Green liquor dregs-amended till to cover sulfidic mine waste

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Cover image:

Picture of the GLD deposition at BillerudKorsnäs papermill in Karlsborg, Sweden.
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Abstract

The mining industry produces massive amounts of waste that without treatment and in contact with oxygen can result in acid rock drainage (ARD) and increased leaching of metals. In Sweden, the common way to manage the mine waste after closure is to apply a dry coverage on top of the waste deposition. The access to a suitable cover-material is however limited and can lead to extensive costs for the mining company. This creates a great driving force for alternative solutions, e.g. bentonite amendment to till. However, bentonite production is costly both economically and environmentally due to time- and resource-consuming production. Recycling of industrial residues as a cover material is another option. The recycling is beneficial for many parts, i.e. the industry producing the residue, the mining industry that will use the residue and the society in its strive for zero waste. An industrial residue that has potential to improve the sealing layer qualities of a local till is Green Liquor Dregs (GLD), a residue from pulp production in a paper mill. In this study 5 to 20 wt. % of GLD from two different paper mills, with varying total solid content (TSC) and particle size distributions (PSD), were mixed with three tills also with varying PSD and TSC. The objectives of the study were to investigate if GLD addition can improve a till so it can be used in a sealing layer, how the fines- and clay content in the tills affects the hydraulic conductivity (HC) and the water retention capacity (WRC) of the till-GLD mixtures, and how the initial water content will affect the HC in the different mixtures. The HC of the mixtures based most of the tills studied improved, but not enough to reach the required $10^{-8}$ m/s. Only the material based on a more clayey silty till had an HC below $10^{-8}$ m/s, even if it increased with the addition of GLD. The WRC, on the other hand, shows promising results, especially for 15 wt. % GLD addition. An increase in WRC is seen the more GLD is added. However, due to compaction difficulties more than 20 wt. % addition is not recommended. There is a difference in both HC and WRC between the mixtures based on the different tills, and a characterization of the till as a bulk material and its mixture with GLD is suggested before using it in mine remediation purposes. The HC of the till is affected by the content of fines and especially clay. The results from the WRC is more complicated and a mineralogical study on the GLD and the clay fraction of the tills is recommended. This study also shows that the initial water content of the materials rather than dry density has an impact on the HC of the mixtures. The initial water content determines the degree wet of optimum that in this and previous studies has shown to greatly affect the HC. A drier till and GLD leads to that more GLD should be added to the mixture when reaching for optimal HC. Even if the study did not result in low enough HC in most of the cases, the WRC shows promising results and is the most important parameter minimizing the oxygen diffusion to the mine waste. The greatest challenge when working with GLD is, however, its heterogeneity which makes it difficult to predict how it will behave in a sealing layer. Even though, the recycling of GLD in mine waste remediation should be strived for as it is beneficial for both the industry that provides the residue and the mining company. It is also a great profit for the environment as it minimizes the waste disposed and reduce use of natural resources.
List of papers

This licentiate thesis includes a synthesis chapter and the following two manuscripts:

I. Nigéus, Susanne. and Maurice, C. 2018. Opportunities to use Green Liquor Dregs as an amendment in till to cover sulfidic mine waste (to be submitted).

II. Nigéus, Susanne. and Maurice, C. 2018. The drivers of hydraulic conductivity in till-GLD mixtures to be used in a sealing layer on top of mine waste (to be submitted).

The following articles represent international conference contributions related to the thesis, but are not included in it:


3. Nigéus, Susanne. and Maurice, C. 2018. Monitoring a field application of a Green Liquor Dregs-till mixture in a sealing layer on top of sulfidic mine waste. Accepted in: 11th International Conference on Acid Rock Drainage (ICARD), Pretoria, South Africa.
Abbreviations

ARD – Acid Rock Drainage
e - Void ratio
Dv – Diffusion coefficient
GLD – Green Liquor Dregs
GARD-guide – Global Acid Rock Drainage guide
HC – Hydraulic conductivity (m/s)
LC – Light Proctor compaction
LTU – Luleå University of Technology
m – Mass of a moist sample (g)
m0 – Mass of a dry sample (g)
mR – Mass of the ring (g)
mR+w – Mass of the wet sample and the ring (g)
n – Porosity
PC – Proctor compaction
PSD – Particle size distribution
RD – Compaction degree
Sr – Water saturation (%)
SWCC – Soil water characteristic curve
TSC – Total solid content
V – Volume (cm³)
Vv – Void volume (cm³)
Vs – Soil volume (cm³)
Vt – Total volume (cm³)
Vw – Water volume (cm³)
w – Gravimetric water content (%)
wL – Wilting point
wP – Plasticity index
WRC – Water retention capacity
wt. % – Weight percentage
ψ – Matric suction (mH₂O)
ψa – Air entry suction (mH₂O)
ψ85 – Matric suction for 85 % Sr (mH₂O)
ρd – Dry density (g/cm³)
ρr – Relative density of the grains (g/cm³)
θ – Volumetric water content (%)
θr – Residual volumetric water content (%)
θns – Near saturated volumetric water content (%)
θe – Effective saturation (%)
α – Fitting parameter van Genuchten equation
n – Fitting parameter van Genuchten equation
m – Fitting parameter van Genuchten equation, = 1-n⁻¹
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1 Introduction

Massive amounts of mine wastes are generated worldwide. Sweden alone generated 110-million-ton mine waste in 2017, accounting for 77% of all waste produced during that year (Swedish EPA 2018). These wastes need to be managed in an environmentally-, technically- and economically sustainable way. About 70% of the mine waste in Sweden contains sulfide minerals (SGU and Swedish EPA 2017). If sulfidic mine waste is left in contact with oxygen and humidity the sulfides oxidize and have the potential to produce acid rock drainage (ARD; Figure 1).

Figure 1. Descriptive figure of acid rock drainage (ARD).

ARD is a major long-term threat to the environment as it is associated with low pH and high concentrations of metals and metalloids (Akcil and Koldas 2006; Saria et al. 2006). Weathering of minerals is a natural process, with increasing intensity of the processes in mining creating new surfaces. ARD is generated by the following reactions (Akcil and Koldas 2006):

\[
\text{FeS}_2 + \frac{7}{2}\text{O}_2 + \text{H}_2\text{O} \rightarrow \text{Fe}^{2+} + 2\text{SO}_4^{2-} + 2\text{H}^+ \tag{1}
\]

With further oxidizing conditions the Fe\(^{2+}\) can oxidize to Fe\(^{3+}\) by the following reaction:

\[
\text{Fe}^{2+} + \frac{1}{4}\text{O}_2 + \text{H}^+ \rightarrow \text{Fe}^{3+} + \frac{1}{2}\text{H}_2\text{O} \tag{2}
\]

At relatively low pH Fe\(^{3+}\) precipitates as Fe(OH)\(_3\) and jarosite (KFe\(_3\)(OH)\(_6\)(SO\(_4\))\(_2\)):

\[
\text{Fe}^{3+} + 3\text{H}_2\text{O} \rightarrow \text{Fe(OH)}_3\text{, solid} + 3\text{H}^+ \tag{3}
\]

The remaining Fe\(^{3+}\) in solution after reaction 3 may be used to oxidize additional pyrite according to the following reaction:

\[
\text{FeS}_2 + 14\text{Fe}^{3+} + 8\text{H}_2\text{O} \rightarrow 15\text{Fe}^{2+} + 2\text{SO}_4^{2-} + 16\text{H}^+ \tag{4}
\]

A combination of the reaction 1-3 is:
FeS₂ + \frac{15}{4}O₂ + \frac{7}{2}H₂O \rightarrow Fe(OH)₃ + 2SO₄²⁻ + 4H⁺ \quad (5)

1.1 Soil cover solution

There are several ways to prevent ARD both in situ while the mine is still operating and after mine closure. The GARD guide (Global Acid Rock Drainage guide; Verburg et al. 2009) categorize different methods to prevent ARD after closure, into two main categories; engineered barriers and water covers (Figure 2). Engineered barriers can be divided into liners and dry covers, where liners are typically designed to act as a barrier for contaminant flow from the overlying waste into the receiving environment. Dry covers are typically designed to limit the ingress of water and oxygen into the underlying waste. Under the relatively humid climatic conditions in Sweden a soil cover can be used to reduce oxygen flux to the underlying reactive wastes, and thus reduce ARD (Collin and Rasmuson 1990; Bussière et al. 2003; Dagenais et al. 2006). A typical soil cover in Sweden consists of a sealing layer made of a compacted fine-grained material with a low HC and high WRC. On top of the sealing layer, an uncompacted protection layer is placed, usually made of till. The function of the protection layer is to protect the sealing layer from penetration of roots and frost. Above the protection layer, a nutrient-rich vegetation layer is usually placed, to easier reestablishment of vegetation (Figure 3). A sealing layer in Sweden is usually made of a clayey till. The availability of a clayey till nearby a mine is however often limited, and till can either be improved or replaced to construct a sealing layer. There is, therefore, a great need for alternative solutions, e.g. bentonite amendment to till. However, bentonite production is costly both economically and environmentally due to time- and resource-consuming production. Recycling of industrial residues as a sealing layer material is another option. Sweden produces 28 million ton wastes, excluding mine waste, of which about 26 million ton is classified as a non-hazardous waste by the Swedish EPA and of that 6 million ton is residues from industrial production (Swedish EPA 2016). Most of these residues are deposited and only a small amount recycled. Using an industrial residue in a mine remediation program is useful for both the mining industry and the industry providing the residue and a large benefit for the environment as less waste is being deposited. The system serves as a circular economy as the industries aim towards reducing waste deposition. Some examples of industrial residues that previously have been used as a mine waste cover are sewage sludge, fly ash, desulfurized tailings, coal combustion by-products (CCB) and steel slag (Hallberg et al. 2005; Bäckström et al. 2009; Dobchuk et al. 2013; Lu et al. 2013; Nason et al. 2013; Park et al. 2014; de Almeida et al. 2015; Phanikumar and Shankar 2016).
Figure 2. Different methods to prevent ARD from mine waste according to the GARD-guide (Verburg et al. 2009). The typical method in Sweden is a soil cover (red circle).

Figure 3. A describing figure of a typical soil cover system in Sweden.
1.2 Green Liquor Dregs

Previous studies have shown that a residue from pulp production, Green Liquor Dregs (GLD), has properties suitable for a sealing layer i.e. it is fine-grained (d100 < 63μm), commonly has an HC in the range of 10⁻⁸ and 10⁻⁹ m/s and a higher water retention capacity (WRC) compared to materials with similar particle size, such as clayey/sandy silt (Mäkitalo et al. 2014). A general trend in the pulp and paper industry to minimize landfill disposal is seen (Monte et al. 2009) due to extensive costs (Eroglu et al. 2006). This is a great driving force for the industries to find recycling options for their waste. In 2003 Sweden produced about 200 000 tons of GLD annually (SGI 2003), and production has slightly increased based on a survey made 2017 (unpublished data).

GLD is an alkaline, inorganic waste originating from the recycling process at sulfate pulp and paper mills. Pictures of GLD and an overview of the generating process can be seen in Figure 4 and Figure 5. The main solid compounds of GLD consists of CaCO₃, Mg(OH)₂, C and metal sulfides, especially FeS (Sanchez and Tran 2005; Jia et al. 2014). The liquid phase of the GLD consists of Na₂CO₃ and NaOH, which generates its characteristic high pH. GLD is regarded as an inert material (Mäkelä and Höynälä 2000) and has the same grain size distribution as silt (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/m³, a compact density of 2.47 to 2.60 g/cm³, a low HC (10⁻⁸ and 10⁻⁹ m/s) and a high WRC (Mäkitalo et al. 2014). Sequential extraction has been performed on GLD and indicates relatively low bioavailability of metals in general (Nurmesniemi et al. 2005). GLD has been used in a few sanitary landfill cover applications (Pousette and Mácsik 2000; Hargelius 2008) and the possibility to use GLD as a cover material on top of mine waste have been investigated in several pilot-scale studies (Chtaini et al. 2001; Lu et al. 2013; Ragnvaldsson et al. 2014). The studies show a reduced effluent volume by 40% (Ragnvaldsson et al. 2014), a reduction of leaching metals (Chtaini et al. 2001; Lu et al. 2013; Ragnvaldsson et al. 2014) and reduced oxygen diffusion to the mine wastes (Chtaini et al. 2001). A long-term stability of the GLD has been studied by Mäkitalo et al. (2016). The study concluded that aging of GLD mainly depends on the amount of water passing through the material, which is changing its pH and chemical composition, primarily by leaching S and K. The GLD did not show any physical or mineralogical changes in the study that will affect its ability to prevent oxygen diffusion to the underlying mine waste. The WRC observed to be >85% under field conditions and maintained a high degree of saturation with low HC independently of its age. The shear strength of the GLD increased over time, but not enough to ensure a long-term physical stability. The main concern was expected to be the chemical stability of the GLD due to the high content of calcite, which might dissolve and profoundly change its properties. However, Mäkitalo et al. (2016) saw from estimations of the density and HC of the GLD as well as their performance in the batch leaching experiments, no evidence that this will occur within the next thousands of years.

Considering its lack of long-term physical stability, due to a low shear strength, it is not reasonable to solely use GLD in the sealing layer where there are slopes, from a geotechnical point of view. Neither is it reasonable from an economic point of view, due to its relatively high transportation costs, which are affected by its high water-content (leading to higher weight) and
relatively long distances to the mine sites (Alakangas et al. 2014). Previous field studies on mixing GLD with till have shown promising results for the mixtures to be used as a sealing layer, with low HC, high WRC and increased compaction properties (Hargelius 2008; Jia et al. 2013; Mäkitalo et al. 2015a).

At Luleå University of Technology (LTU) several studies on GLD has been conducted, mainly by Maria Mäkitalo (Mäkitalo et al. 2014; Mäkitalo 2015; Mäkitalo et al. 2015a; Mäkitalo et al. 2015b; Mäkitalo et al. 2016). An overview of previous-, on-going- and future studies on GLD at LTU is presented in figure 6.

Figure 4. Overview of the production process that generates green liquor dregs (Reworked from Mäkitalo 2015).
Figure 5. Pictures of GLD from four different paper mills, from top left to bottom right; Iggesund, Smurfit Kappa, Vallvik and BillerudKorsnäs papermill in Sweden. The bottom two pictures are also from BillerudKorsnäs paper mill.
1.3 Factors affecting the function of a sealing layer

The main function of the sealing layer is usually to limit oxygen diffusion through the cover. Oxygen has two main ways of transport in a porous medium, dissolved in the pore water or in gaseous form in the unsaturated porosity. Often, oxygen diffusion coefficient of the sealing layer material is not measured, and hydraulic conductivity (HC) is usually used as an indicator to evaluate the effectiveness of a sealing layer as it is easier to measure. The requirements for HC in a sealing layer is site-specific but in Sweden commonly set to $10^{-8}$ m/s. To reach a low HC the sealing layer should be compacted to a high density (Höglund et al. 2004). Studies by Leroueil (2002) and Watabe (2000) on glacial till shows that the HC is highly dependent on the degree of compaction, with significantly decreasing values with decreased void ratio during loading.

Another feature connected to compaction that is affecting the HC is the molding water content of the material (Benson and Trast 1995; Leroueil et al. 2002), i.e. the water content after compaction. A study conducted by Benson and Trast (1995) on thirteen compacted clays shows that the lowest HC was reached at molding water content of 1-2 % wet of the line of optimums. The optimum water content is the molding water content at which the highest dry density can be reached. The presence of fine-grained material is another important factor that influences the HC. An increasing amount of fines in the material decreases the HC (Benson et al. 1994; Benson and Trast 1995; Leroueil et al. 2002), as the micro-porosity of the material decreases. In a study on tills conducted by Leroueil et al. (2002) the HC decreased several orders of magnitude as the clay-size fraction increased from 2 to 12 %. Also, a study on clay conducted by Benson et al. (1994) showed a strong relationship between clay content and HC and a weak relationship between the contents of fines and HC.

