Evaluation of mixtures of green liquor dregs and till for application as a sealing layer on sulfidic mine waste.

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Abstract

The oxidation of sulphides in mine wastes is a possible threat to the environment as it might have potential to generate acid and release metals. Recent research has shown that a residue from the paper production, Green Liquor Dregs (GLD) has properties suitable for a sealing layer on top of mine waste, i.e. it is fine grained and has a low hydraulic conductivity (HC). A sealing layer mitigates oxidation of metals by deterring oxygen to enter the waste. In this study, sieved till (<20mm) with fines (<63 µm) of 30-40 % was mixed with 5, 10 and 15 wt. % of GLD. HC and maximal density after compaction were determined for each mixture. The maximal density in the sieved till studied decreased with increasing addition of GLD (1.95 to 1.89 t/m^3). The decrease is likely due to that the high flow stress and the low shear strength of the GLD inhibits compaction. Increasing water content and amount of GLD in the mixture prevents compaction of the samples, leading to an increase in HC ($5.5*10^{-10}$ to $2.2*10^{-9}$ m/s). The study concludes that an addition of 5 % GLD is acceptable in a fine grained till regarding compaction properties and HC.

Keywords: Green Liquor Dregs; Sealing layer; Mine waste; Acid mine drainage; Industrial residues

1 Introduction

The oxidation of sulphides in mine wastes and the production of acid rock drainage (ARD) is a major long-term threat to the environment as metals may become mobile with access to oxygen (Saria et al 2006). One method to reduce sulphide oxidation is to apply a dry cover on top of the mine waste deposit. A dry cover usually consists of a sealing layer placed on top of the mine waste and above this, a protective layer. The sealing layer is usually made of till and its purpose is to mitigate oxidation of metals by reducing infiltrating water and oxygen to reach the mine waste. To fulfil these requirements, the sealing layer should be compacted to a high density which in turn leads to lower hydraulic conductivity (Höglund et al 2004). Studies conducted by Leroueil et al (2002) and Watabe et al (2000) conducted on a glacial till shows that the hydraulic conductivity is highly dependent on the degree of compaction, with significantly decreasing values with decreased void ratio during loading. The presence of fine grained material is also an important factor that influences the hydraulic conductivity (HC). An increasing amount of clay in the material decreases the HC (Leroueil et al 2002).

Previous studies have shown that a residue from pulp and paper production, Green Liquor Dregs (GLD), has properties suitable for a sealing layer i.e. it is fine grained (d100 < 63µm) and commonly has a HC in the range of 10^{-8} and 10^{-9} m/s (Mäkitalo et al 2014). Other characteristics of GLD are a high pH (10-11), relatively high porosity (73 - 82 %), a bulk density of 0.44-0.67 g/m^3, a compact density of 2.47 to 2.60 g/cm^3 and consist of up to 75 % of $CaCO_3$ (Mäkitalo et al 2014). The high $CaCO_3$ generates from the retrieving process where a pre-coat lime mud filter (mixture of $CaCO_3$, CaO and $Ca(OH)_2$) is used leading to various amounts of lime mud mixed with the green liquor. This strongly influences the composition of the GLD. Due to its high carbonate content GLD has a high buffering capacity (Mäkitalo et al 2014). A high buffering capacity is a good property for a sealing layer over mining waste, as it can act as an alkaline barrier. Furthermore, GLD is classified as a non-hazardous chemical waste by the Swedish EPA (SFS 2001). However, to solely use GLD in the sealing layer is not reasonable neither from economical or a geotechnical point of view. In a geotechnical perspective GLD is not suitable due to its stickiness and low shear strength, which makes it difficult to compact (Mäkitalo et al 2014).

Previous field studies have shown that mixing till with GLD can improve both of the materials properties as sealing layer (Mácsik and Maurice 2015). A pilot scale experiment using a till/GLD mixture was conducted in 2014, where a pilot landfill (50 x 10 m) in Boden, Sweden was constructed to test production at industrial scale. The results from the constructed pilot cell are to be evaluated.

