Permanent under Water Storing of Weathered Mine Waste after Removal of Fine Fraction and Addition of Ash

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Abstract Weathered sulphidic mine waste is a major environmental problem. An experiment was performed in order to study water covers for oxidized mine waste. In two experiments oxidized mine waste were covered with water, in one experiment the fine fraction was removed and in one experiment alkaline ash was also added prior to water covering. It was found that removal of the fine fraction decreased pH and increased trace element concentrations. Water covering of the mine waste with and without ash decreased trace element concentrations indicating that co disposing oxidized sulphidic mine waste and ash under water might be a promising remediation method.

Keywords acid rock drainage, oxidized mine waste, alkaline, remediation, copper

Introduction

Water cover is a very common method used for modern waste (mainly tailings) in order to reduce further weathering and oxidation of sulfides to a minimum (Vigneault et al. 2001). Due to the significantly lowered diffusion of oxygen in water (around $2*10^{-9}$ m²/s) compared with in air $(1.78*10^{-5} \text{ m}^2/\text{s})$ the oxidation is lowered with several orders of magnitude. In the case with pre-oxidized mine waste there is, however, a large uncertainty concerning reductive dissolution of pre-formed secondary iron(hydr)oxides and their associated trace metals. It has been argued that flooding of pre-oxidized mine waste will result in high trace metal concentrations and acid pH due to the dissolution of secondary precipitates (Ribet et al. 1995; Simms et al. 2000; Simms et al. 2001). To avoid this potential disadvantage for pre-oxidized mine tailings protective layers (Simms et al. 2001) or layers with alkaline materials (Catalan and Kumari 2005) have been suggested (Simms et al. 2001).

Performed laboratory experiments on the flooding of pre-oxidized tailings (Simms et al. 2000) showed that even if the pore water quality improved considerably the overlying surface water still had low pH and high metal concentrations after 600 days. Both low pH and high metal concentrations from historical oxidized mine waste can, however, be prevented by the addition of an alkaline amendment (preferably an industrial by-product) (Sartz et al. 2009). Performed experiments by Catalan and Kumari (2005) using lime mud residue covers showed that the quality of the surface water increased.

The present flooding experiment was to simulating flooding of an oxidized waste rock (both with and without the fine fraction) amended with an ash with a permanent water cover (in a natural lake or in a flooded open pit). During the experiment possible reductive dissolution of ferric(hydr)oxides were studied as well as ongoing oxidation of sulfides.

Materials and methods

The flooding experiments were designed to simulate the chemical changes taking place when a weathered and acidic historical mine waste is mixed with a strongly alkaline material and flooded with water.

The historical mine waste, from Ljusnarsbergsfältet in Kopparberg, Sweden (Sartz et al. 2009), is heavily weathered and 200-300 years old. It contains elevated concentrations of primarily copper, lead and zinc. Before it was used in the experiments aggregates were crushed and the fraction smaller than 13 mm (referred to as the fine fraction) was removed in some experiments. Ash used in the experiments was from a boiler fuelled by municipal solid waste in Ludvika, Sweden. Due to its content of trace elements it is classified as hazardous waste. Eight experiments were set up in 1 m^3 plastic containers according to table 1 and fig. 1. Water for covering was retrieved from a nearby stream.

Exp.	Fractions	Cover	Ash
1	Coarse	Water	10 %
2	Coarse	-	10 %
3	Coarse/fine	Water	-
4	Coarse/fine	-	-
5	Coarse	Water	-
6	Coarse	Water	-
7	Coarse	-	-
8	Coarse	-	-

Table 1 Description of the eight experiments and their content



Fig.1 Experimental set up during the winter of 2013

Sampling was performed regularly from July 1 until mid November 2013. Sampling was performed both from the water cover and from the bottom of the containers. From systems not covered with water, sampling was performed less regularly (i.e. after precipitation events). Analysis was performed with respect to pH, electrical conductivity, acidity/alkalinity and redox potential. Metal analysis was made with an Agilent 7500 cx ICP-MS that was equipped with a MicroMist nebulizer and a Scott type double pass spray chamber. The spray chamber temperature was kept at 2 °C to minimize the formation of oxides. An internal standard of ¹⁰³Rh was added manually to all solutions to give a concentration of 10 μ g/L.

Results and discussion

Only results from the bottom of the containers are presented here. In most systems the chemistry in the bottom and in the surface waters was fairly similar. Systems 4, 7 and 8 have the lowest pH and the highest acidity of all systems (table 2). These are the non sieved and exposed to the air systems. Acidity values indicate a higher rate of weathering in these

systems compared to the water covered systems (compare systems 3, 5 and 6). Water covering only increases pH with around a half unit. Addition of ash and water covering (system 1) increases the pH significantly (around 3 orders of magnitude). It should, however, be noted that in system 1 there was a great difference in pH between bottom water (pH 6.38) and surface water (pH 3.65). Similar results have been noted in other experiments (Simms et al., 2000). Addition of ash in an open system resulted in a much lower pH (system 2). It should be noted that the concentrations in open and water covered systems are not directly comparable due to the large difference in water volume.

