

## ARD treatment in sequential filter sections – efficiency of different alkaline waste materials

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**Abstract** Six alkaline waste materials are tested as potential filter materials for ARD treatment. The study is an ongoing project since 2.5 years on a mine waste remediation test field in Kopparberg, Sweden. The filters (0.5 m<sup>3</sup>) are operating under field conditions and general parameters (pH, electric conductivity, redox potential (Eh), alkalinity, acidity and sulphate) are measured immediately after sampling in a mobile laboratory. The reactive (alkaline) materials are followed by other filter materials in sequence with non-reactive support materials, to ensure iron/aluminum precipitation and trace metal sorption.

**Key Words** Oxidized mine waste, paper mill by-products, fly ash, lime kiln dust, steel slag

### Introduction

Abiotic remediation of ARD with barriers and filters (passive systems) are often used in order to minimize the impact on receiving streams, rivers and the wider environment (Johnson and Hallberg 2005). Generally, treatment of ARD with high concentrations of iron causes problems with iron precipitation subsequently inhibiting neutralizing surfaces (Santomartino and Webb 2007). By keeping reducing conditions while increasing the pH, i.e. RAPS (reducing alkalinity producing systems; Younger et al. 2003) or ALD:s (anoxic limestone drains), these problems can be overcome (Cravotta 2003). However, problems with keeping reducing conditions as well as latent acidity in the recipient, once iron and aluminium precipitates are common. Aeration of ARD before raising pH, hence excluding iron from the remaining treatment is an alternative method.

This paper reports on the effectiveness (in terms of acid neutralization and metal reduction) of six alkaline by-products as reactive filter materials. The filters are operating under field conditions and are kept open to the atmosphere, i.e. no measures are taken against surface passivation of iron precipitates. The alkaline materials are placed first out of three in sequential filter sections. Only the first section in each filter is reactive (the one containing an alkaline material). The following sections contain supportive, non-reactive materials (e.g. crushed brick and peat).

### Methods

The alkaline materials are: lime kiln dust (LKD), LD-slag (LD), green liquor dreg (GLD), fresh fly ash (FFA), carbonated fly ash (CFA) and a mixture of lime mud and fresh fly ash (LM/FFA). Since the start of the experiments in June 2008, some 2000 liters of acidic (pH 3, acidity 1.8 meq/L) and metal-rich leachates have passed each filter.

The filters have received ARD from two reactors during two seasons (May to October). Each reactor is coupled to three filters. pH, acidity and metal load varied between the first and second season, however not considerably between the two reactors. The filters consist of three coupled tubes (each 0.5 m<sup>3</sup>; fig. 1) receiving ARD which is successively passing the tubes through gravity (the filters are built on a slope, approximately 10° incline). An obstacle is placed in the middle of each filter section to achieve good contact between the alkaline materials and the ARD.

### Results

General characteristics of the alkaline materials are presented in tab. 1. Acidity loadings from the reactors were 3 000 meq (FFA, CFA, LM/FA) and 2 500 meq (LKD, GLD, LD) during season 1 and 550 meq for season 2 (all filters). Added acidity is well below the total alkalinity in the filters (tab. 1). During season 1, only the LKD and LD filters were able to produce net alkalinity (tab. 2), while during season 2, in addition to LKD and LD, also CFA and GLD filters produced net alkalinity (tab. 2). Net alkalinity production in the FFA filter was low; average values of 0.3 and 0.1 meq/l for season 1 and 2 respectively. The LM/FFA filter was not able to produce alkalinity in any of the seasons. In

**Table 1** Total amount (kg) of alkaline material in each barrel, total alkalinity (based on analysis of available lime index and carbonates) and estimated surface area (estimated from the particle size distribution)

	Amount (kg)	Total alkalinity (meq/kg)	Surface area (m <sup>2</sup> /kg)
FFA	320	3 200	25
LM/FFA	570	6 600	30
CFA	310	2 500	40
LKD	610	9 000	50
GLD	330	7 400	50
LD	960	1 600	5



**Figure 1** View of two of the filters and the ARD reactors. Each filter consists of three coupled plastic barrels (each 0.5 m<sup>3</sup>)

connection with the large pH increase between season 1 and 2 for LKD and LD filters (tab. 2), a decrease in redox potential was noticed.

Performance in terms of acidity neutralization and trace element reduction (tab. 3, fig. 2) increase in season 2 compared to season 1, even for the low performance filters LM/FFA and CFA. One exception is however the FFA filter, where both average pH, acidity neutralization and trace element reduction decrease in season 2 compared to season 1 (tab. 3). This could be due to increased carbonation of the fly ash, suggesting the performance will decrease over time and eventually be comparable with the CFA filter.