In this study GLD was mixed with sieved (<20 mm) fine grained (c. 40 % <0.063 mm) till. The aim of the study was to find the optimal percentage of GLD to add to a sieved fine grained till and to improve the materials properties considering HC and compaction. The purpose was to be able to use the mixture of till and GLD as a sealing layer in a dry cover application over mine waste.

2 Materials

Till was collected at a till carrier at the Brännkläppen waste plant in Boden (northern Sweden) after the first frost. The material was collected in 10 liter plastic buckets with a lid. At the laboratory, the till was sieved through a 20 mm sieve and air dried in the buckets (lids off) to lower its water content.

GLD from Smurfit Kappa paper mill in Piteå (northern Sweden) was collected in sealed plastic containers to preserve the water content of the material. The GLD from Smurfit Kappa has previously been characterized and the summary is presented in table 1 (Jia et al 2014).

Table 1. Characterization of GLD from Smurfit Kappa paper mill. The values for the chemical composition are expressed as mean ± SE (n = 2), where SE is standard error, TS total solid, LOI loss on ignition. The values for the physicochemical characterization are expressed as mean ± SD (n = 3), where SD is standard deviation (Jia et al 2014).

TS	(%)	52.1 ± 0.1	TiO_2	(TS %)	0.012 ± 0.002	Hg	(mg/kg)	<0.03
SiO_2	(TS %)	0.84 ± 0.01	LOI	(TS %)	41.3 ± 0.1	Mo	(mg/kg)	<0.5
Al_2O_3	(TS %)	0.49 ± 0.01	As	(mg/kg)	<1.0	Ni	(mg/kg)	41.3 ± 0.8
CaO	(TS %)	41.7 ± 0.3	Ba	(mg/kg)	316 ± 1	Pb	(mg/kg)	3.0 ± 0.1
Fe_2O_3	(TS %)	0.29 ± 0.01	Be	(mg/kg)	<0.5	S	(mg/kg)	9,335 ± 361
K_2O	(TS %)	0.18 ± 0.00	Cd	(mg/kg)	3.6 ± 0.1	Zn	(mg/kg)	812 ± 11
MgO	(TS %)	6.5 ± 0.1	Co	(mg/kg)	2.8 ± 0.1	pH		11.0 ± 0.0
MnO	(TS %)	1.2 ± 0.0	Cr	(mg/kg)	66.9 ± 0.4	Eh	(mV)	244.8 ± 9.9
Na_2O	(TS %)	2.37 ± 0.03	Cu	(mg/kg)	109 ± 3	EC	(mS/cm)	18.0 ± 1.2
P_2O_5	(TS %)	0.86 ± 0.00						

3 Methods

3.1 Sieving

The till was washed and dry sieved according to SS-EN 933-1:2012 to obtain the weight percentage of fines in the material. After washing, the material was mechanically sieved for 10 min with a Retsch AS 200 sieve with an amplitude of 2.2 mm/"g". The sizes of the sieves were 12.5, 10, 8, 5, 4, 2, 1, 0.5, 0.25, 0.125 and 0.063 mm. Five replicates were used to determine the particle size distribution.

3.2 Mixing of till and GLD

The sieved till was mixed with 5, 10 and 15 wt. % of GLD. The mixing was carried out by passing the material five times through a riffle splitter with eight 3.6 cm chutes.

3.3 Total solids content

The TS-content (total solids content) of three replicates of each material and mixtures were decided by drying them in an oven (105°C for 24 h) according to the SIS standard SS-EN 14346:2007.

3.4 Proctor compaction

Proctor compaction was carried out according to standard SS-EN 13286-2:2010. For the proctor compaction experiments the till was air dried at least 24 hours prior to mixing with the naturally moist GLD. The dry density (ρ_d) and the water content (w) were then calculated with equation 1 and 2:

$$\rho_d = m/((1+w)+V) \quad (1)$$

$$w = (m-m_d)/m_d \quad (2)$$

where m is the mass of the moist sample, m_d is the mass of the dry sample and V is the volume.

