WASCON 2012 Conference proceedings.
M. Arm, C. Vandecasteele, J. Heynen, P. Suer and B. Lind (Editors)

Use of rest products as additive in tailings paste for the mitigation of ARD: Effect of green liquid dregs and fly ash addition on geotechnical stabilization of tailings

Yu JIA[1,*], David STENMAN[1], Maria MAKITALO[1], Christian MAURICE[1], Björn ÖHLANDER[1]

[1] Division of Geosciences and Environmental Engineering, Luleå University of Technology, SE-971 87 Luleå, Sweden
**Postdoc fellow, Division of Geosciences and Environmental Engineering, Luleå University of Technology, SE-97187 Luleå, Sweden, Email: yu.jia@ltu.se*

Abstract

This study focuses on the effect of green liquor dregs (GLD) and fly ash (FA) on geotechnical stabilization of tailings, using shear strength test.

The objectives were to evaluate thickening of tailings using GLD to paste to improve its water retention and to reduce waste percolation, to identify optimal proportion of additives regarding geotechnical stabilization of tailings, and to study the role of FA in the decrease of porosity and thus to reduce oxygen diffusion via hardening process.

The results showed that GLD is a potential material for tailings stabilization by decreasing water percolation and improving water retention property. However, the paste of GLD amended tailings withstands low shear strength. The addition of FA to the paste greatly improves shear strength which is up to 2-3 times higher. Moreover, the fly ash hardening process effectively reinforces the strength of GLD amended tailings paste. Longer curing period (3 months) for specimens leads to up to 2-3 folds higher strength compared to that of 1 month. Hydraulic conductivity is reduced as a result of GLD and FA addition, since the porosity of the tailings decreased. If reduced porosity, improved water retention capacity and no cracks are ensured in the tailings, oxygen diffusion can be limited.

Keywords: Acid rock drainage; Fly ash; Green liquor dregs; Shear strength test; Tailings stabilization.

1 Introduction

1.1 Introduction to mine waste

Mining of sulphidic metal ores results in waste rock and tailings, which contain sulphide–bearing minerals. In the presence of water and oxygen, sulphide minerals are unstable and oxidize. The oxidation of iron-sulphides such as pyrite and pyrrhotite lead to generation of acid rock drainage (ARD), which can last for hundreds or even thousands of years. ARD may cause soil erosion, deteriorate water (surface water and ground water) quality, posing an adverse impact to ecosystems as well (Lottermoser, 2010). Surface dry cover application, liming and water management (e.g. pit lake) are common remediation options for tailings sites.

Sweden is one of the most active European countries in mining activities, and approximately 59 million tons of reactive sulphidic mine tailings are generated annually (Statistics-Sweden, 2008). Attention has been therefore focused on tailings management with respect to monitoring of contaminated sites, development of new reliable strategies for tailings remediation and prediction of environmental fate of the contaminants especially heavy metals (see e.g. Carlsson et al., 2003; Alakangas et al., 2010).

One method to remediate ARD used in Sweden is to add lime in the mine waste, however ARD neutralization in lime stations involves high costs, and generates large amount of waste (sludge mainly consisting of gypsum and Fe hydroxides) (Blowes, 1997). Various less expensive alkaline material such as red mud, cement kiln dust and sodium bicarbonate are added to mine wastes to neutralize it and enhance hydroxide precipitation (see e.g. Hallberg et al., 2005; Bertocchi et al., 2006; Kalin et al., 2006).

Thickening of tailings to paste improves the hydro-geotechnical properties of tailings i.e. reducing water percolation, improving water retention and decreasing free oxygen transport. Thus it hinders sulphide oxidation in the heaps to prevent ARD generation in situ, also on long time scales.

Therefore, a number of alkaline materials such as fly ash (FA) and paper wastes as alkaline additives to neutralize and stabilize sulphide-rich mining wastes have been assessed (e.g., Bellaloui et al., 1999; Yeheyis et al., 2010; Pérez-López et al., 2011). Using alternative materials such as industrial waste as paste additives to mitigate the negative effects of ARD would solve two waste problems simultaneously.