For material that is to be used as an oxygen barrier on top of mine waste, the water retention capacity (WRC) is another important feature, as high WRC usually corresponds to a high water-saturation ($S_i$). The transport of gaseous oxygen through fine-grained materials are mainly by molecular diffusion (Yanful 1993). At a relatively low degree of saturation, most oxygen transport occurs through the partially air-filled pores (Aachib et al. 2004), due to the 10 000 times higher diffusion coefficient ($D_o$) in the air than in water (Yanful 1993). In addition to a higher diffusion coefficient, the dissolved $O_2$ concentrations in the air are around 20 000 times higher than in water (Höglund et al. 2004; Verburg et al. 2009). In general, the oxygen flux rates are at a...
minimum when the degree of saturation is greater than 85–90 %, this as the air-phase at a saturation greater than 85 % becomes discontinuous (Corey 1957). The oxygen is then transported through the water phase (Aubertin and Mbonimpa 2001; Aachib et al. 2004) and in a layer that is kept close to saturation, the $D_1$ in the soil can be comparable to the $D_1$ in water. It is then small enough to reduce the oxygen flux to a level comparable to that of a water cover (Yanful 1993; Aachib et al. 2004).

One way to illustrate the WRC is to use a soil-water characteristic curve (SWCC), which shows the correlation between matric suction ($\psi$) and water content (gravimetric $[w]$ or volumetric $[\theta]$) or degree of saturation ($Sr$; Figure 7:A). The typical SWCC consist of three stages: capillary saturation, desaturation and residual saturation (Figure 7:A; Sillers et al. 2001). The matric suction required to remove water from the largest pores corresponds to the break in the SWCC, where the soil is nearly saturated ($\theta_s$; Figure 7:A). This break in the curve is generally referred to as the air-entry suction ($\psi_a$; Figure 7:A). The water content corresponding to the asymptote of the SWCC at low degrees of saturation is called the residual water content ($\theta_r$; Figure 7:A). The SWCC is hysteretic, which means that matric suction for desorption (drying) is higher than for sorption (wetting), for a given water content (Figure 7:B; Tinjum et al. 1997). This so-called hysteretic effect is caused by size differences between the bulk-pores and the interface-pores that connects the main-pores, which changes the contact angle between water and air during wetting (Tinjum et al. 1997). Due to experimentally practical reasons usually only the desorption curve is used when measuring WRC in laboratory (Tinjum et al. 1997). The shape of the SWCC is related to the soil type. Soils with smaller pores and finer particles usually have a higher $\psi_a$ (Figure 7:C; Fredlund and Rahardjo 1993; Tinjum et al. 1997; Sillers et al. 2001). Also, the slope of the curve in the desaturation zone tends to become flatter as the soil particles become finer (Figure 7:C; Sillers et al. 2001). Except from particle size distribution also the molding water content is known to affect the SWCC of fine-grained soils, with a higher $\psi_a$ and a steeper slope for suctions greater than the $\psi_a$ as the molding water content increases (Tinjum et al. 1997; Vanapalli et al. 1996; Vanapalli et al. 1999). Vanapalli et al. (1999) studied the SWCC in fine-grained soils and found that if they are compacted dry of optimum water content, steeper SWCC are generated compared to those compacted at or wet of optimum water content. Dry of optimum the specimen is influenced by its macrostructure and acts like a coarse-grained soil due to aggregation. When compacted wet of optimum the microstructure is more important, leading to a higher $\psi_a$ than in soil compacted dry of optimum (Vanapalli et al. 1999). In addition to molding water content, the initial density has a significant impact on the SWCC due to a decrease in the volume of voids with increasing dry density, leading to faster saturation (Vanapalli et al. 1999).
1.4 The objectives and hypotheses of his study

In this study 5 to 20 wt. % of GLD from two different paper mills were mixed with three sieved tills (<20 mm) to investigate if GLD addition can improve the HC and WRC of till that alone does not meet the in Sweden commonly required HC of $10^{-8}$ m/s. The study also investigated how the content of fines and clays affect the HC and WRC of the different mixtures and how initial water-content affect the HC. Maximum 20 % GLD addition was set as a limit as a mixture with more GLD becomes difficult to compact and handle, due to increased water content over the optimum molding water content and decreased shear strength. Figure 8 shows an overview of the different mixtures used and the analyses performed on them.

The HC in tills, which alone does not reach a low enough HC, was expected to decrease below $10^{-8}$ m/s with addition of GLD, to make it suitable for a sealing layer. The GLD consist of material in the silt fraction which is expected to fill the pores of the till leading to a decreased HC. However, after a certain limit of GLD addition (10-15 wt. %), the high water-content and low shear strength of the GLD is expected to decrease the compaction degree and therefore increase the HC in the mixtures. The till as a heterogenic material and a dominant part of the mixture is expected to have a significant impact on the HC and to some extent also the WRC of the mixtures. The finer the till is, the lower HC and higher WRC in the mixture are expected. The WRC of the mixtures is expected to increase with higher amount of GLD added and the GLD addition is expected to have a greater influence on the WRC of the mixtures than the PSD of the tills. This due to the high initial WRC of the GLD. A decrease in initial water-content of till and GLD is expected to decrease the HC with higher amounts of GLD added, as the molding water-content will decrease to a value closer to the optimal water-content. The mixtures are expected to reach the lowest HC at water contents a few percent wet of the optimum water-content.

The objectives of the laboratory study were to find out (i) if GLD can improve the HC and WRC of a till that is to be used in a sealing layer on top of sulfidic mine waste, (ii) how the contents of fines and clays in three different tills will affect the HC and WRC in different tills.
GLD-mixtures, and (iii) how the initial water content of the materials affects the HC of the mixtures.

This study is of great importance as the access to a suitable cover-material is limited and can lead to extensive costs for the mining company. This creates a great driving force for alternative solutions where recycling of industrial residues is one example. The recycling is beneficial for many parts, i.e. the industry producing the residue, the mining industry that will use the residue and the society in its strive for zero waste.

**Figure 8.** An overview of the different mixtures of tills and GLD and the analysis performed on them. PC = Proctor compaction, HC = hydraulic conductivity, WRC = water retention capacity, w = water content and PSD = particle size distribution.
2 Materials and methods

2.1 Materials

Two silty tills and a sandy till was used as the bulk material. The silty tills were collected at two different till quarries at the Brännkläppen facility in Boden, Sweden. The sandy till was collected at a quarry at Sunderbyn, Luleå, Sweden. GLD from two different paper mills were used in this study; GLD1 from Smurfit Kappa paper mill in Piteå (Sweden) and GLD2 from Billerud Korsnäs paper mill in Karlsborg (Sweden). The GLD were collected in sealed plastic containers to preserve the water content of the materials. The two different GLD were chemically characterized by an accredited commercial laboratory (ALS Scandinavia AB in Luleå, Sweden) by a semi-quantitative screening analysis by ICP-SMS (HR-ICP-MS). The results are presented in Table 5 and the main constitutes of the two GLD are Ca, Na and Si, followed by Mg, K, Al and S. None of the metals are above the limits for the material to be classified as hazardous waste according to the Swedish Waste Management Association (SWMA 2007).

2.2 Mixing

The tills were air-dried (Figure 9:A) and sieved through a 20-mm sieve and the particles above 20 mm removed. This as particles above 20 mm will affect negatively on the effectiveness of the materials used as the sealing layer. Then the tills were mixed with 5, 10, 15 and 20 wt. % of GLD. The weight percentage was calculated towards a dry till and a naturally moist GLD. A naturally moist GLD was used as the material is challenging to rewet which affects its physical properties. Mixing was carried out by hand with a small shovel until the mixture was visibly estimated as homogenized. An overview of the different mixtures can be seen in Figure 8.

![Figure 9. Pictures of air drying of the till (A), the sieves used for PSD-analysis on the particles >0.063 mm (B), and the proctor compacting (C).](image)
2.3 **Particle size distribution**

A complete method description on the PSD is presented in article 1 and a picture of the sieving tower in Figure 9:B.

2.4 **Particle density**

A methods description of the particle density is presented in article 2.

2.5 **Proctor compaction**

A methods description on the PC-test performed is presented in article 2 and a picture of the experiment in Figure 9:C.

2.6 **Hydraulic conductivity**

HC measurements were conducted on a moist (TSC 91±1 %, n=24) and a drier (TSC 96 %) till with 5, 10, 15 and 20 wt. % addition of GLD. The TSC (total solid content) of each material were decided by drying them in an oven (105°C for 24 h) according to the SIS standard SS-EN 14346:2007. The results from the TSC are presented in Table 3. The constant head-method was used to measure HC (Figure 10) and a methods description is presented in article 2.

![Figure 10. Pictures of the HC-experiment stand and the equipment used.](image)

2.7 **Water retention capacity**

WRC was estimated by an SWCC, conducted with the results derived from a pressure plate extractor test described further in article 1.
3 Findings

The results from this study are summarized in Table 1 and Table 2.

3.1 The possibility for GLD to improve the HC and WRC of a till that is to be used in a sealing layer on top of sulfidic mine waste

The hypothesis was that the addition of GLD would improve the HC of the tills that alone would not reach the in Sweden common requirement of $10^{-8}$ m/s. However, after a certain limit of GLD addition (10-15 wt. %), the high water-content and low shear strength of the GLD was expected to decrease the compaction degree and therefore increase the HC in the mixtures. Unexpectedly only the mixture based on a more clayey till (silty till 2) had an HC below $10^{-8}$ m/s (Figure 11:A). The HC of the mixtures based on the silty till 1 and the sandy till did improve (decrease), but not enough to reach below $10^{-8}$ m/s. One reason for the unexpectedly low improvement of HC with the addition of GLD might be the method of compaction that was chosen, which is discussed further in section 3.4.1. The hypothesized decreasing trend followed by an increase is seen in this study. First, the HC of the silty till-1 decreased from 3.5E-08 to 1.5E-08 m/s at an addition of 5 wt. % GLD, to then increase up to 7E-08 m/s at up to 20 wt. % GLD in the mixtures (Figure 11:A). In the sandy till the HC decreased from 7E-08 to 2E-08 m/s when adding 5 to 10 wt. % of GLD. The HC then increased to 4E-08 m/s when adding up to 20 wt. % of GLD. HC in silty till-2 increased from 3E-10 to 5E-09 m/s with up to 20 wt. % of GLD addition (Figure 11:A). The decrease with low percentages of GLD addition was expected due to an increase in fine-grained material in the mixtures, leading to a decrease in micro-porosity (Benson et al. 1994; Benson and Trast 1995; Leroueil et al. 2002). With more than 10-15 wt. % of GLD addition, the HC increases. Likely due to a deterioration of the compaction degree with an increase in water content and lower shear strength (discussed further in section 3.3). Silty till 2 unexpectedly does not decrease with GLD addition. This might be due to its higher content of clay compared to the other two tills (discussed further in section 3.2). It might also be due to a different mineralogy compared to the, in PSD, similar silty till 1. It is therefore suggested that mineralogy of the clay fraction of the tills are analyzed in a future study.
Table 1: Summary of the results from the laboratory study conducted in this thesis, i.e. hydraulic conductivity (HC), compaction properties and fines/clay content.