3.5 Hydraulic conductivity

HC measurements were conducted on a moist till (TS 90.4±0.4) with 5, 10 and 15 wt. % added GLD. The constant head-method was used in air tight cylinders with a volume of 943 cm^3. The walls of the cylinders were sealed with a thin layer of benthonite. The mixtures inside the cylinder were compacted with proctor compaction in five equally thick layers with a falling weight of 4.54 kg, falling 45 cm 25 times on each layer. The mixture with 15 % GLD was hand compacted, due to high water content which made proctor compaction impossible. The hand compaction was done in a similar way as proctor compaction (five layers with 25 "hits" each).

Water was lead to the bottom of the cylinder with a hydraulic gradient of 150 cm. The water passing through the cylinder was collected in a plastic bottle, sealed from the top to prohibit evaporation. The plastic bottle was weighed regularly and the time was noted to measure the velocity of the water passing through the sample. HC was calculated using Darcy's law.

4 Results

4.1 Particle size distribution

The particle size distribution measurement of the sieved till, show a percentage of fines (<0.63 µm) between 34 and 41 %.

4.2 Total solid-content

The total solid contents (TS) of the till was 90.4±0.3 %, GLD had 40.8±1.6 %, till with 5 % GLD had 88.0±0.6 %, till with 10 % GLD had 85.5±1.6 % and till with 15 % GLD had a TS content of 83.5±0.1 %.

4.3 Proctor compaction

The performed proctor compaction shows that the highest dry density was reached in the mixture with the lowest weight percentage (5%) of added GLD (1.95 g/cm^3). The highest density that can be reached in the different mixes decreases with increasing addition of GLD in the mixture; 1.95, 1.94 and 1.89 g/cm^3 adding respectively 5, 10 and 15 % of GLD (Figure 1).

The optimal water content was approximately 9 % in all three mixtures (Figure 1). However the optimal water content after compaction for the mixture with 5 % GLD ranged between 5-10 %, and between 9-14 % for the mixture with 15 % GLD added. The difference between 5 and 10 % GLD was small and the materials behaved similar to proctor compaction. When the water content of the mixtures increased above 14 %, all three mixtures behaved similarly to proctor compaction.

4.4 Hydraulic conductivity

The HC increased with increasing addition of GLD in the mixtures (Figure 2). With 5 % GLD added the HC was 5.5*10^{-10}±0.7*10^{-10} m/s (average value after stabilization), with 10 % GLD added 1.6*10^{-9}±0.2*10^{-9} m/s and with 15 % GLD added the HC was 2.2*10^{-9}±0.2*10^{-9}m/s.

The dry density after compaction of the HC samples decreased with increasing percentage of GLD added to the till, 1.90 g/cm^3 in the mixture with 5 % GLD, 1.84 g/cm^3 in the mixture with 10 % of GLD and 1.74 g/cm^3 in the mixture with 15 % GLD.

The water content of the HC samples increased with increasing percentage of GLD added. 14, 15 and 20 % for 5, 10 and 15 % added GLD respectively. The water content of the mixtures was higher than the optimal water content which was obtained by proctor compaction (Figure 1).

