613	10.2	74.3	45.2	543.0	9.2	403.3	84.0	0.6	BD	0.6	0.6	0.4	6.1	5.3
	36	528	624	5.1	50.0	35.0	661.2	4.7	404.8	279.0	0.4	BD	0.4	0.8	0.3	4.4	6.1

BD: below detection limit.