<table>
<thead>
<tr>
<th>GLD</th>
<th>HC (wt.%)</th>
<th>Average HC (m/s)</th>
<th>Average ρd (g/cm³)</th>
<th>Average w (%)</th>
<th>Wet of opt. w (%)</th>
<th>Average max. ρd (g/cm³)</th>
<th>Opt. molding w (%)</th>
<th>Fines (wt.%)&lt;63μm</th>
<th>Clay size (%)&lt;2μm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silty till - GLD1</td>
<td>0%</td>
<td>3E-08±1E-09</td>
<td>9.8E-09</td>
<td>1</td>
<td>2.0±0.1</td>
<td>6</td>
<td>3.6±0.5</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>5%</td>
<td>2E-08±1E-09</td>
<td>1.97±0.02</td>
<td>10±0.6</td>
<td>2</td>
<td>2.05±0.01</td>
<td>8</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>10%</td>
<td>1.86±0.01</td>
<td>14±1.3</td>
<td>5</td>
<td>1.99±0.01</td>
<td>9</td>
<td>(n=2)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>20%</td>
<td>5E-08±1E-08</td>
<td>1.74±0.05</td>
<td>18±0.4</td>
<td>8</td>
<td>1.95±0.00</td>
<td>10</td>
<td>(n=2)</td>
<td></td>
</tr>
<tr>
<td>Sandy till - GLD1</td>
<td>0%</td>
<td>7E-08±1E-08</td>
<td>1.65±0.02</td>
<td>20±0.7</td>
<td>1</td>
<td>2.08±0.01</td>
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<td>(n=9)</td>
<td>0.7</td>
</tr>
<tr>
<td>Silty till - GLD2</td>
<td>0%</td>
<td>3E-10±1E-11</td>
<td>10±1.0</td>
<td>6</td>
<td>2.03±0.01</td>
<td>8</td>
<td>3.6±1</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>5%</td>
<td>2E-08±1E-09</td>
<td>1.99±0.03</td>
<td>11±1.0</td>
<td>2</td>
<td>2.0±0.00</td>
<td>9</td>
<td>(n=3)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>10%</td>
<td>1.92±0.02</td>
<td>14±0.3</td>
<td>3</td>
<td>1.9±0.02</td>
<td>11</td>
<td>(n=3)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>20%</td>
<td>4E-08±1E-08</td>
<td>1.64±0.04</td>
<td>21±0.4</td>
<td>6</td>
<td>1.9±0.02</td>
<td>12</td>
<td>(n=3)</td>
<td></td>
</tr>
<tr>
<td>Silty till 1 - GLD1</td>
<td>0%</td>
<td>3E-08±1E-09</td>
<td>1.86±0.02</td>
<td>20±0.7</td>
<td>1</td>
<td>2.08±0.01</td>
<td>8</td>
<td>(n=2)</td>
<td>4.3</td>
</tr>
<tr>
<td></td>
<td>5%</td>
<td>2E-08±1E-09</td>
<td>1.99±0.03</td>
<td>11±1.0</td>
<td>2</td>
<td>2.0±0.00</td>
<td>9</td>
<td>(n=2)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>10%</td>
<td>1.92±0.02</td>
<td>14±0.3</td>
<td>3</td>
<td>1.9±0.02</td>
<td>11</td>
<td>(n=3)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>20%</td>
<td>4E-08±1E-08</td>
<td>1.64±0.04</td>
<td>21±0.4</td>
<td>6</td>
<td>1.9±0.02</td>
<td>12</td>
<td>(n=3)</td>
<td></td>
</tr>
<tr>
<td>Silty till 1 - GLD2</td>
<td>0%</td>
<td>3E-08±1E-09</td>
<td>1.86±0.02</td>
<td>20±0.7</td>
<td>1</td>
<td>2.0±0.00</td>
<td>9</td>
<td>(n=1)</td>
<td>4.3</td>
</tr>
<tr>
<td></td>
<td>5%</td>
<td>2E-08±1E-09</td>
<td>1.99±0.03</td>
<td>11±1.0</td>
<td>2</td>
<td>2.0±0.00</td>
<td>9</td>
<td>(n=1)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>10%</td>
<td>1.92±0.02</td>
<td>14±0.3</td>
<td>3</td>
<td>1.9±0.02</td>
<td>11</td>
<td>(n=1)</td>
<td></td>
<td></td>
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<tr>
<td></td>
<td>20%</td>
<td>4E-08±1E-08</td>
<td>1.64±0.04</td>
<td>21±0.4</td>
<td>6</td>
<td>1.9±0.02</td>
<td>12</td>
<td>(n=1)</td>
<td></td>
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<tr>
<td>Silty till 1 - GLD3</td>
<td>0%</td>
<td>3E-08±1E-09</td>
<td>1.86±0.02</td>
<td>20±0.7</td>
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<td>2.0±0.00</td>
<td>9</td>
<td>(n=1)</td>
<td>4.3</td>
</tr>
<tr>
<td></td>
<td>5%</td>
<td>2E-08±1E-09</td>
<td>1.99±0.03</td>
<td>11±1.0</td>
<td>2</td>
<td>2.0±0.00</td>
<td>9</td>
<td>(n=1)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>10%</td>
<td>1.92±0.02</td>
<td>14±0.3</td>
<td>3</td>
<td>1.9±0.02</td>
<td>11</td>
<td>(n=1)</td>
<td></td>
<td></td>
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<tr>
<td></td>
<td>20%</td>
<td>4E-08±1E-08</td>
<td>1.64±0.04</td>
<td>21±0.4</td>
<td>6</td>
<td>1.9±0.02</td>
<td>12</td>
<td>(n=1)</td>
<td></td>
</tr>
<tr>
<td>Silty till 1 - LC</td>
<td>0%</td>
<td>3E-08±1E-09</td>
<td>1.86±0.02</td>
<td>20±0.7</td>
<td>1</td>
<td>2.0±0.00</td>
<td>9</td>
<td>(n=1)</td>
<td>4.3</td>
</tr>
<tr>
<td></td>
<td>5%</td>
<td>2E-08±1E-09</td>
<td>1.99±0.03</td>
<td>11±1.0</td>
<td>2</td>
<td>2.0±0.00</td>
<td>9</td>
<td>(n=1)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>10%</td>
<td>1.92±0.02</td>
<td>14±0.3</td>
<td>3</td>
<td>1.9±0.02</td>
<td>11</td>
<td>(n=1)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>20%</td>
<td>4E-08±1E-08</td>
<td>1.64±0.04</td>
<td>21±0.4</td>
<td>6</td>
<td>1.9±0.02</td>
<td>12</td>
<td>(n=1)</td>
<td></td>
</tr>
<tr>
<td>GLD1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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</table>
| GLD2 | 1E-08±1E-09 | (n=3) | * | (Mäkitalo et al. 2014).
Figure 11. Hydraulic conductivity (HC), dry density after compaction ($\rho_d$), and molding water content ($w$) in the different mixtures of till and GLD. Silty till 1 – GLD1 - drier till represents a till with a TSC of 96 % compared to 91 %. Silty till 1–GLD1-LC represents the same as Silty till 1-GLD1 except that it has been compacted with less compaction effort (lower weight and drop height). The transparent thick lines are $w$ 1-2 % wet of optimum for each material.
For WRC, it was expected that the high initial WRC of the GLD (Mäkitalo et al. 2014) would increase the WRC of the till with GLD addition. The SWCC for the different till-GLD mixtures studied here shows that WRC does improve with GLD addition (Figure 12; Table 2). At 85 % Sr, the matric suction ($\psi_{85}$) increased from 3.2 to 6 mH2O with 15 wt. % GLD addition to the silty till 1 (Figure 12:A-D; Table 2), 3.3 to 26 mH2O in the sandy till mixtures (Figure 12:E-H; Table 2) and from 3.6 to 50 mH2O in the silty till 2 mixtures (Figure 12:I-L; Table 2). The more GLD that is added the higher WRC is achieved in the mixtures. To minimize oxygen diffusion the sealing layer should be kept at a saturation degree of at least 85 % (Corey 1957, Aubertin and Mbonimpa 2001; Achib et al. 2004) and with 15 wt. % GLD addition to the tills a high saturation is kept even at high suction (Figure 12).

Table 2. Van Genuchten parameters, matric suction for 85 % Sr ($\psi_{85}$), the air entry suction ($\psi_a$), dry density ($\rho_d$), water content ($w$), compaction degree ($R_D$) and if the water content is dry or wet of optimal water content.

<table>
<thead>
<tr>
<th></th>
<th>$\psi_{85}$</th>
<th>$\theta_r$</th>
<th>$\theta_s$</th>
<th>n</th>
<th>m</th>
<th>$\alpha$</th>
<th>$\beta$</th>
<th>$\psi_a$</th>
<th>$\rho_d$</th>
<th>$w$</th>
<th>$R_D$</th>
<th>Dry/wet of opt. w</th>
</tr>
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<tr>
<td>Silty till 1</td>
<td>3.2</td>
<td>0.025</td>
<td>0.34</td>
<td>4</td>
<td>0.75</td>
<td>0.22</td>
<td>2.5</td>
<td>1.8</td>
<td>0.09</td>
<td>87%</td>
<td>w</td>
<td></td>
</tr>
<tr>
<td>Silty till 1 + 5 wt. % GLD</td>
<td>3.4</td>
<td>0.05</td>
<td>0.34</td>
<td>2.5</td>
<td>0.65</td>
<td>0.2</td>
<td>2</td>
<td>1.95</td>
<td>0.12</td>
<td>95%</td>
<td>w</td>
<td></td>
</tr>
<tr>
<td>Silty till 1 + 10 wt. % GLD</td>
<td>4.8</td>
<td>0.06</td>
<td>0.34</td>
<td>2.6</td>
<td>0.62</td>
<td>0.15</td>
<td>3</td>
<td>1.83</td>
<td>0.15</td>
<td>92%</td>
<td>w</td>
<td></td>
</tr>
<tr>
<td>Silty till 1 + 15 wt. % GLD</td>
<td>6</td>
<td>0.07</td>
<td>0.34</td>
<td>2.6</td>
<td>0.62</td>
<td>0.12</td>
<td>3</td>
<td>1.78</td>
<td>0.19</td>
<td>91%</td>
<td>w</td>
<td></td>
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<tr>
<td>Sandy till</td>
<td>3.3</td>
<td>0.04</td>
<td>0.34</td>
<td>3.6</td>
<td>0.72</td>
<td>0.27</td>
<td>3</td>
<td>1.77</td>
<td>0.07</td>
<td>87%</td>
<td>w</td>
<td></td>
</tr>
<tr>
<td>Sandy till + 5 wt. % GLD</td>
<td>1.2</td>
<td>0.045</td>
<td>0.34</td>
<td>1.8</td>
<td>0.44</td>
<td>0.7</td>
<td>0.7</td>
<td>1.83</td>
<td>0.12</td>
<td>91%</td>
<td>w</td>
<td></td>
</tr>
<tr>
<td>Sandy till + 10 wt. % GLD</td>
<td>3.3</td>
<td>0.02</td>
<td>0.34</td>
<td>1.35</td>
<td>0.26</td>
<td>0.3</td>
<td>1</td>
<td>1.81</td>
<td>0.16</td>
<td>91%</td>
<td>w</td>
<td></td>
</tr>
<tr>
<td>Sandy till + 15 wt. % GLD</td>
<td>26</td>
<td>0.03</td>
<td>0.34</td>
<td>1.15</td>
<td>0.13</td>
<td>0.1</td>
<td>4</td>
<td>1.77</td>
<td>0.19</td>
<td>92%</td>
<td>w</td>
<td></td>
</tr>
<tr>
<td>Silty till 2</td>
<td>3.6</td>
<td>0.09</td>
<td>0.34</td>
<td>1.5</td>
<td>0.33</td>
<td>0.9</td>
<td>2</td>
<td>1.92</td>
<td>0.07</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Silty till 2 + 5 wt. % GLD</td>
<td>11</td>
<td>0.01</td>
<td>0.34</td>
<td>1.28</td>
<td>0.22</td>
<td>0.1</td>
<td>4</td>
<td>1.89</td>
<td>0.13</td>
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</tr>
<tr>
<td>Silty till 2 + 10 wt. % GLD</td>
<td>22</td>
<td>0.00001</td>
<td>0.34</td>
<td>1.37</td>
<td>0.27</td>
<td>0.04</td>
<td>10</td>
<td>1.86</td>
<td>0.17</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Silty till 2 + 15 wt. % GLD</td>
<td>50</td>
<td>0.005</td>
<td>0.34</td>
<td>3</td>
<td>0.67</td>
<td>0.032</td>
<td>25</td>
<td>1.75</td>
<td>0.2</td>
<td></td>
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</table>
Figure 12. WRC in the mixtures of silty till 1-, sandy till- and silty till 2 and GLD. On the x-axis is the matric suction ($\psi$) in meter water column and on the y-axis the water saturation ($S_r$). Each mixture has been run 1-3 times and each run is marked with a different geometric figure in the graph. The red line is the van Genuchten parameter (best fit) and the black line represent 85 % water saturation. The van Genuchten line was set to start at 100 % $S_r$. 0 m suction is in the graphs 0.1 m suction due to the logarithmic scale.
3.2 How the content of fines and clay in till affects the HC and WRC of till-GLD mixtures

The PSD of the GLD showed that the majority was finer than 63 μm, but only 5.4±4.1 % and 9.5±1.6 % were in the clay fraction for GLD1 and GLD2 respectively (Figure 13 and Table 3). The silty till 1 consisted of 35 % fines of which 2.6 % was in the clay fraction. Silty till 2 had the same percentage of fines as the silty till-1, but a higher percentage of these were in the clay fraction, 4.3 %. The sandy till had a lower percentage of fines and clays (14 % fines and 0.7 % clays; Figure 13 and Table 3).

A decrease in HC is seen for the silty till 1 and sandy till with addition of GLD up to a wt. % of 5-10 (Figure 11:A). This was expected as the GLD increases the fine-grained material in the mixtures and results in decreasing HC (Benson et al. 1994; Benson and Trast 1995; Leroueil et al. 2002). The HC of the silty till 2 does not decrease at the addition of GLD as the tills initial clay content leaves no space for the GLD in its micropores. Therefore, the addition of GLD to
the silty till 2 is only deteriorating the compaction degree of the mixture due to an increase in water content, leading to a higher HC. The same mechanism increases the HC when more than 5 and 10 wt. % of GLD is added for the silty till 1 and the sandy till.

The till as a heterogenic material and a dominant part of the mixture, was expected to have a significant impact on especially the HC of the mixtures. The finer the till is, the lower the HC in the mixture was expected. The higher clay content in silty till 2 is likely the reason for the much lower HC compared to the silty till 1 (Figure 11:A). In a study conducted by Leroueil (2002) the HC decreased several orders of magnitude when the clay size fraction increased from 2 to 12 %). Also, a study conducted by Benson et al. (1994) showed a strong relationship between clay content and HC and a weak relationship between fines and HC when studying clay.

For WRC, an increase in the retention capacity, i.e. an increase in $\psi_a$ and a decrease in $n$ was expected, with higher amounts of GLD added to the tills. This as materials with smaller pores generally has a higher $\psi_a$ (Fredlund and Rahardjo 1993; Tinjum et al. 1997; Sillers et al. 2001) and a flatter slope of the SWCC in the desaturation zone (Sillers et al. 2001). A similar trend is seen in these experiments, i.e. a higher $\psi_a$ and a flatter slope with increasing GLD addition (Table 2; Figure 12). However, comparing the pure tills, the silty till 2 should have the highest $\psi_a$, this is not the case. The tills have similar $\psi_a$, 2-3 (Table 2). The dry density is another factor that has shown to have a significant impact on the SWCC due to a decrease in the number of voids with increasing dry density, leading to faster saturation (Vanapalli et al. 1999). However, the differences in dry density between mixtures is limited (Table 2) and cannot explain the lack of a higher $\psi_{at}$ of the silty till 2, compared to the silty till 1 and the sandy till (Table 2). Neither can it explain the higher $\psi_{at}$ in the sandy till + 15 wt. % GLD compared to the silty till 1 + 15 wt. % GLD. However, the $\psi_{at}$ is not the only value explaining the SWCC. The behavior of the material at higher pressures is another important feature. Silty till 2 keeps a higher saturation at higher suction (Figure 12:I-L). This might be due to that this till is highly aggregated and the inner forces between soil aggregates have been shown to be very strong in resisting de-saturation behavior at higher suction values (Vanapalli et al. 1999). Mineralogy is another factor that is known to affect the SWCC of a soil (Mitchell 1993; Tinjum et al. 1997) and needs further investigation.

### 3.3 How the initial water content of the materials affects the HC in the till-GLD mixtures

A lower initial water-content of the till was expected to improve the HC when higher amounts of GLD was added, as the final molding water-content of the mixture would be closer to the optimal water-content. The hypothesis is confirmed in this study. The mixture with the wetter till (Silty till – GLD1) decreases from 3 to 2E-08 m/s with an addition of 5 wt. % of GLD, to then steadily increase with the further addition of GLD (from 2 to 7E-08 m/s; Figure 11:A and Table 1). In the drier till mixture (Silty till 1–GLD1–drier till) the initial HC was higher, 10E-08 compared to 3E-08 m/s (Figure 11:B). The HC then decreases steadily with increasing addition of GLD (from 10 to 3E-08 m/s) until 20 wt. % of GLD addition, where a slight increase in HC is seen (from 3 to 4E-08 m/s; Figure 11:B). When the GLD addition reaches over 10 wt. % in the drier mixture the HC becomes lower than in the moister mixture, 3E-08 compared to
The decrease in HC in the drier till-mixture can be explained by the correlation between HC and the degree of compaction (Benson et al. 1994; Watabe et al. 2000; Leroueil et al. 2002). Compacting a drier till results in higher dry densities in mixtures with 5-15 wt. % GLD addition, approximately 0.1 to 0.2 g/cm³ higher compared to the moist till (Figure 11:B). However, the HC in the drier mixture only reaches lower values than in the wetter mixture with 15-20 wt. % GLD addition. Adding 15 wt. % GLD to the moist till increases the water-content far beyond the optimum water-content (Figure 11:E), leading to a lower dry density and a higher HC than in a mixture with a drier till. Benson and Trast (1995) showed that the HC is sensitive to the molding water-content, where the lowest HC was reached at molding water-content of 1-2 % wet of the optimum water content. Around 5 wt. % of GLD1 seems to be a threshold for tills with a TSC of approximately 91 % and with an increasing amount of GLD above that the water-content increases beyond the 1-2 % wet of optimum. In a drier till, a higher amount of GLD-addition is needed to reach the optimum HC compared to a wetter till (Figure 11:A and B). At 15 wt. % of GLD1 addition the 1-2 % wet of optimum water level is reached and here is where the lowest HC is seen in the drier till mixture.

An addition of a drier GLD was expected to decrease the HC of the mixture. In this study, the HC stays around 3E-08 m/s with an addition of up to 15 wt. % of the drier and more fine-grained GLD2 and does not follow the same u-shaped trend as when using GLD1 (Figure 11:A and B; Table 1). As for the mixture with the drier till there seems to be a higher tolerance of how much drier GLD can be added to the silty till compared to when adding a more wet GLD. This is likely due to the optimal water content, that for 10-15 wt. % addition of GLD1 reaches 5-8 % wet of optimum water-content after compaction, for GLD2 the corresponding number is 4 % (Figure 11:E and F; Table 1). Another explanation to that a higher amount of GLD2 can be added to the mixture before an increase in HC occurs may be that GLD2 is more fine-grained than GLD1. Even if the dry density decreases with more addition of GLD, the increase of fine-grained material levels out the expected increase in HC.