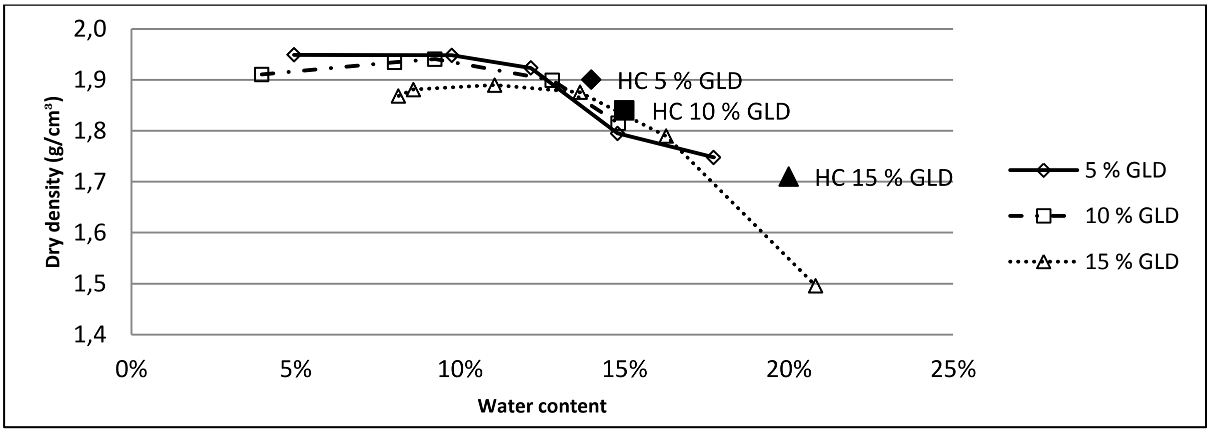

Figure 1. Dry density with increasing water content after proctor compaction in sieved fine grained till with 5, 10 and 15 wt. % of added GLD. The optimal density decreases with an increasing percentage of GLD added to the till. The filled dots marked with HC represent the samples which were proctor- (5 and 10 % GLD) and hand compacted (15 % GLD) for hydraulic conductivity.

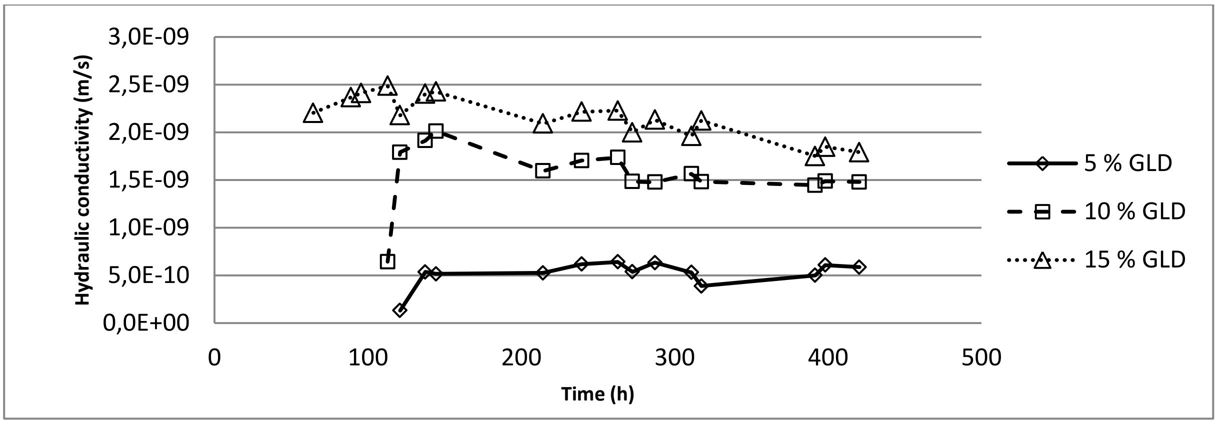

Figure 2. Hydraulic conductivity (HC) in till with 5, 10 and 15 wt. % of added GLD. The HC increases with increasing percentage of GLD added to the till.

5 Discussion

The proctor compaction curve in Figure 1 shows that the mixtures are more difficult to compact the more GLD is added to the sieved fine grained till (< 20 mm). This is likely due to the properties of the GLD, i.e. its high water content, low shear strength (Mäkitalo et al 2014) and high flow stress (0.8 – 2.00; unpublished data). The high porosity and water content of the GLD leads to liquefaction when intensive compaction energy is applied and water is released which makes the material difficult to compact (Mäkitalo et al 2014). However, the difference between 5 and 10 % GLD addition was low, which indicate that the threshold for the fine grained till used in this study appears to be 10 % of added GLD seen to compaction properties.

