Water Retention Capacity as a Measure to Evaluate the Effectiveness of a Green Liquor Dregs-Amended Till to Cover Sulfidic Mine Waste

Susanne Nigéus[1], Christian Maurice[1,2], Jenny Lindblom[1]

[1]*Luleå University of Technology, Department of Civil, Environmental and Natural Resources Engineering, 971 87 Luleå, Sweden, susanne.nigeus@ltu.se*

[2]*Ramböll Sverige AB, Kyrkogatan 2, Box 850, SE-97126 Luleå, Sweden*

Abstract

Hydraulic conductivity is often used as a measure to evaluate the effectiveness of a sealing layer material on top of mine waste. However, the most important soil parameter minimizing the oxygen diffusion to the mine waste is the water retention capacity (WRC) of the sealing layer, as a high saturation corresponds to a low oxygen diffusion rate. This study shows that an amendment of Green Liquor Dregs (GLD), an industrial residue, to a till increases its WRC and has therefore potential to be a more accurate method to evaluate the effectiveness of a sealing layer than hydraulic conductivity.

Keywords: Green Liquor Dregs, Sealing Layer, Water Retention Capacity, Industrial Residue, Recycling, Circular Economy

Introduction

Massive amounts of mine wastes are generated all over the world. Sweden alone generated 104-million-ton mine waste in 2018 (Avfall Sverige 2020). These wastes need to be managed in an environmentally-, technically- and economically sustainable way. About 70% of the mine waste in Sweden contain sulfide minerals (SGU and Swedish EPA 2017) which if left in contact with oxygen and humidity the sulfides oxidize and may produce acid rock drainage (ARD; Saria *et al*. 2006). One way to limit ARD production is to prevent oxygen and moisture to enter the waste (Collin and Rasmuson 1990; Bussière *et al*. 2003; Dagenais *et al*. 2006). This can be done by applying a dry cover. In Sweden this cover usually consists of a sealing layer of a fine-grained material compacted to a high density and a protection layer on top. The availability of a clayey till to construct a sealing layer 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. The use of an industrial residue as an amendment to a local till is therefore highly motivating. The European Union produces 2.5 billion tons of waste each year (Avfall Sverige 2020) and Sweden produced 139 million tons wastes year 2018, excluding mine waste, of which about 2.9 million tons is classified as a non-hazardous waste. Using an industrial residue in a mine remediation program would serve as a large benefit for the environment, for the industry providing the residue, and the mining industry.

In this study Green Liquor Dregs (GLD), a fine-grained residue from pulp production was used as an amendment to a local till. Previous studies on mixing GLD with till have shown promising results for the mixtures to be used as a sealing layer, with low hydraulic conductivity, high water retention capacity (WRC), low oxygen diffusion and increased compaction degree (Hargelius 2008; Mäkitalo *et al*. 2015a and b; Virolainen *et al* 2020). Hydraulic conductivity is often used as a measure to evaluate the effectiveness of a sealing layer material. However, the most important parameter minimizing the oxygen diffusion to the mine waste is the water retention capacity of the material as a high saturation corresponds to a low oxygen diffusion rate. In general, the oxygen flux rates become negigble when the degree of saturation is greater than 85-90%, this since the air phase pathways becomes discontinuous (Corey 1957) and the oxygen is then transported through the water phase (Aubertin and Mbonimpa 2001; Aachib *et al*. 2004). One way to estimate the water retention capacity is to use a soil-water characteristic curve (SWCC), which describes the water content (w) or the degree of saturation (S_r) retained in the soil as a function of the matric suction (ψ; Sillers *et al.* 2001). The matric suction required to begin draining a fully saturated soil is called the air-entry suction (ψ_a). The residual water content (θ_r) is defined as the amount of water in the soil that cannot be removed even at great suction heads, due to adhesive forces between the water molecules and the soil particles, or to entrapment in disconnected pores. Factors controlling the shape of the curve are mainly the type of the soil (Fredlund and Rahardjo 1993; Tinjum *et al* 1997; Sillers *et al*. 2001), but also the molding water content is an important factor (Vanapalli *et al*. 1996; Tinjum *et al*. 1997; Vanapalli *et al*. 1999).

In this study 5 to 20 wt.% of GLD from Smurfit Kappa paper mill, Sweden was mixed with three tills with different particle size distributions. A maximum 20% of addition of GLD was set as a limit in this study, as a mixture with more GLD was shown by Mäkitalo *et al* (2015b) to be difficult to compact and handle, due to increased water content above the optimum molding water content and due to decreased shear strength. The addition of GLD to tills was expected to increase the WRC of the tills the more GLD was added. The objectives of the study was to find out (i) if GLD can improve the WRC of till with the purpose of using the mixture in a sealing layer on top of mine waste, and (ii) if WRC can be used as a measure to evaluate the effectiveness of that sealing layer.

Methods and materials

Three different tills were used in this study, a sandy till (SaT) and two silty tills with different clay contents (SiT1 and SiT2; Table 1). The silty till with a lower clay content is named SiT1 and the silty till with higher clay content SiT2. SiT1 was collected at a till quarry at the Brännkläppen facility in Boden, Sweden, SiT2 from another till quarry outside Boden, Sweden and the sandy till from Sunderbyn outside Luleå, Sweden. The GLD used in the study derived from Smurfit Kappa paper mill in Piteå, Sweden and was collected in sealed plastic containers to preserve the water content of the material (TSC 43%; Table 1).