### 3.4 Challenges with the methods and the materials

**3.4.1 Hydraulic conductivity**

There are several parameters in the laboratory conditions that can affect the measured HC and complicates the comparison between different studies. One key issue is the **compaction method** used to prepare the samples to be tested. The degree of compaction, expressed as dry density is known to affect the HC in porous media. Hand compaction is a non-standardized compaction procedure creating differences between samples, and therefore the dry density and HC may also be different. Thus, a PC method was preferred as it uses the same effort between samples. One downside of the PC method is that the compaction hammer does not reach the sides of the sample cylinder. Therefore, the millimeters closest to the walls are not compacted and there is a risk of preferred wall-flow when measuring HC. Indications of this were seen in the cylinders after HC-test, as the samples appeared wetter at the edges. Using a hand-driven compaction hammer attached to a frame enables a standardized compaction effort between the samples and the hammer can reach the sides of the cylinder. The disadvantage with this method is that it is a challenge to make sure that the whole sample is compacted in the same degree when comparing to the PC method where the hammer moves around the sample in a standardized fashion.
The compaction effort has previously shown to have an impact on HC. A study conducted on thirteen compacted clays show a decrease in HC with an increase in compaction effort (Benson and Trast 1995). However, studies by Mäkitalo et al. (2015b) indicate that mixing time and mixing effort releases the water bound in the GLD leading to an increase in porosity of the till-GLD mixtures. The authors suggested that standard PC might not be the best choice when working with GLD, as the effort and contact with the GLD are quite high with a standard PC method. The hypothesis was that a lower compaction effort might decrease the HC of the till-GLD mixtures. However, the results from this study do not corroborate this argumentation, as a decreased compaction effort does not significantly affect the HC, neither positively or negatively. A difference in the mixtures for which light compaction was used compared to standard PC is only seen with an addition of 5 wt. % of GLD1, where the HC is higher when using a light PC than when using standard PC (4E-08 compared to 2E-08 m/s; Figure 11: A and B). This can likely be explained by the decrease in compaction effort that, as explained before, has been shown to increase HC (Benson and Trast 1995). However, a water release leading to a liquefaction of the GLD that Mäkitalo (2015b) found when increasing the mixing effort can also be seen in this study. Even if it does not seem to affect the HC negatively. Despite the lack of positive effects on HC when using a lighter compaction effort, it might still be advisable, as laboratory tests tend to underpredict HC compared to actual values in the field. There is evidence of an underprediction of HC by a factor of 10 and sometimes more (Daniel 1984). When comparing HC in the lab and in the field, the compaction effort in the field is usually less than in the lab, leading to an increase in optimum water content (Daniel 1984). Therefore, when compacting in the field, the compaction is made dry of optimum water content instead of at the optimum, resulting in a larger HC than expected. This can be avoided by compacting at the same compaction effort that will be used in the field when compacting in the lab (Daniel 1984). Other reasons for underestimating of field HC are desiccation cracks, difficulties getting a representative sample of soil for laboratory test and poor construction (Daniel 1984). The unsaturated properties with a soil in the field is another challenge. An unsaturated soil can vary 10 orders of magnitude in HC, making it difficult to analyze and interpret the results (Fredlund et al. 1994) as the HC in the laboratory is often measured in a saturated soil.

Another parameter in the HC-method that can affect the outcome is the hydraulic gradient. In geotechnical engineering, the common practice is to apply high hydraulic gradients for quick determination of the HC, which can cause an increase in HC as an increased flow rate at high hydraulic gradients widens the void space by erosion (Al-Taie et al. 2014). High hydraulic gradients can also cause reduction of the HC if the pressure is high enough to erode aggregates of colloidal size from the flow channels (Pusch and Weston 2003; Al-Taie et al. 2014). However, in this study, the hydraulic gradient of 8.7 is close to natural conditions and much lower than 100, that by Al-Taie et al. (2014) was suggested as a limit, or 30 which by ASTM is the recommended maximum hydraulic gradient.

3.4.2 Water retention capacity

For many mixtures the calculated saturation is greater than 100 % and for these cases, the best-fit van Genuchten curve was set to start at 100 % saturation (Figure 12), as a saturation above 100 % is not possible. Calculated saturation values above 100 % might be explained by the GLD’s tendencies to shrink when drying. Volumetric shrinkage is due to the rearrangement of particles
and aggregates during drying causing a decrease in bulk volume (Gapak et al. 2017). The shrinkage has an impact on the SWCC as the total volume that is used in the saturation calculations is not constant. If the material has tendencies towards volumetric shrinkage behavior when dried, they would first swell of the water addition when saturated, leading to an increase in volume and Sr. However, also the pure tills with no GLD addition have Sr higher than 100 %, so the shrinkage behavior might not be the only reason. An incorrect measurement of the ring-volumes is not a likely explanation, as the Sr never reaches below 100 % at the first pressure step. Therefore, another explanation that could lead to increased Sr is a loss of material when the samples are taken out to be weighed. Some material was left underneath the ceramic plates that are placed on top of the samples. This even though “separators” higher than the rings were placed between the plates. However, this is not a likely explanation for the oversaturated samples as they were placed on the top ceramic plate. Also, two ways of calculating Sr was used, one with the water content calculated from the material before compaction and one with the water content in the samples that was oven dried after the pressure steps. Both values show oversaturated samples in the first pressure step. The common parameters between the two ways of calculating Sr is the volume of the samples and the particle density of the materials used. An error in the estimation of the particle density is not likely the only explanation as it, to decrease the Sr to 100 %, needs to be increased to a value that is not likely for a till (3 g/cm³). Volumetric shrinkage is therefore the likely explanation to the overestimated calculated saturation of the materials. The presence of material on the bottom of the ceramic plates might be evidence for that volumetric shrinkage is in fact occurring and a volumetric shrinkage curve (VSC) should be obtained and the results from that included in the SWCC (Fredlund 2002; Wijaya et al. 2015). The VSC present the relationship between the volume of the specimen and the corresponding water content (Fredlund 2002; Wijaya et al. 2015).

The hysteretic effect is a parameter that will impact the interpretation of the SWCC and the actual WRC of the mixtures studied. The hysteretic effect will give higher matric suction for drying than for wetting for the same water content (Tinjum et al. 1997), and the $\psi_{\text{sat}}$ would be higher. This lead to an underestimation of the WRC in laboratory experiments compared to actual values in the field.

3.4.3 Variability of the GLD

A challenge when working with GLD is the heterogeneity in both physical and chemical properties between different paper mills, but also with time for the same paper mill. The differences in the material are visible by observation, see pictures in Figure 5. To illustrate this variability, the results from the characterization of GLD from 15 Swedish papermills, producing 1 200 to 35 000 ton per year, are presented in Table 4 and 5, where also the two GLD used in this study are shown as comparison. The TSC of the GLD differs between 33 and 75 %, clay content between 3 and 28 %, plasticity index (wP) between 46 and 170 %, the yield point (wL) between 60 and 330 % and the HC between 1E-07 and 4E-09 m/s (Table 4). All these properties are factors controlling the compaction properties, HC, WRC and finally also the oxygen deterring function of a sealing layer.
There is also a great chemical variation between the different GLD, up to approximately 2500 order of magnitude difference (Zn; Table 5). Other elements that differ significantly between the different GLD are Cd, Co, Cu and W (Table 5). The chemistry of the material creates the mineralogy which in turn affects the HC, WRC and oxygen diffusion.

The chemical and physical variation within a GLD from the same papermill is also extensive, which is demonstrated in a study by Mäkitalo et al. (2014). The great variability creates challenges when GLD is to be used in a sealing layer. There is no guarantee that the properties of the GLD in the planning phase of a project is the same as when the GLD is delivered to the mine site, even though a characterization of the materials has been made.

Table 4. Physical characterization of GLD from 15 different paper mills. TSC = total solids content, PSD = particle size distribution, \( \rho_s \) = particle density, \( w_P \) = plasticity index, \( w_L \) = wilting point and HC = hydraulic conductivity.
Apart from the variation in GLD the variation in physical properties in till within the same till quarry is also a factor that needs consideration. For natural reasons, the PSD and TSC vary in the same till. Till is created by the ice from the latest ice age eroding bedrock. The PSD in a till depends on the way it was formed and on the kind of bedrock it originates from. In Sweden the till below the highest coast line have been affected by the sea due to the uplift of the crust after the latest ice age. The waves have then sorted out the finest material of the tills.

The PSD in till from a quarry at Näsliden mine in Sweden was investigated and show contents of fines (<0.063 mm) between 24 and 35 % (Figure 14) and TSC of 90-94 %. These two factors have in this study shown to affect the final HC and this variation makes an estimation of field HC in a sealing layer where the till is to be used challenging.
Figure 14. Variation in PSD (<0.063 mm) in till from the same till quarry.
4 Conclusions

The hypothesis formulated when starting this study was that the addition of GLD would improve the HC of the tills that alone would not reach the required $10^{-8}$ m/s. The HC of the mixtures based on most of the tills did improve (decrease), but not enough to reach the required HC.

However, even if the study did not result in low enough HC in most of the cases, the WRC of the GLD-till mixtures show promising results, especially at 15 wt. % addition and can be seen as the most important parameter minimizing the oxygen diffusion through a sealing layer, by keeping the layer saturated.

This study indicates that the initial HC of the till that is used in the mixtures plays a major role in controlling the HC of the final mixture. The HC of the till is in turn affected by the content of fines and especially clay in the till.

The study also show that the initial water content of the materials has a greater impact on the HC of the mixtures compared to dry density after compaction. It determines the degree wet of optimum that in this and previous studies has shown to greatly affect the HC, 1-2 % wet of optimum water content generates the lowest HC. If the till is dry, a higher amount of GLD is required to reach optimum HC. If the GLD is drier, the HC is not affected by the amount of GLD added.

The greatest challenge when working with GLD is its heterogeneity, which makes it difficult to predict how it will behave in a sealing layer. Even though the recycling of industrial residues is something to strive for and an important step towards a circular economy. The use of GLD in mine waste remediation is beneficial for both the industry that provides the residue and the mining company. It is also a great profit for the environment as it minimizes the waste produced.
Future research

A better understanding of the mineralogical composition of the fine particle fraction of both till and GLD used in the mixture is necessary to be able to predict how the sealing layer will age. The oxygen diffusion is controlled by the degree of saturation of the sealing layer, which in turn is controlled by the amount and the properties of the finest particle fraction of the mixture. The durability of the cover is therefore dependant of this fraction and its ability to decrease the porosity, adsorb water or dissolve.

Future laboratory studies should focus on connecting the HC and WRC of the materials to oxygen diffusion. As it can be difficult to reliably monitor oxygen in the field, there is a need to study how the different parameters studied here are connected to the oxygen diffusion.

Ongoing work related to this study is the evaluation of a cover system constructed on top of the waste rock dump at the closed Näsliden mine in northern Sweden (Figure 15). The sealing layer here consists of a bentonite-till mixture, but a small test surface at the site consists of a till-GLD mixture. In both of these surfaces instruments like lysimeters, temperature-, oxygen-, soil moisture-, and pore pressure probes have been installed. Data is being collected and will be analyzed to evaluate the effectiveness of the sealing layer.

Prior to the covering of the Näsliden mine, a pilot cell in Boden, Sweden was constructed to evaluate the methods of mixing and compacting together with the use of GLD from different paper mills. The results from these tests are presented in a paper by Mäkitalo et al (2015a). At the pilot cell, lysimeters, temperature-, soil moisture and pore pressure probes were installed (Figure 16) and data from these instruments are collected and are analyzed to evaluate the effectiveness of the sealing layer and will be presented in a future study.
Figure 15. Instrumentation of the covered waste rock dump at the closed Näsliden mine in northern Sweden.
Figure 16. Pictures and overview of the pilot cell in Boden, Sweden, made of 10 wt. % GLD and a silty till.
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Opportunities to use Green Liquor Dregs as an amendment in till to cover sulfidic mine waste

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Opportunities to use Green Liquor Dregs as an amendment in till to cover sulfidic mine waste

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Abstract

The oxidation of sulfide minerals in mine wastes is a possible threat to the environment as it might have potential to generate acid leachate and release metals. A common method to reduce sulfide oxidation is to apply a dry cover on the mine waste deposit. A typical dry cover in Sweden consists of a compacted sealing layer of a clayey till with a protection layer on top. The sealing layer mitigates oxidation of metals by deterring oxygen to enter the waste. Due to a lack of suitable natural materials in the vicinity of the mine and an increasing amount of mine waste produced, there is a drive to use alternative solutions in the mine remediation program. An inert industrial residue would be a suitable alternative and serves as a large benefit for the environment, but also for the mining industry and the industry providing the residue, decreasing their environmental footprint. In this study, tills with different contents of fines and clays were mixed with 5-20 wt. % of Green Liquor Dregs (GLD), a residue from the pulp production. Hydraulic conductivity (HC) and water retention capacity (WRC) were determined for each mixture with the objective to investigate if GLD can improve the HC and WRC of a local till that is to be used in a sealing layer on top of sulfidic mine waste. Another objective was to find out how the HC and WRC are affected by the contents of fines and clays in the tills. This study concludes that the HC and the WRC of tills that does not meet the requirements of a sealing layer, improves with GLD addition. However, the improvement in HC was not enough to reach the requirements. Even though, a high WRC can be seen as the most important factor in deterring ARD by keeping the sealing layer close to saturation. The study shows that the till-GLD mixtures can keep a high saturation even with high matric suction, especially with 15 wt. % GLD addition. Recycling of GLD in mine remediation is something to strive for as it is beneficial for both the mining company and the pulp and paper industry and a great profit for the community as it minimizes the waste produced.

Keywords: Green Liquor Dregs; Industrial residue; Sealing layer; Mine reclamation; Till; Hydraulic conductivity; Water retention capacity
1 Introduction

Massive amounts of mine wastes are generated all over the world. Sweden alone generated 139-million-ton mine waste in 2014, accounting for 83 % of all waste produced during that year (Swedish EPA 2016). These wastes need to be managed in an environmentally-, technically- and economically sustainable way. About 70 % of the mine waste in Sweden contains sulfide minerals (SGU and Swedish EPA 2017). If sulfidic mine waste is left in contact with oxygen and humidity the sulfides oxidize and have the potential to produce acid rock drainage (ARD). ARD is a major long-term threat to the environment as metals and metalloids may become mobile (Saria et al. 2006). The GARD guide (Global Acid Rock Drainage guide; Verburg et al. 2009) categorize different methods to prevent ARD after closure, into two main categories: engineered barriers and water covers. Engineered barriers can be divided into liners and dry covers, where liners are typically designed to act as a barrier for contaminant flow from the overlying waste into the receiving environment. Dry covers are typically designed to limit the ingress of water and oxygen into the underlying waste. Under the relatively humid climatic conditions in Sweden a soil cover can be used to reduce oxygen flux to the underlying reactive wastes, and thus reduce ARD (Collin and Rasmuson 1990; Bussière et al. 2003; Dagenais et al. 2006). A soil cover in Sweden usually consists of a sealing layer placed on top of the mine waste and above this, a protective layer. The sealing layer is made of a fine grained compacted material to prevent oxygen to diffuse to the waste underneath by keeping it close to saturation. The purpose of the protection layer is to protect the sealing layer from erosion, frost- and/or root penetration. The main function of the sealing layer is usually to limit oxygen diffusion through the cover and oxygen has two main ways of transport in a soil, through the water or the air in the pores. Instead of oxygen diffusion, the hydraulic conductivity (HC) is usually measured to evaluate the effectiveness of a sealing layer as it is easier to measure. The requirements for HC in a sealing is site specific, but commonly the requirement in Sweden is <10⁻⁸ m/s.