1.2 Introduction to fly ash

FA has a potential to serve as a barrier material for mine wastes, attributing to its pozzolanic and highly alkaline properties resulting from its high calcium content (Nehdi and Tariq, 2007). With respect to solid consolidation and soil remediation, the commonly used FA is coal fly ash generated from power plant. However FA produced from biofuel incineration is common in Sweden. Although energy produced from biomaterials today is sufficiently effective and the process leaving a little residue occurs, large amount of FA is generated (Lindgren, 2005). The substances contained in the ash are calcium and magnesium, with a low nitrogen content. Wood ash generated from municipal heating plants and forest industry has a hydraulic conductivity of between 10^{-7} and 10^{-11} s/m (Lindgren, 2005).

1.3 Introduction to green liquor dregs

Each year large amount of Green liquor dregs (GLD) are produced, and landfilled in Sweden (Lindgren, 2005). GLD are a residual product generated by the pulp and paper industry from sulphate pulping process. Typically GLD are composed of sodium carbonate, sodium hydroxide calcium carbonate, unburned carbon, sulphides and trace amount of heavy metals. They are normally with high pH and possess a fine clayey texture, and the hydraulic conductivity for GLD is in the range of 10^{-8} to 10^{-9} m/s (Lindgren, 1995; Maurice et al., 2009). Limited work has been performed for ARD mitigation using of paper mill wastes (see e.g. Pérez-López et al., 2011).

1.4 Previous findings

Results from earlier studies indicate that addition of 10% GLD (dry weight basis) reduced the hydraulic conductivity of the tailings by a factor of two. Addition of a 30% mixture (dry weight basis) of GLD and FA to tailing collected at an abandoned copper mine, reduced the hydraulic conductivity by more than one order of magnitude. Thanks to its alkaline properties, the rest products had a direct effect on the metal leaching from tailings. The alkaline capacity of the pulping wastes raised pH in the stabilised tailings reducing leaching of metals e.g. Cu, Co, Cd, and Ni. Addition of 10% pulping waste to tailings on dry weight basis was efficient to reduce copper leaching by a factor 4 to 10 (Maurice et al., 2010).

1.5 The objectives

The objectives of the study were

i) to evaluate thickening of tailings using GLD to paste to improve its water retention and to reduce waste percolation;
ii) to identify optimal proportion of additives regarding geotechnical stabilization of tailings;

iii) to study the role of FA in the decrease of void space and thus to reduce oxygen diffusion via hardening process.

2 Materials and methods

This work was carried out in Complab at Luleå University of Technology.
To enhance cohesive properties of tailings and to increase its compressive strength, addition of GLD and FA was performed, and shear strength test (uni-axis pressure test) was carried out for samples after a curing period of 1 and 3 months. Tailings used in this study were collected from Boliden and was a blend of ore from Maurliden and Kristineberg. Green liquor dregs and FA were obtained from Billerud in Kalix. Upon received, GLD from the containers (see Fig 1A) were mixed and filled back to the container, and then were hand-shaken to achieve a homogeneous distribution.

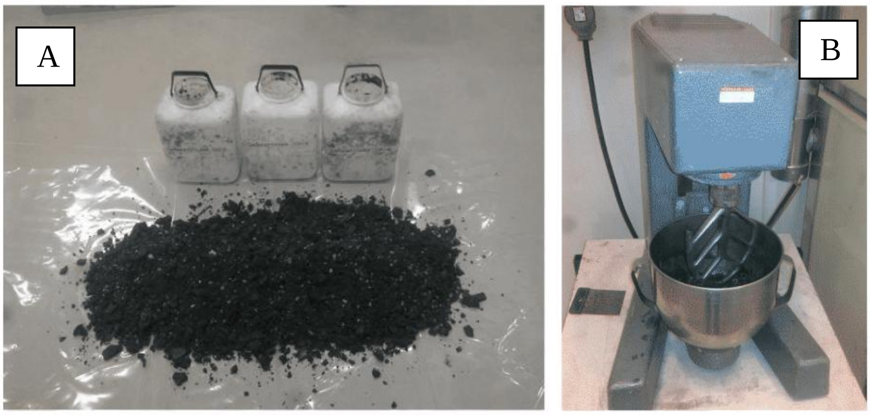

Figure 1. Sample of green liquor dregs and mixing of tailings with additives.
A: green liquor dregs. B: mixing of samples.