Exp.	pН	El. Cond (µS/cm)	Alk (meq/L)	Redox (mV)	Na (mg/L)	Ca (mg/L)	Fe (mg/L)
1	6.38	12 100	2.73	-110	708	934	2.17
2	4.21	23 700	-1.27	210	3 080	1 640	28.6
3	2.88	2 060	-3.22	93	7.6	520	94.1
4	2.46	3 320	-45	460	20.3	446	406
5	3.07	3 020	-3.07	160	15.3	499	909
6	3.18	2 920	-2.59	110	17.5	516	925
7	2.47	4 790	-91	440	4.21	415	1 520
8	2.39	5 940	-160	450	3.97	427	2 0 2 0

 Table 2 General chemistry (average values) for all systems tapped at the bottom. Grey highlight indicate water covered systems

It is also clear that adding ash to the systems increased the electrical conductivity substantially indicating a release of soluble salts from the ash (see for instance Na in table 1).

 Table 3 Trace element concentrations (average values) for all systems tapped at the bottom. Grey highlight indicate water covered systems

Exp.	Cu	Cd	Pb	Zn	Cr	Мо
	(µg/L)	(µg/L)	(µg/L)	(µg/L)	(µg/L)	(µg/L)
1	269	889	84.8	247 000	16.7	4.62
2	105 000	3 450	4 860	518 000	41.2	Bdl
3	7 490	19.9	1 130	7 340	11.6	Bdl
4	85 800	233	1 150	93 400	54.7	Bdl
5	52 400	730	1 100	98 200	10.1	1.24
6	32 300	859	1 020	88 700	10.4	1.68
7	1 090 000	4 470	259	612 000	31.6	Bdl
8	1 440 000	5 710	262	887 000	36.2	Bdl

System 4 can be considered the unamended system (no fine fraction removed, no water cover and no addition of alkaline ash) and should thus represent the worst case scenario.

When covering the mine waste with water (system 3) pH increases somewhat (table 2) and the trace element concentrations decrease (table 3).

However, when removing the fine fraction (<13 mm) (systems 7 and 8) acidity and iron increases substantially compared to system 4 (table 2) indicating an increase in oxidation of acid generating minerals (FeS and FeS₂). In the open systems containing only the coarse fraction, the concentrations of copper, cadmium and zinc are very high compared to the other systems indicating an increased weathering. However, lead concentrations decrease in these systems. This is probably due to precipitation of anglesite (PbSO₄) as sulphate concentrations probably have increased.

Systems 5 and 6 (water covered coarse fraction) have higher pH and much lower acidity compared to systems 7 and 8. Trace element concentrations are also significantly lowered, except for lead and molybdenum (table 3). Cadmium decreased 84 %, zinc 88 % and copper 97 %, while lead increased with around 300 % (no anglesite precipitation). This indicates that water covering lowers the weathering rate and the reductive dissolution is not as pronounced

as would be expected even though the iron concentration is quite high in the water (above 900 mg/L). Copper concentrations in systems 5 and 6, decrease during the experiment from around 140-240 mg/L after water covering to around 2-3 mg/L after a couple of months. Iron concentrations, on the other hand, increase from around 400-500 mg/L in the beginning to almost 1 000 mg/L after a couple of months.

Water covering of coarse and fine fraction (system 3 compared with system 4) also supports the conclusions that water covering reduces weathering and lowers trace element concentrations. Trace element concentrations are lowered by around 90 % except for lead, which probably is governed by anglesite dissolution (table 3).

When adding ash to the coarse fraction (system 2 compared to systems 7 and 8) pH increases from around 2.4 to around 4.2. Copper concentrations are lowered by around 90 % while cadmium and zinc concentrations are only slightly lowered. Lead concentration increase from around 260 μ g/L to around 4 900 μ g/L when ash is added to the system. It is likely that the low pH releases cadmium, lead and zinc from the ash. No significant release of other typical ash elements (chromium, molybdenum) was noted.

Adding ash to the coarse fraction during water covered conditions (system 1 compared to systems 5 and 6) increased the pH from 3 to above pH 6. Copper and lead concentrations decreased 99 % and 92 %, respectively, while cadmium and zinc concentrations increased (11 % and 164 %) despite higher pH. Increased cadmium and zinc concentrations are most likely due to leaching from the waste ash.

Conclusions

This study has demonstrated that removing the fine fraction from a weathered mine waste prior to reclamation has no positive benefits. This seems to be true for both remediation strategies of traditional covering and water covering. In both cases pH dropped and trace element concentrations increased when the fine fraction was removed.

Water covering of weathered mine waste decrease trace element concentrations by around 90 %. Addition of ash decreased the trace element concentrations even more, as long as the ash itself did not release trace elements. Preliminary results indicate that co-disposing oxidized sulphidic mine waste and ash under water might be a promising remediation method.

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