The presence of fine grained material is an important factor that influences the HC. An increasing amount of fines in the material decreases the HC (Benson et al. 1994; Benson and Trast 1995; Leroueil et al. 2002), as the micro-porosity of the material decreases. For materials that is to be used as an oxygen barrier on top of mine waste, their water retention capacity (WRC) is another important feature, as a high WRC usually corresponds to a high water saturation (S_τ). The transport of gaseous oxygen through fine-grained materials is mainly by molecular diffusion (Yanful 1993). At a relatively low degree of saturation, most oxygen transport occurs through the partially air-filled pores (Aachib et al. 2004), due to the 10 000 times higher diffusion coefficient (D_τ) in the air than in water (Yanful 1993). In addition to a higher diffusion coefficient, the dissolved O₂ concentrations in the air is around 20 000 times higher than in water (Höglund et al. 2004; Verburg et al. 2009). In general, the oxygen flux rates are at a minimum when the degree of saturation is greater than 85-90 %, this as the air-phase at a saturation greater than 85 % becomes discontinuous (Corey 1957). The oxygen is then transported through the water phase (Aubertin and Mbonimpa 2001; Aachib et al. 2004) and in a layer that is kept close to saturation, the D_τ in the soil can be comparable to the D_τ in water. It is then small enough to reduce the oxygen flux to a level comparable to that of a water cover (Yanful 1993; Aachib et al. 2004). One way to estimate the WRC is to use a soil-water characteristic curve (SWCC), which demonstrates the correlation between matric suction (ψ) and water content (gravimetric [w] or volumetric [θ]) or degree of saturation [Sr]; Figure 1:A). The typical SWCC consist of three stages: capillary saturation, desaturation and residual saturation (Sillers et al. 2001). The matric suction required to remove water from the largest pores corresponds to the break in the SWCC, where the soil is nearly saturated (θ_s; Figure 1:A). This brake in the curve is generally referred to as the air-entry suction (ψ_a; Figure 1:A). The water content corresponding to the asymptote of the SWCC at low degrees of saturation is called the residual water content (θ_r; Figure 1:A). The SWCC is hysteretic, which means that for a given water content, higher matric suction exists for desorption (drying) than for sorption (wetting; Figure 1:B; Tinjum et al. 1997). This so called hysteretic effect is caused by size differences between the bulk-pores and the interface-pores that connects the bulk-pores, which changes the contact angle between water and air during wetting (Tinjum et al. 1997). Due to experimentally practical reasons usually only the desorption curve is used in laboratory studies (Tinjum et al. 1997). The shape of the SWCC is related to the soil type. Soils with smaller pores and...
finer particles have a higher $\psi_a$ (1:C; Fredlund and Rahardjo 1993; Tinjum et al. 1997; Sillers et al. 2001). Also, the slope of the curve in the desaturation zone tends to become flatter as the soil particles become finer (Figure 1:C; Sillers et al. 2001). Except from particle size distribution also the molding water content is known to affect the SWCC of fine grained soils, with a higher $\psi_a$ and a steeper slope for suctions greater than the $\psi_a$ as the molding water content increases (Vanapalli et al. 1996; Tinjum et al. 1997; Vanapalli et al. 1999). A fine-grained soil compacted dry of optimum water content generates steeper SWCC compared to those compacted at or wet of optimum water content (Vanapalli et al. 1999). Dry of optimum the specimen is influenced by its macrostructure and acts like a coarse-grained soil due to aggregation. When compacted wet of optimum the microstructure is more important, leading to a higher $\psi_a$ than in soil compacted dry of optimum (Vanapalli et al. 1999). In addition to molding water content the initial density has a significant impact on the SWCC due to a decrease in the number of voids with increasing dry density, leading to faster saturation (Vanapalli et al. 1999).

![Figure 1. A) A typical SWCC, where $\psi_a$ is the air-entry suction, $\psi_r$ the water entry value, $\theta_s$ the near saturated water content and $\theta_r$ the residual water content (reworked from Tinjum et al. 1997). B) A typical SWCC for desorption (drying) and sorption (wetting; reworked from Zhou et al. 2012). C) A typical SWCC for clay, silt and sand (reworked from Sillers et al. 2001).](image)

Because of the Swedish geology most sealing layers are usually made of a till, ideally a clayey till. The availability of clayey till nearby a mine is often limited and the till can either be improved or replaced to construct a sealing layer. Bentonite amendments to till is one solution to improve the sealing layer-qualities of the local till. However, bentonite is costly both economically and environmentally due to time- and resource consuming production. Another suitable amendment to the till could be an industrial residue. Sweden produces 28 million tons wastes, excluding mine waste, of which about 26 million tons is classified as a non-hazardous waste by the Swedish Environmental protection Agency (EPA). Thereof 6 million tons are residues from industrial production (Swedish EPA 2016). Using an industrial residue in a mine remediation program would serve as a large benefit for the environment, and for the industry providing the residue together with the mining industry. Some examples of industrial residues that previously have been used as a mine waste cover are sewage sludge, fly ash, desulfurized tailings, coal combustion by-products (CCB), and steel slag (Hallberg et al. 2005; Bäckström et al. 2009; Dobchuk et al. 2013; Lu et al. 2013; Nason et al. 2013; Park et al. 2014; de Almeida et al. 2015; Phanikumar and Shankar 2016). Previous studies have shown that a residue of pulp production, Green Liquor Dregs (GLD), has properties suitable as sealing layer i.e. it is fine grained (d100 < 63μm), commonly has a HC in the range of 10^{-8} and 10^{-9} m/s and a higher water retention capacity (WRC) compared to materials with similar particle size, such as clayey/sandy silt (Mäkitalo et al. 2014). In 2003, about 200 000 tons of GLD were produced annually in Sweden (SGI 2003), and production has increased based on a survey made 2017 (unpublished data). GLD is an alkaline, inorganic waste originating from the recycling process at sulfate pulp and paper mills. The main solid compounds of GLD consists of CaCO₃, Mg(OH)₂, C and metal sulphides, especially FeS (Sanchez and Tran 2005; Jia et al. 2014). The liquid phase of the GLD consists of Na₂CO₃ and NaOH,
which generates its characteristic high pH. GLD is regarded as an inert material (Mäkelä and Höynälä 2000) and has the same grain size distribution as silt (Mäkitalo et al. 2014). Other characteristics of GLD are a high pH (10-11), relatively high porosity (73 - 82 %), a compact density of 2.47 to 2.60 g/cm³, a low HC (10⁻⁸ and 10⁻⁹ m/s) and a high WRC (Mäkitalo et al. 2014).

GLD has been used in a few sanitary landfill cover applications (Hargelius 2008; Pousette and Mácsik 2000). It has the possibility to act as a neutralizing agent for acidic wastewater (Pöykiö et al. 2006) and for sulfidic mine waste (Bäckström et al. 2010; Bellaloui et al. 1999; Lu et al. 2013), as the increased pH enhances the metal removal (Pérez-López et al. 2011). The possibilities to use GLD as a cover material on top of mine waste have been investigated in several pilot scale studies (Chtaini et al. 2001; Lu et al. 2013; Ragnvaldsson et al. 2014). The studies show a reduced effluent volume by 40 % (Ragnvaldsson et al. 2014), a reduction of leaching metals (Chtaini et al. 2001; Lu et al. 2013; Ragnvaldsson et al. 2014) and reduced oxygen diffusion to the mine wastes (Chtaini et al. 2001). Sequential extraction performed on GLD indicates relatively low bioavailability of metals (Nurmesniemi et al. 2005).

The long-term stability of the GLD has been studied by Mäkitalo et al. (2016) and suggested that the GLD did not show any physical or mineralogical changes in the study that will affect its ability to prevent oxygen diffusion to the underlying mine waste. The water retention capacity (WRC) observed to be >85 % under field conditions and maintained a high degree of saturation with low HC independently of its age (Mäkitalo et al. 2016). The shear strength of the GLD increased over time, but not enough to ensure a long-term physical stability in slopes (Mäkitalo et al. 2016).

Considering its lack of long-term physical stability, it is not reasonable to solely use GLD in the sealing layer from a geotechnical point of view. In this study GLD is used as an amendment in till with different PSD. Previous field studies on mixing GLD with till have shown promising results for the mixtures to be used as a sealing layer, with low HC, high WRC, and increased compaction properties (Hargelius 2008; Mäkitalo et al. 2015a and b). The properties of the till and GLD are however not homogeneous. The contents of fines and clays are one example of factors that varies in the materials and has a great potential to affect the final HC of the mixture. There is, therefore, a great need for further studies on how these factors control the final HC of the mixture and at which percentages the materials should be mixed with each other. In this study 5 to 20 wt. % of GLD from Smurfit Kappa paper mill were mixed with three sieved tills (<20 mm) with different contents of fines and clays. A maximum 20 % of addition of GLD was set as a limit as a mixture with more GLD becomes very difficult to compact and handle, due to increased water content over the optimum molding water content and decreased shear strength. The addition of GLD to tills, which alone does not reach a low enough HC, is expected to decrease its HC below 10⁻⁸ m/s to make it suitable for a sealing layer. The GLD consist of fine material which will fill the pores of the till, leading to a decreased HC. However, after a certain limit of GLD addition (10-15 wt. %) the high water-content and low shear strength of the GLD is expected to decrease the compaction degree and therefore increase the HC in the mixtures. The till as a heterogenic material and a dominant part of the mixture, is expected to have a significant impact on the HC and WRC of the mixtures. The finer the till is, the lower HC and higher WRC in the mixture is expected. The WRC of the mixtures is expected to increase with increasing amount of GLD added and the GLD addition is expected to have a greater influence on the WRC of the mixtures than the PSD of the tills. This due to the high initial WRC of the GLD. The objectives of the study were to find out (i) if GLD can improve the HC and WRC of till so it can be used in a sealing layer on top of sulfidic mine waste, and (ii) how the contents of fines and clays in three different tills will affect the HC and WRC in till-GLD-mixtures.

2 Materials and methods

2.1 Materials

Three different tills were used in this study, a sandy till and two silty tills with different clay contents. The silty till with a lower clay content is named silty till-1 and the silty till with higher clay content silty till-2. Silty till-1 was collected at a till quarry at the Brännkläppen facility in Boden (northern
Sweden), silty till-2 from another till quarry outside Boden and the sandy till from Sunderbyn outside Luleå (northern Sweden). The GLD used in the study derived from Smurfit Kappa paper mill in Piteå (northern Sweden) and was collected in sealed plastic containers to preserve the water content of the material.

2.2 Mixing of till and GLD

The tills were air dried and sieved through a 20-mm sieve and particles above 20 mm were removed. The sieved tills were then mixed with 5, 10, 15 and 20 wt. % of GLD. The weight percentage was calculated towards a dry till and a naturally moist GLD. The GLD was kept naturally moist as it is difficult to rewet which affects its physical properties. The mixing was carried out by hand with a small shovel until the mixture was estimated as homogenized.

2.3 Particle size distribution

The tills were washed and dry sieved according to SS-EN 933-1:2012 to obtain the weight percentage of fines. A mechanical sieve tower (Retsch AS 200) with an amplitude of 2.2 mm/g was used. The cut-off sizes were 12.5, 10, 8, 5, 4, 2, 1, 0.5, 0.25, 0.125 and 0.063 mm.

PSD for the fines (<0.063 mm) in the tills and GLD were done by laser diffraction analysis on triplicate samples of each material by a CILAS Granulometer 1064 (CILAS, Orléans, France). With a compact cast iron optical bench, the CILAS 1064 integrates 2 sequenced laser sources positioned at 0° and 45°, to produce a diffraction pattern analyzed on a 64-channel silicon detector. The PSD is then calculated using the CILAS software.

2.4 Hydraulic conductivity

Prior to HC-analysis, water was added to the tills until they reached a TSC (total solid content) of 91% (TSC 91±1%, n=24). 5, 10, 15 and 20 wt. % of GLD was den mixed in with the till. The constant head-method was used in air tight cylinders with a volume of 996 cm³. The walls of the cylinders were coated with a thin layer of bentonite to prevent the preferential wall flow. The mixtures inside the cylinder were compacted with standard proctor compaction (PC) method in five equally thick layers with a falling weight of 4.54 kg, falling 45 cm 25 times on each layer.

Water was lead to the bottom of the cylinder with a hydraulic gradient of 8.7 for silty till 1 and the sandy till. The hydraulic gradient for the silty till 2 was 13. The water passing through the cylinders were collected in plastic bottles, 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. The HC was calculated using Darcy’s law.

2.5 Water retention capacity

WRC was measured with a pressure plate test using a 1500F1 Pressure Plate Extractor. The objective of the test was to apply a succession of increasing gas pressure steps to samples enclosed in a tight cell, and to measure the variation of its saturation. 12 samples were placed on saturated ceramic plates with a high air entry value, which base is connected to the atmosphere. Gas pressure in 12 increasing steps were applied to the samples inside the cell and one sample was taken out for each pressure step and characterized for its water content. The pressure gradient applied was the same as if suction was applied to it in its normal environment. The ceramic plates were saturated for at least 48 h. The air-dried till used in the mixtures to be tested was wetted to reach a TSC of approximately 91 %. The plastic rings that were to contain the samples were prepared with taping Whatman filter paper of grade 41 to the bottom, measuring the volume and weighing them with the filter paper on. The materials to be tested were then compacted by hand with a metallic compactor with the weight of 2 kg. The compaction was made with 25 “hits” in five layers with the goal to reach an even compaction between the samples. When compaction was done the samples were levelled off with a knife and weighed. The TSC of the remaining material was determined using standard SS-EN 14346:2007. The results from this could then be used to estimate the void ratio of the sample according to formula (1):

$$e = \frac{V_v}{V_s} = \frac{(\rho_s \times V_t) - m_t - 1}{(\rho_s \times V_t)}\\= \frac{(\rho_s \times V_t) - m_t}{((\rho_r + \rho_s) + 1) + m_t}$$

(1)
Where $e =$ void ratio [-], $\rho_s =$ relative density of the grains [g/cm$^3$], $m_d =$ mass of dry soil [g], $m_r =$ mass of the ring, $m_w =$ mass of the wet sample and the ring [g], $w =$ gravimetric water content [-], $V_v =$ void volume [cm$^3$], $V_s =$ soil volume [cm$^3$] and $V_t =$ total volume [cm$^3$].

The saturated ceramic plates were then placed in the pressure cell and connected to the outflow tube. The rings with the samples were placed on the ceramic plates and saturated by adding water on the top of the ceramic and the cell was closed for at least 48 h. After saturation, water in excess was removed and the first pressure step of atmospheric pressure was applied. The system was then left to run for 48 h and after that one sample was removed from the cell and weighed. Its gravimetric water content was estimated by removing the soil sample from the ring, weighed, oven dried in 105˚C for 24 h and the weighed again. Thereafter the cell was closed, and the next pressure step applied. The procedure continued in the same manner for each pressure step that is presented in Table 1.

Table 1. Pressure steps used for the pressure plate test.