Seven samples were prepared for the strength test with varying weight percentage and curing time. Mixing of samples is shown in Fig. 1B. The experiments were conducted in duplicate to ensure the reliability of the test results resulting in a total number of 14 samples (Table 1). For FA two samples were stored for three month to study the effect of curing time. The packing of each sample was performed systematically in stages by adding approx. 1 cm material in a cylindrical test tube each time. The same procedures were repeated until a length of 11 cm samples was packed. Afterwards the samples were stored at +5 °C (equivalent to soil temperature in the field) under a pressure of 20 kPa, which simulates a load of about 1 m soil column.

Table 1. Experiment setup for strength test

Sample nr.	**Tailings (%)***	**GLD (%)**	**FA (%)**	**Curing time (month)**	**Replicate**
1	-	100	-	1	1x2
2	70	30	-	1	1x2
3	40	60	-	1	1x2
4	70	20	10	1	1x2
5	70	20	10	3	1x2
6	30	60	10	1	1x2
7	30	60	10	3	1x2
Total	-	-	-	-	7x2=14

* Percentage in dry weight.

To measure the compressive strength of the samples, cylindrical specimens (with a diameter of 5 mm and an adjusted height of 10 mm) were prepared. During the strength test, rammers applied with Vaseline on the perpendicular surface where force acted on. The Vaseline was used to reduce frictional forces when strength was tested for the samples. The pressure was set at a deformation rate of 1.5% / min. The force to measure was registered every 10 seconds until "breaking" occurred. The term "breaking" refers to a sample failing to take increased loads.

3 Results and discussion

Results from the shear strength tests (Table 2) showed that very low shear strength (12-19 kPa) was recorded when only GLD or GLD (30% and 60%) mixed with tailings (40% and 60%) was tested, represented by samples nr.1-3. In contrast if 10% of either tailings or GLD is replaced by FA, the samples (represented by samples nr.4 and 6) withstood ca. 4 times higher shear strength (ca. 58-92 kPa) which is in the range of medium to high level based on designation by Larsson (2008). Moreover, it showed that fly ash-hardening processes affect strength efficiently. Samples stored for 3 months withstood strength of ca. 116-250 kPa (high to very high) in comparison to that of ca. 58-92 kPa (medium to high) for samples cured for 1 month, which is up to 2-3 times higher. After storage, large cracks in one of the duplicates of sample nr.3 were observed, and thus it withstood lower shear strength (9.5 kPa) compared to its counterpart (28.4 kPa). Although with a low FA level (10%) in the additive, very high shear strength has been recorded for sample nr.5.

It is expected that if GLD amended tailings is placed on the top of a heap, 15-16 kPa (very low shear strength) might be sufficient. However, if the GLD amended tailings is placed on the slope of a hillside, FA might be required to withstand medium to high shear strength at 60-90 kPa regarding the physical stabilization.

Table 2. Laboratory experimental results from shear strength test

Sample nr.	**τ (kPa) Avg±SE (n=2)**	**Classification (designation values*, kPa)**	**Water ratio (%)**	**ρ (t/m^3)**
1	11.8±2.2	Very low (10-20)	117	0.63
2	15.7±4.3	Very low (10-20)	49	1.26
3	19.0±13.4	Very low (10-20)	78	0.93
4	91.5±7.0	High (75-150)	35	1.57
5	250±23	Very high (150-300)	35	1.57
6	58.4±10.1	Medium (40-75)	62	1.07
7	116±35	High (75-150)	62	1.07

*Larsson, 2008. τ=shear strength; ρ=dry density; Avg=average, SE=standard error.

The results from previous studies regarding hydraulic conductivity and physical stabilization showed the potential application of GLD in tailings stabilization. The addition of GLD may decrease the leaching of heavy metals and a mixture of GLD and fly ash has the potential to decrease the hydraulic conductivity of the tailings (Villain, 2008; Maurice et al., 2010).

This study showed that the paste of GLD and tailings withstands low shear strength, however the addition of biofuel incineration generated FA to the paste greatly improves shear strength. This is in agreement with the findings of Prashanth et al. (2001), who showed that pozzolanic fly ashes develop good strength properties with time because the ashes contained sufficient lime, while Non-pozzolanic fly ashes do not develop strength even with addition of lime.

Moreover the results of this study showed that the fly ash-hardening process effectively reinforces the strength of GLD amended tailings. Based on Termkhajornkit et al. (2009), FA has the ability to seal the cracks caused by both autogenous and drying shrinkage in concrete, since hydrated products from FA may modify microstructure. It showed in that study that most of the cracks occur within 28 days, while FA continued to hydrate after 28 days and therefore seal these cracks.