The HC increased with increasing percentage of GLD added to the sieved fine grained till (Figure 2). Only the mixture with 5 % GLD ($5*10^{-10}$ m/s) was below the recommended minimum HC of $1*10^{-9}$ m/s in a sealing layer (Höglund et al 2004). In contradiction to this study a decrease of the HC was expected with a higher fraction of GLD in the mixture due to increased amount of fine grained material. A study conducted by Leroueil et al (2002) showed that the HC of glacial till is very sensitive to minor changes in clay-sized particle fractions with decreasing values with increasing amount of clay. However, in this study the HC did not decrease with increasing amount of fine grained material. Likely due to decreased degree of compaction as a result of increased water content as more GLD was added. The till used in this experiment was already moist (TS c. 90 %) and with added GLD (TS c. 41 %) the water content of the mixtures increased to 14 - 20 %. A water content of this magnitude is higher than the optimal water content in the mixtures (Figure 1), resulting in a decreasing dry density after compaction. This in turn may increase the HC. HC has shown to be highly dependent on the degree of compaction, with decreasing HC with increasing degree of compaction (Leroueil et al 2002; Watabe et al 2000). However, low HC values cannot be reached if the material is unable to be compacted due to excessively high water content. As discussed above the properties of the GLD is likely also contributing to the materials incapability to compact.

It is important to note that the mixture with 15 % GLD was compacted with hand, compared to a proctor machine which was used for the mixtures with 5 and 10 % GLD. Proctor compaction was not possible to use for 15 % GLD, as the material flowed over the walls of the cylinder when compacted because of excessively high water content in the mixture. Hand compaction might have led to a higher degree of compaction compared to proctor compaction which may have underestimated the HC. When compacting by hand, it is possible to compact closer to the cylinder walls, increasing the degree of compaction of the overall sample which in turn decreases the HC.

The specific GLD used in these experiments was not characterized in this study, but assumed to have similar characteristics as the GLD from the same paper mill characterized by Jia (2014).

Supplementary research with a drier till and a less fine grained till will be conducted in future studies for optimal percentages of GLD for different properties of the till. Furthermore, the HC and the compaction properties of the till and the GLD in itself will be investigated. The GLD will also be further characterized chemically and physically.

6 Conclusions

- A moist fine grained (~ 40 % <63 µm, TS ~ 90 %) till mixed with 5 % GLD fulfil the requirement as a sealing layer in a dry cover application. When more GLD is added to the mixture the hydraulic conductivity increases.
- The compaction properties for the mixtures deteriorate with increasing amount of GLD added to the sieved fine grained till. The optimal percentage of added GLD is 5-10 % considering compaction properties.
- The optimal water content in the sieved fine grained till-GLD mixture is around 9-10 % if it is to be compacted.

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References

Höglund, L.-O., Herbert, R., Lövgren, L., Öhlander, B., Neretniks, I., Moreno, L., Malmström, M., Elander, P., Lindvall, M., Lindström, B. MiMi—Performance Assessment, Main Report. MiMi-Report 2003:3. MiMi Print. Stockholm, Sweden. (2004).

Jia, Y., Maurice, C., Öhlander, B.: Metal Mobilization in Tailings Covered with Alkaline Residue Products: Results from a Leaching Test Using Fly Ash, Green Liquor Dregs, and Lime Mud. Mine Water Environment (2014).

Leroueil, S., Le Bihan, J.-P., Sebaihi, S., Alicescu, V. Hydraulic conductivity of compacted tills from northern Quebec. Canadian Geotechnical Journal. 39, pp 1039-1049. (2002).

Mácsik, J., Maurice, C.: (in Swedish) Teknisk Rapport. Inblandning av GLS vid tillverkning av tätskikt för efterbehandling av gruvavfall. Dimensioneringskriterier och kvalitetskrav. Stockholm/Luleå, pp 40. (2015).

Mäkitalo, M., Maurice, C., Jia, Y., Öhlander, B.: Characterization of green liquor dregs, potentially useful for prevention of the formation of acid rock drainage. Minerals 4, pp 330-344. (2014).

Saria, L., Shimaoka T., Miyawaki K.: Leaching of heavy metals in acid mine drainage. Waste manage Res. 24, pp 134-140. (2006).

SFS: Swedish Ordinance of Waste. Sweden Ministry of Environment. Stockholm, Sweden (in Swedish). (2001).

Watabe, Y., Leroueil, S., Le Bihan, J.-P. Influence of compaction conditions on pore-size distribution and saturated hydraulic conductivity of a glacial till. Canadian Geotechnical Journal. 37, pp 1184-1194. (2000).