<table>
<thead>
<tr>
<th>Pressure step</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>8</th>
<th>9</th>
<th>10</th>
<th>11</th>
<th>12</th>
</tr>
</thead>
<tbody>
<tr>
<td>m H$_2$O</td>
<td>0</td>
<td>1.5</td>
<td>3</td>
<td>6</td>
<td>9</td>
<td>12</td>
<td>17</td>
<td>25</td>
<td>40</td>
<td>60</td>
<td>80</td>
<td>145</td>
</tr>
</tbody>
</table>

After the test was completed the porosity ($n$), volumetric water content ($\theta_w$) and degree of saturation ($S_r$) was calculated using formula 2-4.

$$n = \frac{V_v}{V_t} = \frac{e}{1+e}$$  \hspace{1cm} (2)

$$\theta_w = \frac{V_w}{V_t}$$  \hspace{1cm} (3)

$$S_r = \frac{\theta_w}{n}$$  \hspace{1cm} (4)

where $V_w =$ volume of water [cm$^3$].

The measured values from the experiment were plotted together best fit curve from Van Genuchten equation (Formula 5; Van Genuchten 1980).

$$\Theta = (\theta - \theta_r)/(\theta_s - \theta_r) = \left\{1+[\alpha \psi]^{1/n}\right\}^{-m}$$  \hspace{1cm} (5)

Where the $\Theta$ is the effective saturation and $a, n, m$ are fitting parameters. $m$ is set to 1-n$^{-1}$ (Van Genuchten 1980)

3 Results and discussion

3.1 The possibility for GLD to improve the HC and WRC of a till that is to be used in a sealing layer on top of sulfidic mine waste

The hypothesis was that the addition of GLD would improve the HC of the tills that alone would not reach the requirement. Previous results have shown promising results for a till-GLD mixture to be used as a sealing layer, with low HC, high WRC and increased compaction properties (Hargelius 2008; Mäkitalo et al. 2015 a and b). After a certain limit of GLD addition (10-15 w. %) the high water-content and low shear strength of the GLD was expected to decrease the compaction degree and therefore increase the HC in the mixtures. The HC of the mixtures with the silty till 1 and the sandy till did improve (decrease), but not enough to reach below the in Sweden commonly required 10$^{-8}$ m/s (Figure 2). This study confirms the hypothesis that the initial HC of the till that is used in the mixtures plays a major role in controlling the HC of the final mixture. The HC decreases only slightly with addition of GLD to two of tills studied and increases in one of the tills. One reason for the unexpectedly low decrease of HC with addition of GLD might be the method of compaction that was chosen. In this study a standardized PC method was selected to limit the differences between the samples to imitate compaction in field. One disadvantage with using proctor compaction, is that a small margin on the sides of the cylinder is not properly compacted as the weight does not reach all the way to the sides. This can lead to a preferential flow along the edges of the cylinder and a higher HC than in the field. In the study by Mäkitalo et al. 2015 the compaction was made by hand However,
if and how much the compaction method affects HC needs further investigation. The study by Mäkitalo et al. 2015 conducted no HC measurements on pure till, which makes comparison between the studies difficult. Also it can not be said if the HC improved with GLD addition to the till used in that study.

The hypothesized increasing trend followed by a decrease is seen in this study. First the HC of the silty till-1 decreased from 3.5 E-08 to 1.5E-08 m/s at an addition of 5 wt. % GLD to then increase up to 7E-08 m/s at up to 20 wt. % GLD in the mixtures (Figure 2). In the sandy till the HC decreased from 7E-08 to 2E-08 m/s when adding 5 to 10 wt. % of GLD. The HC then increased to 4E-08 m/s when adding up to 20 wt. % of GLD. However, only the HC in silty till-2 meets the common requirements of a sealing layer in Sweden, i.e. a HC below 10^-8 m/s, and with addition of GLD the HC increased from 3E-10 to 5E-09 m/s with up to 20 wt. % of GLD (Figure 2).

Figure 2. The (HC) in the different mixtures of till and GLD. The HC of the silty till 1- and the sandy till-mixtures are on the left y-axis and the silty till 2-mixture on the right y-axis.

Considering WRC, the hypothesis was that the addition of GLD would improve the WRC of the tills that alone would not reach the requirement of a sealing layer (HC <10^-8 m/s). This due to the high initial WRC of the GLD (Mäkitalo et al. 2014). The SWCC for the different till-GLD mixtures shows that WRC does improve with GLD addition (Figure 3; Table 2). At 85 % S, the matric suction increased from 3.2 to 6 mH2O with 15 wt. % GLD addition to the silty till 1 (Figure 3:A-D; Table 2), 3.3 to 26 mH2O in the sandy till mixtures (Figure 3:E-H; Table 2) and from 3.6 to 50 mH2O in the silty till 2 mixtures (Figure 3:I-L; Table 2). The more GLD that is added the higher WRC is achieved in the mixtures. To minimize oxygen diffusion the sealing layer should be kept at a saturation degree of at least 85 % (Corey 1957, Aubertin and Mbonimpa 2001; Achib et al. 2004) and with 15 wt. % GLD addition to the tills a high saturation is kept even at high suction (Figure 3).

For many mixtures the saturation is greater than 100 %, for these cases the best fit van Genuchten curve were set to start at 100 % saturation (Figure 3), as a saturation above 100 % is not possible. Calculated saturation values above 100 % might be explained by the GLDs tendencies to shrink when drying. Volumetric shrinkage is due to the rearrangement of particles and aggregates during drying causing a decrease in bulk volume (Gapak et al. 2017). The shrinkage has an impact on the SWCC as
the total volume that is used in the saturation calculations is not constant. If the material has tendencies towards volumetric shrinkage behavior when dried, they would first swell of the water addition when saturated, leading to an increase in volume and Sr. However, also the pure tills with no GLD addition have Sr higher than 100 %, so the shrinkage behavior might not be the only explanation. An incorrect measurement of the ring-volumes is not a likely explanation, as the Sr never reaches below 100 % for the first pressure step. Therefore, another explanation that could lead to increased Sr is a loss of material when the samples are taken out to be weighed. Some material was left underneath the ceramic plates that are placed on top of the samples. This even though “separators” higher than the rings were placed between the plates. However, this is not a likely explanation for the oversaturated samples as they were placed on the top ceramic plate. Also, two ways of calculating Sr was used, one with the water content calculated from the material before compaction and one where the water content in the samples was oven dried after the pressure steps. Both values show oversaturated samples in the first pressure step. The common parameters between the two ways of calculating Sr is the volume of the samples and the particle density of the materials used. An error in the estimation of the particle density is not likely the only explanation as, to decrease the Sr to 100 %, needs to be increased to a value that is not likely for a till (3 g/cm³). Volumetric shrinkage is therefore the likely explanation to the overestimated calculated saturation of the materials. The presence of material on the bottom of the ceramic plates might be evidence for that volumetric shrinkage is occurring and a volumetric shrinkage curve (VSC) should be obtained and the results from that included in the SWCC (Fredlund 2002; Wijaya et al. 2015). The VSC present the relationship between the volume of the specimen and the corresponding water content.

The hysteretic effect is also a parameter that will impact the interpretation of the SWCC and the actual WRC of the mixtures studied. The hysteretic effect will give higher matric suction for drying than for wetting for the same water content (Tinjum et al. 1997), and the \( \psi_{85} \) will be higher. This will lead to an underestimation of the WRC in laboratory experiments compared to actual values in field.

To summarize, the HC and the WRC for tills that does not meet the HC requirements for a sealing layer material improves with GLD addition. However, the improvement in HC is not enough to reach the requirements. The WRC on the other hand shows promising results, especially for 15 wt. % GLD addition. The WRC can be seen as the most important parameter minimizing the oxygen diffusion through a sealing layer, by keeping the layer saturated. The WRC results show an increase the more GLD is added. However, due to compaction difficulties more than 20 wt. % is not recommended.

There is a difference in both HC and WRC between the mixtures of the different tills, and a characterization of the till as a bulk material and its mixture with GLD, both considering HC and WRC is needed. It would be interesting that in future studies connect the WRC with actual results from oxygen diffusion. This to see what WRC values are required to get minimum oxygen diffusion through the material.

Table 2. Van Genuchten parameters, matric suction for 85 % Sr (\( \psi_{85} \)), the air entry suction (\( \psi_a \)), dry density (\( \rho_d \)), water content (\( w \)), compaction degree (\( R_d \)) and if the water content is dry or wet of optimal water content.

<table>
<thead>
<tr>
<th>Dry/wet of opt. w</th>
<th>( \psi_{85} )</th>
<th>( \theta_r )</th>
<th>( \theta_s )</th>
<th>n</th>
<th>m</th>
<th>( \alpha )</th>
<th>( \psi_a )</th>
<th>( \rho_d )</th>
<th>w</th>
<th>( R_d )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silty till 1</td>
<td>3.2 0.025 0.34 4 0.75 0.22</td>
<td>2.5 1.8</td>
<td>0.09</td>
<td>87% w</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Silty till 1 + 5 wt. % GLD</td>
<td>3.4 0.05 0.34 2.5 0.60 0.2 2</td>
<td>1.95 0.12</td>
<td>95% w</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Silty till 1 + 10 wt. % GLD</td>
<td>4.8 0.06 0.34 2.6 0.62 0.15</td>
<td>3</td>
<td>1.83 0.15</td>
<td>92% w</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Silty till 1 + 15 wt. % GLD</td>
<td>6 0.07 0.34 2.6 0.62</td>
<td>12</td>
<td>3</td>
<td>1.78 0.19</td>
<td>91% w</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sandy till</td>
<td>3.3 0.04 0.34 3.6 0.72 0.27</td>
<td>3</td>
<td>1.77 0.07</td>
<td>87% w</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sandy till + 5 wt. % GLD</td>
<td>1.2 0.045 0.34 1.8 0.44</td>
<td>0.7</td>
<td>0.7</td>
<td>1.83 0.12</td>
<td>91% w</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sandy till + 10 wt. % GLD</td>
<td>3.3 0.02 0.34 1.35 0.26</td>
<td>0.3</td>
<td>1</td>
<td>1.81 0.16</td>
<td>91% w</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sandy till + 15 wt. % GLD</td>
<td>26 0.03 0.34 1.15</td>
<td>0.13</td>
<td>0.1</td>
<td>4</td>
<td>1.77 0.19</td>
<td>92% w</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Silty till 2</td>
<td>3.6 0.09 0.34 1.5 0.33</td>
<td>0.9</td>
<td>2</td>
<td>1.92 0.07</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Silty till 2 + 5 wt. % GLD</td>
<td>11 0.01 0.34 1.28 0.22</td>
<td>0.1</td>
<td>4</td>
<td>1.89 0.13</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Silty till 2 + 10 wt. % GLD</td>
<td>22 0.00001 0.34 1.37 0.27</td>
<td>0.04</td>
<td>10</td>
<td>1.86 0.17</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Silty till 2 + 15 wt. % GLD</td>
<td>50 0.005 0.34 3 0.67</td>
<td>0.012</td>
<td>25</td>
<td>1.75 0.2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
3.2 The effect of fines- and clay content in till on HC and WRC in till-GLD-mixtures.

The PSD of the GLD showed that the majority was finer than 63 μm (Figure 4 and Table 3). The Silty till-1 consisted of 35 % fines of which 2.6 % was in the clay fraction. Silty till-2 had the same percentage of fines as the silty till-1, but a higher percentage of these were in the clay fraction, 4.3 %. The sandy till had a lower percentage of fines and clays (14 % fines and 0.7 % clays; Figure 4 and Table 3).

A decrease in HC is seen for the silty till 1 and sandy till with addition of GLD up to a wt. % of 5-10 (Figure 2). This was expected as the GLD increases the fine-grained material in the mixtures and
results in decreasing HC (Benson et al. 1994; Benson and Trast 1995; Leroueil et al. 2002). The HC of the silty till 2 does not decrease at the addition of GLD as the tills initial clay content leaves no space for the GLD in its micropores. Therefore, the addition of GLD to the silty till 2 is only deteriorating the compaction degree of the mixture due to an increase in water content, leading to a higher HC. The same mechanism increases the HC when more than 5 and 10 wt. % of GLD is added for the silty till 1 and the sandy till.

The till as a heterogenic material and a dominant part of the mixture, was expected to have a significant impact on especially the HC of the mixtures. The finer the till is, the lower the HC in the mixture was expected. The higher clay content in silty till 2 is likely the reason for the much lower HC compared to the silty till 1 (Figure 2). In a study conducted by Leroueil (2002) the HC decreased several orders of magnitude when the clay size fraction increased from 2 to 12 %. Also, a study conducted by Benson et al. (1994) showed a strong relationship between clay content and HC and a weak relationship between fines and HC when studying clay.

![PSD of the three different tills and the GLD](image)

Figure 4. PSD of the three different tills and the GLD.

Table 3. The total solid content (TSC), the fines- and clay contents in the mixtures. n= number of analyzed samples.

<table>
<thead>
<tr>
<th>Material</th>
<th>TSC (%)</th>
<th>Content of fines (&lt;63µm)</th>
<th>Content of clays (&lt;2µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silty till 1</td>
<td>91.5±0.4  (n=9)</td>
<td>34±5 % (n=9)</td>
<td>2.6 % (n=1)</td>
</tr>
<tr>
<td>Sandy till</td>
<td>91.7±0.4  (n=9)</td>
<td>14±1 % (n=3)</td>
<td>0.7 % (n=1)</td>
</tr>
<tr>
<td>Silty till 2</td>
<td>91.1±0.8  (n=6)</td>
<td>35±1 % (n=2)</td>
<td>4.3 % (n=1)</td>
</tr>
<tr>
<td>GLD</td>
<td>43±4  (n=12)</td>
<td>76±29 % (n=3)</td>
<td>5.4±4.1 % (n=3)</td>
</tr>
</tbody>
</table>

For the WRC, an increase in $\psi_a$ and a decrease in $n$ was expected, with higher GLD addition. This as materials with smaller pores generally has a higher $\psi_a$ (Fredlund and Rahardjo 1993; Sillers et al. 2001; Tinjum et al. 1997) and a flatter slope of the SWCC in the desaturation zone (Sillers et al. 2001).
This is a general trend in these experiments as well (Table 2; Figure 3). However, comparing the tills with no GLD added, the silty till 2 should have the highest $\psi_a$, this is not the case. The tills have similar $\psi_a$, 2 -3 (Table 2). The soil macro structure, mainly aggregation has shown to impact the $\psi_a$ and the slope of the SWCC in fine-grained soils compacted dry of optimum water content (Vanapalli et al. 1999). The soil will then act more like a coarse-grained soil due to its highly aggregated macrostructure, with steeper SWCC and lower $\psi_a$ (Vanapalli et al. 1999). The silty till 2 is in fact highly aggregated, but due to shortage on material there is no data on the optimal water content for this material. The dry density is another factor that has shown to have significant impact on the SWCC due to a decrease in the number of voids with increasing dry density, leading to faster saturation (Vanapalli et al. 1999). However, the differences in dry density between mixtures is not great (Table 2) and cannot explain the lack of a higher WRC of the silty till 2, compared to the silty till 1 and the sandy till. Neither can it explain the higher WRC in the sandy till + 15 wt. % GLD compared to the silty till 1 + 15 wt. % GLD. Mineralogy is another factor that is known to affect the SWCC of a soil (Mitchell 1993; Tinjum et al. 1997) and needs further investigation.

The study indicates that there are other factors other than the PSD of the materials that controls the HC of the mixtures, as initial water content, dry density and WRC. The high WRC in the till-GLD mixtures with 15 wt. % GLD addition show promising results for GLD to be sued in a sealing layer, even though the HC did not reach the requirements. A high saturated sealing layer is the key factor of deterring oxygen diffusion to the mine waste. The recycling of GLD for mine remediation purposes is an important step towards circular economy and beneficial for both the industry providing the residue and the mining company. For future studies, it would be interesting to study how dry density and initial water content of the tills and GLD affects the HC. It is also important to study how the compaction method affects the HC of a till and GLD-mixture. In view of WRC the SWCC indicates that the contents of fines and clays in the tills is not the main factor determining the WRC. It would be interesting to study the mineralogy of the tills and the GLD to better explain the behavior of the SWCC.