Nevertheless higher levels of FA tend to increase uncertainty for the strength, as the risk to form cracks in the mixture increases after hardening process. The challenge of the cover for mine waste in cold climate is that subsidence in the ashes during snow-melt period can cause cracking in winter (frost heave). However Prashanth et al. (2001) evaluated the role of FA as a hydraulic barrier in landfills and showed that fly ashes do not crack since they possess low shrinkage.

The main advantages of GLD are that they do not harden, are plastic and are not easily affected by frost heave (Lindgren, 2005). Although both GLD (Stenman, 2011) and FA are difficult to compact (Prashanth et al., 2001), a mixture of GLD and FA may both decrease pore space and increase plastic properties in the tailings since FA has an ability to fill the micro-pores (Termkhajornkit et al., 2009) and GLD has plastic properties (Lindgren, 2005). To achieve a better understanding of the stabilisation mechanisms, a more detailed study is needed to investigate the optimal proposition of tailings and additives, as well as other factors such as curing time.

Addition of GLD and FA at a level of 30% to tailings reduces hydraulic conductivity of the tailings by 1-3 orders of magnitude (Maurice et al., 2009). Addition of FA may decrease porosity of the tailings, which was shown by grain size distribution of the FA, GLD and tailings (Makitalo, 2010). GLD processes a higher water retention capacity than tailings due to that GLD has a finer particle size distribution (d10: 1.4 µm, d90: 20.2 µm) than that of tailings (d10: 3.6 µm, d90: 99.1 µm) (Maurice et al., 2010). The goal to increase water retention capacity for tailings therefore may be met as a result of tailings amendment with GLD. If the following prerequisites such as decrease of porosity, increase water retention capacity and no cracks are met, the goal to limit free oxygen diffusion in tailings can be achieved.

Geotechnical stabilization of amended tailings paste was focused in the above discussion. Note that chemical stability is one of the important factors regarding the stabilization of the tailings.

Results from previous leaching test revealed that GLD has a direct effect on the metal leaching from tailings, attributed to its alkaline properties and its negative zeta potential. The alkaline capacity of GLD raised pH in the stabilized tailings and reduced leaching of heavy metals such as Cu, Co, Cd and Ni. An addition of 10% GLD to tailings on a dry weight basis was sufficient to reduce copper leaching by a factor of 4 to 10 (Maurice et al., 2010).

To qualitatively and quantitatively evaluate water drainage from dry cover with alkaline materials to the tailings, chemical properties e.g. acid neutralization capacity together with chemical characterization of different additives is under the investigation in our laboratory. According to the findings from Makitalo et al. (2012) which is presented in the same conference, fly ash has a higher leaching capacity than GLD in general.

Based on the results from physical- and chemical stabilization, a comprehensive evaluation will be performed to find out optimal proportion of tailings and additives, and selected ratio of paste will be tested in the field trials in the near future.

4 Conclusions

- Green liquor dregs (GLD) is a potential material for tailings stabilization by decreasing waste percolation and improving water retention property, however the paste of tailings and GLD withstand low shear strength (12-19 kPa).
- The addition of fly ash (FA) in GLD amended tailings paste lead to higher shear strength (58-92 kPa) which is up to 2-3 folds in comparison to the case without FA.
- Fly ash hardening process affects positively to reinforce the strength. The specimens stored for 3 months withstood high to very high strength (116-250 kPa) in comparison to that of medium to high (58-92 kPa) for samples cured for 1 month, which is up to 2-3 folds higher.

- Water retention capacity in GLD amended tailings was higher than that of unamended tailings.
- Hydraulic conductivity of tailings paste was reduced as a consequence of GLD and FA addition, as the porosity in the tailings was filled by the additives.
- If the following prerequisites such as reducing of porosity, improvement of water retention capacity and no cracks in the tailings are ensured, free oxygen transports can be limited.

Acknowledgement

The authors wish to thank the Swedish Research Council for Environment, Agricultural Sciences and spatial planning (FORMAS ID 2008-268-12239-54), the European Union Structural Funds, the Northern Sweden Soil Remediation Centre, EDF Objective 2, Contract MCN IO No 43173, Sveriges Ingenjörers environmental fund and the Centre of Advanced Mining and Metallurgy (CAMM) for financial support.

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