**Table 2** pH and alkalinity (average values) during season 1 and 2. Filters with fresh fly ash (FFA), lime mud and fresh fly ash (LM/FFA) and carbonated fly ash (CFA) receive ARD from reactor 1. Filters with lime kiln dust (LKD), green liquor dregs (GLD) and LD-slag (LD) receive ARD from reactor 2

	Season 1		Season 2	
	pH	Alkalinity (meq/l)	pH	Alkalinity (meq/l)
Reactor 1	2.9	-2.4	3.3	-0.8
FFA	5.7	0.3	5.0	0.1
LM/FFA	3.6	-2.2	3.8	-0.5
CFA	3.4	-2.0	5.0	1.4
Reactor 2	3.0	-1.7	3.3	-0.7
LKD	5.8	10.7	10.7	17.1
GLD	3.7	-1.5	5.5	0.3
LD	4.9	3.3	9.7	13.5

**Table 3** Reduced amount of Cd and Pb (in %) through the filters compared to the loading (g) to each filter from the reactors

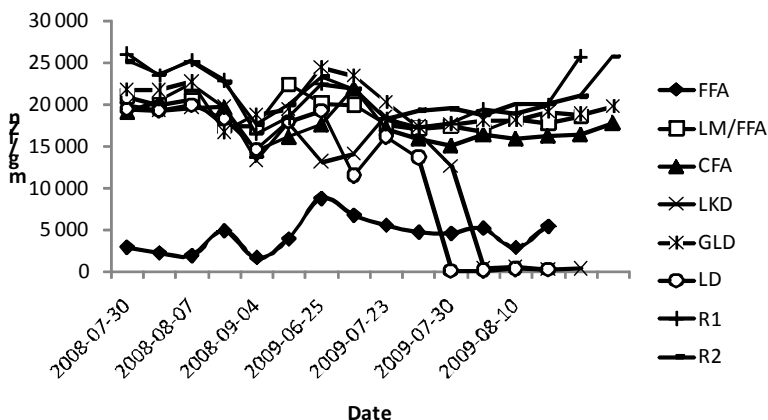
	Season 1		Season 2	
	Cd	Pb	Cd	Pb
FFA	97	89	75	87
LM/FFA	7	46	7	17
CFA	17	32	12	45
LKD	93	54	81	80
GLD	8	25	8	20
LD	19	42	83	81

Concentrations of Fe and Al from the reactors are around 90 mg/L and 25 mg/L, respectively (season 2). Approximately 81% of the Fe and 80% of the Al is precipitated in the FFA filter (season 2). Corresponding numbers for the other filters are: LM/FFA Fe: 14%, Al: 13%; CFA Fe: 30%, Al: 19%; LKD: Fe: 71%, Al: 64%; GLD Fe: 18%, Al: 12% and LD Fe: 79%, Al: 73%. Thus, two groups are recognized: (1) FFA, LKD and LD, where approximately 70–80% of Fe and Al precipitate, creating efficient sorption sites for trace elements and (2) CFA, GLD and LM/FFA, where only 10–30% of Fe and Al precipitate.

Retention of trace metals is shown in table 3 (Cd and Pb) and in figure 2 (Zn). Trace metal immobilization is closely related to pH and precipitation of Fe and Al. Filters with good trace metal retention, approximately 80% of the loading from the reactors, are FFA, LKD and LD (tab. 3, fig. 2). In the first season, the FFA-filter was working well with pH around 7–8. However, in the beginning of the second season the ash underwent carbonation and pH dropped to around 4.5–5 (tab. 2), still though, trace metal retention is rather good in the FFA-filter (tab. 3, fig. 2).

The mixture of lime mud and fresh fly ash as well as the carbonated fly ash are not working as expected (pH around 3 and hence low trace metal immobilization). Probably due to surface passivation (iron (III) precipitates). Overall performance of the GLD-filter is improved from season 1 to season 2, however, pH is still rather low and in combination with low degree of Fe- and Al precipitating, trace metal reduction becomes poor (tab. 3, fig. 2).

In previous laboratory studies (Sjöberg et al 2010) the release of vanadium from LD-slag was found to be pH-dependent, with leaching maximum at pH 10. Therefore, some substantial release of vanadium was expected in the filter system with LD-slag. Despite an average pH of 9.7 during season 2 (tab. 2), vanadium concentrations were however repeatedly low.



**Figure 2** Zinc concentrations (in mg/L) during season 1 and 2 (2008 and 2009)

### Conclusions

Filters with low Eh (LKD and LD) also have the highest pH (10–11) and alkalinity (10–15 meq/l). The best trace element reduction is however seen in the FFA filter during the first season. Despite a rather low average pH, generally too low for quantitative Cd removal, the massive amount of Fe- and Al-precipitates enables a high degree of sorption and/or co-precipitation of the trace elements. Unfortunately, the overall performance of the FFA filter is decreasing, probably due to carbonation of the fly ash. Reasons for low filter performance are not due to low capacity, but probably due to passivation and preferential flows through the alkaline materials. The combination of low acidity consumption (low pH) and low amount of sorbent phases (Fe and Al) are most likely the explanation for the low trace element reduction in filters LM/FFA, CFA and GLD. In summary, filters with hydroxide materials (LKD, LD and, before carbonation, FFA) are superior to filters with carbonate materials.

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