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References


The drivers of hydraulic conductivity in till-GLD mixtures to be used in a sealing layer on top of mine waste

Susanne Nigéus and Christian Maurice
The drivers of hydraulic conductivity in till-GLD mixtures to be used in a sealing layer on top of mine waste

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Abstract

The mining industry produces massive amounts of waste that without treatment and in contact with oxygen can result in acid rock drainage (ARD) and increased leaching of metals. In Sweden, the common way to treat the mine waste after closure is to apply a dry coverage on top of the waste. The access to a suitable cover-material that meets the requirements is however limited, which creates a great driving force for the recycling of industrial residues for these purposes. The recycling of industrial residues is beneficial for many parts, as the industry producing the residue, the mining industry that will use the residue and the society in its strive for zero waste. Green Liquor Dregs (GLD) is an industrial residue from pulp production in a paper mill that has potential to be used in a sealing layer together with till. Hydraulic conductivity (HC) is commonly used as a measure to evaluate the quality of a sealing layer. Estimating HC is however challenging as there are many factors controlling it, both in field, but even more so in laboratory measurements. In this study 5 to 20 wt. % of GLD from two different paper mills, with different total solid content (TSC) and particle size distribution (PSD), was mixed with a silty till of different TSC to investigate how the dry density and initial water content of the materials affect the HC. Furthermore, the compaction effort and its effect on the HC was investigated. This laboratory study concludes that the initial water content of the materials is an important factor determining the HC of the till-GLD mixture. With a drier till and GLD, more GLD can and should be added to the mixture to reach the optimum HC. The dry density and compaction effort on the other hand does not seem to be the driving factor determining the HC of the mixtures in this study.

Keywords: Green Liquor Dregs; Sealing layer; Optimal water-content; Compaction effort; Hydraulic conductivity
Introduction

Mining is worldwide one of the biggest producers of waste. In Sweden 110 million tons of mine waste was produced year 2017, accounting for 77 % of all waste produced during that year (Swedish EPA 2018). Approximately 70 % of the generated mine waste in Sweden contains sulfide minerals (SGU and Swedish EPA 2017). Sulfidic mine waste, if left in contact with oxygen and humidity has the potential to produce acid rock drainage (ARD) by sulfide oxidation (Saria et al. 2006). ARD is a major long-term threat to the environment as metals and metalloids may become mobile (Saria et al. 2006). The GARD guide (Global Acid Rock Drainage guide; Verburg et al. 2009) categorize different methods to prevent ARD after closure, into two main categories; engineered barriers and water covers. Engineered barriers can be divided into liners and dry covers, where liners are typically designed to act as a barrier for contaminant flow from the overlying waste into the receiving environment. Dry covers are typically designed to limit the ingress of water and oxygen into the underlying waste. Under the relatively humid climatic conditions in Sweden, a soil cover can be used to reduce oxygen flux to the underlying reactive wastes, and thus reduce ARD (Collin and Rasmuson 1990; Bussièire et al. 2003; Dagenais et al. 2006). The sealing layer is the key element of the soil cover and consists of fine-grained material and is applied closest to the mine waste. It is followed by the protection layer, protecting the sealing layer from frost and root-penetration. A vegetation layer on top is protecting the protection layer from erosion. The main function of the sealing layer is to limit oxygen diffusion through the cover into the waste dump, thereby preventing the oxidation of sulfides and the generation of acidic drainage and the release of metals (Höglund et al. 2004). Another function with the sealing layer is often to limit the infiltration of precipitation into the waste dump, this requires a low HC. In addition to a low HC, a sealing layer generally shows good water retention properties leading to a high degree of saturation in the barrier (Höglund et al. 2004) which reduces the effective oxygen diffusion coefficient ($D_o$) to the $D_i$ in water. It is then considered to be small enough to reduce the oxygen flux to a level comparable to that of a water cover (Yanful 1993; Aachib et al. 2004). Due to challenges measuring oxygen diffusion, mainly due to oxygen leakage from the atmosphere, HC is often used to estimate the quality of a sealing layer. Studies by Leroueil (2002) and Watabe (2000) on glacial till show that the HC is highly dependent on the degree of compaction, with significantly decreasing values at decreased void ratio during loading. Furthermore, the molding water content in the material is affecting the HC (Benson and Trast 1995; Leroueil et al. 2002), i.e. the water-content after compaction. A study conducted by Benson and Trast (1995) on compacted clays shows that the lowest HC was reached at a molding water-content of 1 to 2 % wet of the line of optimums. The optimum water-content is the molding water-content at which the highest dry density can be reached.

Mäkitalo et al. (2015a; 2015b), Hargelius et al. (2008) and Nigéus et al. (2018) have shown that a residue from pulp production, GLD, has as an amendment to till potential to be used in a sealing layer. GLD is an alkaline, inorganic waste originating from the recycling process at sulfate pulp and paper mills. The main solid compounds of GLD consists of CaCO$_3$, Mg(OH)$_2$, C and metal sulfides, especially FeS (Jia et al. 2014; Sanchez and Tran 2005). The liquid phase of the GLD consists of Na$_2$CO$_3$ and NaOH, which generates its characteristic high pH. GLD is regarded as an inert material (Mäkelä and Höynälä 2000) and has the same grain size distribution as silt (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/cm$^3$, a compact density of 2.47 to 2.60 g/cm$^3$, a low HC (10$^{-8}$ and 10$^{-9}$ m/s) and a high WRC (Mäkitalo et al. 2014). Sequential extraction has been performed on GLD and indicates relatively low bioavailability of metals in general (Nurmesniemi et al. 2005). Previous studies have shown promising results for a till-GLD mixture to be used as a sealing layer, with low HC, high water retention capacity (WRC) and increased compaction properties (Hargelius 2008; Mäkitalo et al. 2015a). The GLD consist of a fine material that will fill up the pores in the till and potentially decrease its HC. Nigéus et al. (2018) found in their study that the decrease in HC with GLD addition was not enough to reach the in Sweden commonly required 10$^{-8}$ m/s, but then again, the mixtures showed good WRC, especially for 15 wt. % of GLD addition, which suggest potential for low oxygen diffusion. They also studied how the HC of a mixture of GLD and till is affected by different contents of fines/clays and found that the contents of clay play a major role in determining the final HC of the mixture, the higher the clay-content the lower the HC (Nigéus et al 2018). The study suggested that...
there might be other factors, as initial water content, dry density after compaction and compaction effort that can affect the HC of the GLD-till mixtures.

In this study 5 to 20 wt. % GLD, from two different paper mills, with different total solid content (TSC) and particle size distributions (PSD), were mixed with a sieved (<20 mm) silty till with different TSC to investigate how the initial water-content, compaction effort and dry density after compaction affect the HC. A maximum of 20 % GLD was added to the mixture, as a mixture with a higher GLD content leads to liquefaction and difficulties to compact.

The hypothesis of this study is that a decrease in initial water-content of till and GLD is expected to lead to lower HC as higher amounts of GLD are added, as the final water-content after compaction will decrease to a value closer to the optimal water-content. The mixtures are expected to reach the lowest HC at water contents a few percent wet of the optimal water-content. Lower amounts of GLD in the mixtures is expected to lead to higher dry density and lower HC, due to a higher amount of finer material in the mixtures that decreases their porosity. However, as the amount of GLD increases, other properties of the GLD, as its low shear strength and high water-content, are anticipated to have more influence in the HC of the mixtures. The mixtures become more difficult to compact as the water-content increases above the optimal molding water-content. A decreased compaction effort was expected to increase the dry density after compaction and decrease the HC in the till-GLD mixtures.

With increased strain of the material, the water retained in the micro-pores of the GLD will be released and lead to a higher porosity. The objectives of this experimental study were to find out i) how the initial water-content of a silty till and GLD respectively will affect the HC of the mixtures, (ii) how dry density after compaction correlates with the HC of a silty till and its mixtures with GLD, (iii) how compaction effort affects the dry density and HC of silty till-GLD mixtures.

2 Materials and methods

2.1 Materials

A silty till from a till quarry (Brännkläppen, Boden, Sweden) was used as the bulk material. The till was air-dried and sieved through a 20-mm sieve and the particles above 20 mm removed. This as particles above 20 mm will affect negatively on the effectiveness of the materials used as the sealing layer.

GLD from two different paper mills (GLD1: Smurfit Kappa paper mill, Piteå, Sweden; GLD2: Billerud Korsnäs paper mill, Kalix, Sweden) were used in the experiments. The GLD was collected in sealed plastic containers to preserve the water content of the material.

The till was mixed with 5, 10, 15, and 20 wt. % of GLD. The weight percentage was calculated towards a dry till and a naturally moist GLD. The GLD was kept naturally moist as it is difficult to rewet which affects its physical properties. The mixing was carried out by hand with a small shovel until the mixture was estimated to be homogenized. An overview of the different mixtures and analyses performed on them can be seen in Figure 1.
2.2 Particle size distribution

The till was washed and dry sieved according to SS-EN 933-1:2012 to obtain the weight percentage of fines in the material. A mechanical sieve tower (Retsch AS 200) with an amplitude of 2.2 mm/g was used. The cut-off sizes were 12.5, 10, 8, 5, 4, 2, 1, 0.5, 0.25, 0.125, and 0.063 mm.

PSD for the fines and the two GLD were done by laser diffraction analysis on triplicate samples of each material by a CILAS Granulometer 1064 (CILAS, Orleans, France). With a compact cast iron optical bench, the CILAS 1064 integrates 2 sequenced laser sources positioned at 0° and 45°, to produce a diffraction pattern analyzed on a 64 channel silicon detector. The PSD is then calculated using the CILAS software. The results are presented in Figure 2.
2.3 Particle density

The particle density of the materials was determined using an AccuPyc II 1340 Pycnometer, which uses the gas displacement method, to measure volume accurately. The samples were sealed in the instrument compartment of known volume, an inert gas was admitted and then expanded into another precision internal volume. The pressures observed upon filling the sample chamber and then discharging it into a second empty chamber allowed computation of the sample solid phase volume. The results are presented in Table 1.

Table 1. Particle density (g/cm³) and total solid content (TSC) for the materials used.

<table>
<thead>
<tr>
<th>Material</th>
<th>Particle density (g/cm³)</th>
<th>TSC (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silty till</td>
<td>2.71</td>
<td>91.5±0.4% (n=9)</td>
</tr>
<tr>
<td>GLD1</td>
<td>2.49</td>
<td>43±4 % (n=12)</td>
</tr>
<tr>
<td>GLD2</td>
<td>2.63</td>
<td>58±0.3 % (n=3)</td>
</tr>
</tbody>
</table>

2.4 Proctor compaction

Proctor compaction (PC) was carried out according to standard SS-EN 13286-2:2010. Also, a lighter compaction weight compared to standard PC and drop length was used in this study. For the light PC, a 2.5 kg weight was dropped from a height of 30 cm on the sample. The PC-tests were started with triplicates, but first results showed almost no variations, wherefore only one or two repetitions were made. The PC was done on air-dried till and naturally moist GLD. The dry density and the water content (w) were calculated after equation 1 and 2:

\[
p_d = \frac{m}{(1+w) + V},
\]

\[
w = \frac{(m-m_d)}{m_d},
\]
where $\rho_d$ is dry density, $w$ is water-content, $m$ is the mass of the moist sample, $m_d$ is the mass of the dry sample and $V$ is the sample volume.

### 2.5 Hydraulic conductivity

HC measurements were conducted on a moist (TSC 91±1 %, n=24) and a drier till (TSC 96 %) with 5, 10, 15 and 20 wt. % addition of GLD. The TSC of each material and mixture used for HC analysis were evaluated by drying them in an oven (105°C for 24 h) according to the SIS standard SS-EN 14346:2007. The constant head-method was used in airtight cylinders with a volume of 996 cm³. The walls of the cylinders were coated with a thin layer of bentonite to prevent the preferential wall flow. The mixtures inside the cylinder were compacted with standard PC. Silty till with an addition of 5-20 wt. % of GLD1 was also compacted with a light PC.

Water was lead to the bottom of the cylinder with a hydraulic gradient of 8.7. The water passing through the cylinders were collected in plastic bottles, 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. The HC was calculated using Darcy’s law. The analysis was done with three sets on each sample.

### 3 Results and discussion

#### 3.1 The effect of initial water content of the materials on HC of the mixtures

A lower initial water-content of till and GLD was expected to improve the HC when higher amounts of GLD was added, as the final molding water-content of the mixture would be closer to the optimal water-content. The hypothesis is confirmed in this study. The mixture with the wetter till (silty till – GLD1) decreases from 3 to 2E-08 m/s with an addition of 5 wt. % of GLD, to then steadily increase with the further addition of GLD (from 2 to 7E-08 m/s; Figure 3: A; Table 2). In the drier till mixture (silty till-GLD1-drier till) the initial HC was higher, 10E-08 compared to 3E-08 m/s (Figure 3:A; Table 2). The HC then decreases steadily with increasing addition of GLD (from 10 to 3E-08 m/s) until 20 wt. % of GLD addition, where a slight increase in HC is seen (from 3 to 4 E-08 m/s; Figure 3:A; Table 2). When the GLD addition to the drier till reached over 10 wt. % the HC became lower than in the moister mixture, 3E-08 compared to 7-8E-08 (Figure 3:A; Table 2).

The decrease in HC in the drier till-mixture can be explained by the correlation between HC and the degree of compaction (Benson et al. 1994; Watabe et al. 2000; Leroueil et al. 2002). Compacting a drier till results in higher dry densities in mixtures with 5-15 wt. % GLD addition, approximately 0.1 to 0.2 g/cm³, compared to a wetter till (Figure 3:B). However, the HC in the drier mixture only reaches lower values than in the wetter with 15-20 wt. % GLD addition. Adding 15 wt. % GLD to moist till increases the water-content far beyond the optimal water-content (Figure 3:C; Table 2), leading to a lower dry density and a higher HC than in a mixture with a drier till. Benson and Trast (1995) showed that the HC is sensitive to the molding water-content, where the lowest HC was reached at molding water-content of 1-2 % wet of optimum water content. Around 5 wt. % of GLD1 seems to be a threshold for tills with a TSC content of approximately 91 % and with an increasing amount of GLD above that the water-content increases beyond the 1-2 % wet of optimum. A drier till can, therefore, tolerate a higher amount of GLD-addition compared to a wetter till (Figure 3:A and C). In fact, a higher amount of GLD is needed to reach the lowest HC in the mixtures with a drier till. At 15 wt. % of GLD1 addition the 1-2 % wet of optimum water level is reached and here is where the lowest HC is seen in the drier till mixture.

An addition of a drier GLD was expected to decrease the HC of the mixture. In this study, the HC stays around 3E-08 m/s with an addition of up to 15 wt. % of the drier and fine-grained GLD2 and does not follow the same u-shaped trend as when using GLD1 (Figure 3:A). As for the mixture with the drier till there seems to be a higher tolerance of how much drier GLD can be added to the silty till compared to the more wet GLD. This is likely due to the optimal water content, that for 10-15 wt. % addition of GLD1 reaches 5-8 % wet of optimum water-content after compaction, for GLD2 the corresponding number is 4 % (Figure 3:C). Another explanation to that a higher amount of GLD2 can be added to the mixture before an increase in HC occurs may be that GLD2 is more fine-grained than
GLD1. Even if the dry density decreases with more addition of GLD, the increase of fine-grained material levels out the expected increase in HC. This trend was also seen in a previous study with till and GLD-mixtures, where the mixtures with till with a higher amount of clay had a lower HC (Nigéus et al. 2018).

Table 2. Hydraulic conductivity (HC), dry density (ρd), water-content (w) and fines/clay of the mixtures tested in this study. The data for the silty till and GLD1 is from Nigéus et al (2018) and is used for comparison.

<table>
<thead>
<tr>
<th>GLD</th>
<th>HC (n,m,s)</th>
<th>p_d (g/cm³)</th>
<th>w (%)</th>
<th>Wet of opt. w (%)</th>
<th>Opt. molding w (%)</th>
<th>Fines (%&lt;63μm)</th>
<th>Clay size (%&lt;2μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silty till - GLD1</td>
<td></td>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0%</td>
<td>3E-08:3E-00</td>
<td>2.01±0.05</td>
<td>9±0.5</td>
<td>1</td>
<td>2.08±0.01</td>
<td>6</td>
<td>34.5±9.0</td>
</tr>
<tr>
<td>5%</td>
<td>2E-08:3E-00</td>
<td>1.97±0.02</td>
<td>10±0.6</td>
<td>2</td>
<td>2.05±0.01</td>
<td>8</td>
<td>34.5±9.0</td>
</tr>
<tr>
<td>10%</td>
<td>4E-08:3E-00</td>
<td>1.88±0.01</td>
<td>14±1.3</td>
<td>5</td>
<td>1.99±0.01</td>
<td>9</td>
<td>34.5±9.0</td>
</tr>
<tr>
<td>15%</td>
<td>6E-08:3E-08</td>
<td>1.74±0.05</td>
<td>18±0.4</td>
<td>8</td>
<td>1.95±0.00</td>
<td>10</td>
<td>34.5±9.0</td>
</tr>
<tr>
<td>20%</td>
<td>7E-08:3E-08</td>
<td>1.65±0.02</td>
<td>20±0.7</td>
<td>8</td>
<td>1.97±0.01</td>
<td>10</td>
<td>34.5±9.0</td>
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<tr>
<td>Silty till - GLD2</td>
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<td></td>
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<td></td>
</tr>
<tr>
<td>0%</td>
<td>3E-08:4E-00</td>
<td>1.96±0.02</td>
<td>10±0.5</td>
<td>3</td>
<td>2.09±0.01</td>
<td>6</td>
<td>34.5±9.0</td>
</tr>
<tr>
<td>5%</td>
<td>3E-08:4E-00</td>
<td>1.91±0.02</td>
<td>12±0.3</td>
<td>4</td>
<td>2.05±0.01</td>
<td>8</td>
<td>34.5±9.0</td>
</tr>
<tr>
<td>10%</td>
<td>3E-08:4E-00</td>
<td>1.72±0.02</td>
<td>14±0.8</td>
<td>4</td>
<td>2.04±0.01</td>
<td>8</td>
<td>34.5±9.0</td>
</tr>
<tr>
<td>15%</td>
<td>3E-08:4E-00</td>
<td>1.54±0.01</td>
<td>19±0.4</td>
<td>4</td>
<td>1.99±0.01</td>
<td>10</td>
<td>34.5±9.0</td>
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<tr>
<td>20%</td>
<td>3E-08:4E-00</td>
<td>1.54±0.01</td>
<td>25</td>
<td>4</td>
<td>1.91±0.01</td>
<td>15</td>
<td>34.5±9.0</td>
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<tr>
<td>Silty till - GLD1</td>
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</tr>
<tr>
<td>0%</td>
<td>4E-08</td>
<td>2.04±0.02</td>
<td>9±0.5</td>
<td>2</td>
<td>2.1±0.01</td>
<td>6</td>
<td>34.5±9.0</td>
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<tr>
<td>5%</td>
<td>4E-08</td>
<td>1.95±0.02</td>
<td>11±0.9</td>
<td>4</td>
<td>2.03±0.01</td>
<td>8</td>
<td>34.5±9.0</td>
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<tr>
<td>10%</td>
<td>4E-08</td>
<td>1.83±0.05</td>
<td>15±0.6</td>
<td>3</td>
<td>1.83±0.01</td>
<td>10</td>
<td>34.5±9.0</td>
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<tr>
<td>15%</td>
<td>4E-08</td>
<td>1.72±0.02</td>
<td>19±0.4</td>
<td>4</td>
<td>1.81±0.01</td>
<td>15</td>
<td>34.5±9.0</td>
</tr>
<tr>
<td>20%</td>
<td>4E-08</td>
<td>1.54±0.01</td>
<td>25</td>
<td>4</td>
<td>1.79±0.01</td>
<td>10</td>
<td>34.5±9.0</td>
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<tr>
<td>Silty till - GLD2</td>
<td></td>
<td></td>
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<td></td>
</tr>
<tr>
<td>0%</td>
<td>4E-08</td>
<td>2.02±0.02</td>
<td>9±0.5</td>
<td>2</td>
<td>2.1±0.01</td>
<td>6</td>
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</tr>
<tr>
<td>5%</td>
<td>4E-08</td>
<td>2.06±0.02</td>
<td>7±0.5</td>
<td>1</td>
<td>2.03±0.01</td>
<td>8</td>
<td>34.5±9.0</td>
</tr>
<tr>
<td>10%</td>
<td>4E-08</td>
<td>2.03±0.04</td>
<td>10±1.3</td>
<td>1</td>
<td>1.83±0.01</td>
<td>10</td>
<td>34.5±9.0</td>
</tr>
<tr>
<td>15%</td>
<td>4E-08</td>
<td>1.93±0.01</td>
<td>14±0.4</td>
<td>4</td>
<td>1.81±0.01</td>
<td>15</td>
<td>34.5±9.0</td>
</tr>
<tr>
<td>20%</td>
<td>4E-08</td>
<td>1.66±0.01</td>
<td>21</td>
<td>4</td>
<td>1.79±0.01</td>
<td>10</td>
<td>34.5±9.0</td>
</tr>
</tbody>
</table>

GLD1: 76±29 (n=3)*
GLD2: 1E-08:3E-00 (n=3) *

*(Mäkitalo et al. 2014).
Figure 3. A) Hydraulic conductivity (HC), B) dry density after compaction (ρ₃), and C) water-content (w) in the different mixtures of till and GLD. Silty till – GLD1 - drier till represents a till with half the water-content as used in the rest of the laboratory tests. Silty till–GLD1-LC is compacted with less compaction effort (lower weight and drop height) than a standard PC. The transparent thick lines are the 1-2 % wet of optimum w for each mixture.

In summary, this study shows that the initial water-contents of the materials play a major role for the dry density that can be reached after compaction and determining the HC of the mixtures. It determines the degree wet of optimum that in this and previous studies has shown to greatly affect the HC (Benson et al. 1994; Benson and Trast 1995). This study confirms the results from Benson and Trast (1995) that 1-2 % wet of the optimum water-content is the target to reach the lowest HC in a material. A drier till and GLD implies that more GLD can be added to the mixture for reaching the optimal HC. If the till is dry, a higher amount of GLD is needed to reach the optimum HC. If the GLD is drier, the HC was independent of the amount of GLD added. However, none of the mixtures reached the in Sweden commonly required HC for sealing layer, <10⁻⁸ m/s.

3.2 The effect of dry density on HC of the till-GLD mixtures

The hypothesis was that lower amounts of GLD addition to the tills would increase the dry density and decrease HC, due to the increase in the finer material in the mixtures that decreases the porosity. However, as the amount of GLD increases, other properties of the GLD as its low shear strength and high water-content, was anticipated to have more influence in the HC of the mixtures as it becomes more difficult to compact with a water-content above the optimal molding water-content of the mixture. The expected increase in dry density with smaller amounts of GLD-addition could not be
seen in any of the mixtures, except from the drier till-mixture where a slight increase can be seen with 5 wt. % of GLD addition. In this mixture, higher dry densities, in general, can be seen when adding GLD, compared to when using a wetter till (Figure 3:B). In the rest of the mixtures, the dry density decreases with increasing addition of GLD. The lack of increasing dry density is most likely due to the higher water contents, beyond the preferred 1-2 % wet of optimum water content (see discussion in section 3.1), and the lower particle densities of GLD (Table 1).

However, regarding HC tests compacted with standard PC-method the hypothesis is confirmed. In the silty till – GLD 1 mixture the HC is reaching its optimum HC at 2E-08 m/s when adding 5 wt. % of GLD1 to the silty till, to then increase up to 7E-08 m/s with 10 wt. % or more GLD in the mixtures (Figure 3:A; Table 2). The HC in the drier till-mixture is as discussed in section 3.1 reaching its optimum HC with 15 wt. % of GLD. Also, a slight decrease is detected in the silty till- GLD 2-mixture, 3.3-2.6E-08 m/s, with the optimum HC at 15 wt. % GLD addition. The decreases in HC were expected and can be explained by the properties of the GLD, i.e. high content of fine particles (Table 1), together with its high WRC (Mäkitalo et al. 2014). So, even if the dry density decreases with increasing amount of GLD in the mixtures the HC is improving due to the increase in fine material. However, when more than 5 wt. % of GLD1 or 10 wt. % of GLD 2 is added to the wetter till and more than 15 wt. % to the drier till, the water-content increases above the optimum water-content where the highest dry density can be reached (Figure 3:C; Table 2; see discussion in section 3.1). When using light proctor compaction (LC) the behavior of the HC is different compared to the rest of the mixtures. The HC is increasing with increasing GLD addition (4E-08 to 9E-08 m/s; Figure 3:A; Table 2). The lack of decrease that could be seen in the other mixtures with lower percentages of GLD is not due to dry density as it is similar between the samples compacted with standard PC and light PC (Figure 3:B). A likely explanation might be the optimal water content. At 5 wt. % GLD addition the molding water content is very near the optimal water content (Figure 3:C) and as discussed in section 3.1, Benson and Trast (1995) found that the optimal HC was reached 1-2 % wet of the optimal water content, suggesting that the mixture here is too dry.

HC is known to be highly dependent on the degree of compaction, with decreasing HC at increasing degrees of compaction (Benson et al. 1994; Watabe et al. 2000; Leroueil et al. 2002). However, this study shows that the dry density after compaction affects the HC in mixtures of silty till and GLD to some extent but does not appear to be the main driving force determining the HC.

### 3.3 The effect of compaction effort on dry density and HC in the till-GLD mixtures

A lower compaction effort was expected to increase the dry density and decrease the HC in the GLD-till mixtures. This as previous studies indicated that an increasing strain on GLD releases the water that is retained by the GLD (Mäkitalo et al. 2015b) and this could lead to a decrease in compaction degree. Evidence of this was visible when mixtures with higher amounts of GLD was compacted. However, no evidence of this was seen in the HC and dry density measurements. Comparing light PC to standard compaction, lower maximum dry densities after compaction was reached when light PC was used in the till-GLD mixtures, 2.03 to 1.81 g/cm³ compared to 2.12 to 1.99 g/cm³ with 5 to 15 wt. % GLD added. The difference increases with increasing amount of GLD added (Figure 3:A; Table 2). For further evaluation of the compaction effort the standard PC, LC, number of compacted layers (three or five), and number of hits with the proctor hammer per layer (5-25 hits) were compared (Figure 4:A). This test showed that the difference between PC and LC with the same number of compacted layers is negligible. The highest maximum dry densities were reached with 25 hits and five layers (Figure 4:A). The decreased maximum dry density after compaction due to a decreased compaction effort was not expected, as indicated by Mäkitalo et al. (2015b). However, Benson and Trast (1995) give evidence for the results shown in this study, i.e. an increased dry density after compaction with increasing compaction effort.

Considering the effect of compaction effort on HC, the hypothesis was that a lower compaction effort might also lead to a lower HC of the till-GLD mixtures, as indicated by the study of Mäkitalo et al. (2015b). However, the results from this study do not agree with this argumentation, as a decreased compaction effort does not significantly affect the HC. A difference in the mixtures where light
Compaction was used compared to standard PC is only seen with an addition of 5 wt. % of GLD1, where the HC is higher than when using standard compaction (4E-08 compared to 2E-08 m/s; Figure 3:A; Table 2). However, despite the lack of positive effects on HC when using a lighter compaction effort, it might still be of consideration, as laboratory tests tend to under-predict HC compared to actual values in the field (Daniel 1984). The compaction effort in the field is usually less than in the lab, leading to increased optimum water-content and a lower HC in a laboratory experiment compared to a field study (Daniel 1984). Therefore, when compacting in the field, the compaction is made dry of optimum water-content instead of at the optimum, resulting in a higher HC than expected (Daniel 1984). Other reasons for underestimating of field HC are desiccation cracks, difficulties getting a representative sample of soil for laboratory test and poor construction (Daniel 1984). The unsaturated properties with a soil in the field is another challenge. An unsaturated soil can vary 10 orders of magnitude in HC, making it difficult to analyze and interpret the results (Fredlund et al. 1994).

In addition to compaction effort, there are several other parameters in the laboratory method that make an estimation of field HC from laboratory test challenging. One example is the compaction method. The dry density is known to affect the HC and the dry density depends on how well compacted the material is. Hand compaction is non-standardized compaction procedure which can lead to differences between samples, and therefore variation in the dry density and HC. Therefore, a proctor compaction method is preferable as it uses the same effort between samples. One downside of the PC method is that the compaction hammer does not reach the sides of the sample cylinder. Therefore, the millimetres closest to the walls are not compacted and there is a risk of preferred wall-flow when measuring HC. Using a hand-operated compaction hammer attached to a frame enables standardization in the compaction effort between the samples and the hammer can reach the sides of the cylinder. The disadvantage with this method is that it is can be challenging to make sure that the whole sample is compacted in the same degree, compared to the PC method with compaction around the sample in a standardized fashion. Another parameter in the HC-method that can affect the outcome is the hydraulic gradient. In geotechnical engineering, the common practice is to apply high hydraulic gradients for quick determination of the HC, which can cause an increase in HC as an increased flow rate at high hydraulic gradients widens the void space by erosion (Al-Taie et al. 2014). High hydraulic gradients can also cause reduction of the HC if the pressure is high enough to erode aggregates of colloidal size from the flow channels (Pusch and Weston 2003; Al-Taie et al. 2014). However, in this study, the hydraulic gradient of 8.7 is comparable to that in field and much lower than 100, that by Al-Taie et al. (2014) was suggested as a limit, or 30 which by ASTM is the recommended hydraulic gradient.

In summary, this study shows that the compaction effort does not seem to be the driving factor controlling HC when working with till-GLD mixtures. However, due to the properties of the GLD and the aim to mimic field conditions, a lighter compaction effort than standard PC is still recommended. Another parameter in the method of measuring HC and its effect on the outcome that needs to be investigated is the compaction method. Especially a comparison between a manual compaction method and the standardized PC-method.
Figure 4. A) Maximum dry density ($\rho_d$) and B) Optimal molding water-content ($w$) after compaction with increasing wt. % GLD added. Changes in max $\rho_d$ (C) and $w$ (D) after compaction with different proctor compaction efforts and hits with the proctor hammer in till-10 wt. % GLD mixture.

For future studies, the oxygen diffusion through till-GLD mixture should be investigated as it together with pH and bacterial activity is the main parameter controlling the oxidation rate in sulfidic mine waste (Akcil and Koldas 2006). As it can be difficult to reliably monitor oxygen in the field, there is a need to study how the different parameters studied in this paper and Nigéus et al (2018) correlate to the oxygen diffusion.

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