The tills were air dried and sieved through a 20-mm sieve and particles above 20 mm were removed, which is praxis in a material that is to be used in a sealing layer. Particles above 20 mm will affect negatively on the effectiveness of materials used as a sealing layer. 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 (TSC 43%; Table 1) as it has been shown by previous unpublished studies to be difficult to rewet without affecting its physical properties. The mixing was carried out by hand with a small shovel until the mixture was estimated as homogenized.

To obtain particle size distribution (PSD) the tills were washed and dry sieved according to SS-EN 933-1:2012. A mechanical sieve tower (Retsch AS 200) with an amplitude of 2.2 mm/"g" and cut-off sizes 12.5, 10, 8, 5, 4,

Table 1 *The total solid content (TSC), particle density, sand-, fines- and clay contents in the mixtures. N= number of analyzed samples.*

Material	TSC (%)	Particle density (g/cm³)	Sand (<2 000 µm)	Fines (<63 µm)	Clays (<2 µm)
SaT	91.7±0.4 (N=9)	2.68	73%	14±1% (N=3)	0.7% (N=1)
SiT1	91.5±0.4 (N=9)	2.71	49%	34±5% (N=9)	2.6% (N=1)
SiT2	91.1±0.8 (N=6)	2.70	54%	35±1% (N=2)	4.3% (N=1)
GLD	43±4 (N=12)	2.63	24±29% (N=3)	76±29% (N=3)	5.4±4.1% (N=3)

2, 1, 0.5, 0.25, 0.125 and 0.063 mm was used. PSD for the fines (<0.063 mm) were done by laser diffraction analysis on triplicate samples of each material by a CILAS Granulometer 1064 (CILAS, Orléans, France). The PSD was then calculated using the CILAS software (Figure 1; Table 1). The total solid content (TSC) was performed according to the SIS standard SS-EN 14346:2007 and the particle density of the materials was determined using an AccuPyc II 1340 Pycnometer.

Water retention capacity was measured with a pressure plate test using a 1500F1 Pressure Plate Extractor. The air-dried till used in the mixtures to be tested was wetted to reach a TSC of approximately 91%. Specimens were compacted by hand with a metallic compactor (2 kg) and with 25 blows in five layers of height with the goal to reach an even compaction between the samples and simulate standard proctor compaction. When compaction was done, the samples were levelled off with a knife and weighed. The samples were placed on saturated ceramic plates with 15 bar air entry value, which base was connected to the atmosphere. Gas pressure was applied in 12 increasing steps, up to a pressure of 145 mH_2O to the samples. One sample was taken out after 48 h for each pressure step and characterized for its water content according to standard SS-EN 14346:2007. Water retention curves were described using van Genuchten model (Van Genuchten 1980).

Results and discussion

The hypothesis was that the addition of GLD would improve the WRC of those tills that alone would not reach the requirement of a sealing layer in Sweden (hydraulic conductivity $<10^{-8}$ 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 2; Table 2). At 85% S_r the matric suction increased from 3.2 to 6 mH_2O with 15 wt.% GLD addition to the silty till 1 (Figure 2:A-D; Table 2), 3.3 to 26 mH_2O in the sandy till (SaT) mixtures (Figure 2:E-H; Table 2) and from 3.6 to 50 mH_2O in the silty till 2-mixtures (Figure 2:I-L; Table 2). The more GLD that is added the higher WRC is achieved in the mixtures. To minimize oxygen diffusion to the mine waste the sealing layer should be kept at a saturation degree of at least 85% (Corey 1957, Aubertin and Mbonimpa 2001; Aachib *et al.* 2004; Virolainen *et al.* 2020) and with 15 wt.% GLD addition to the tills a high saturation is kept even at high suction (Figure 2).

This study indicates an increase in WRC the more GLD is added to the tills (i.e. increasing ψ_{85}; Table 2). However, due to compaction difficulties as a result of a water content above the optimum molding water content, more than 20 wt.% is not recommended if the GLD is not dried before mixing it with till.

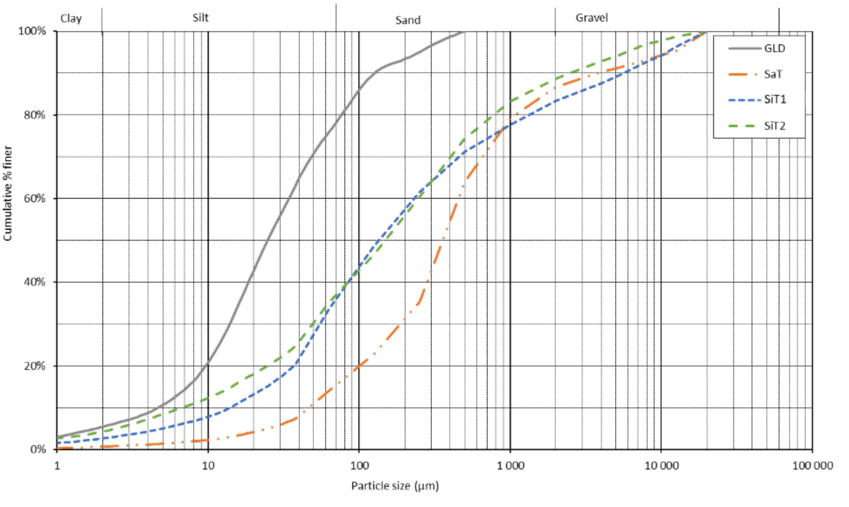

Figure 1 *Particle size distribution curves (PSD) of the three different tills (SaT, SiT1 and SiT2) and the GLD.*

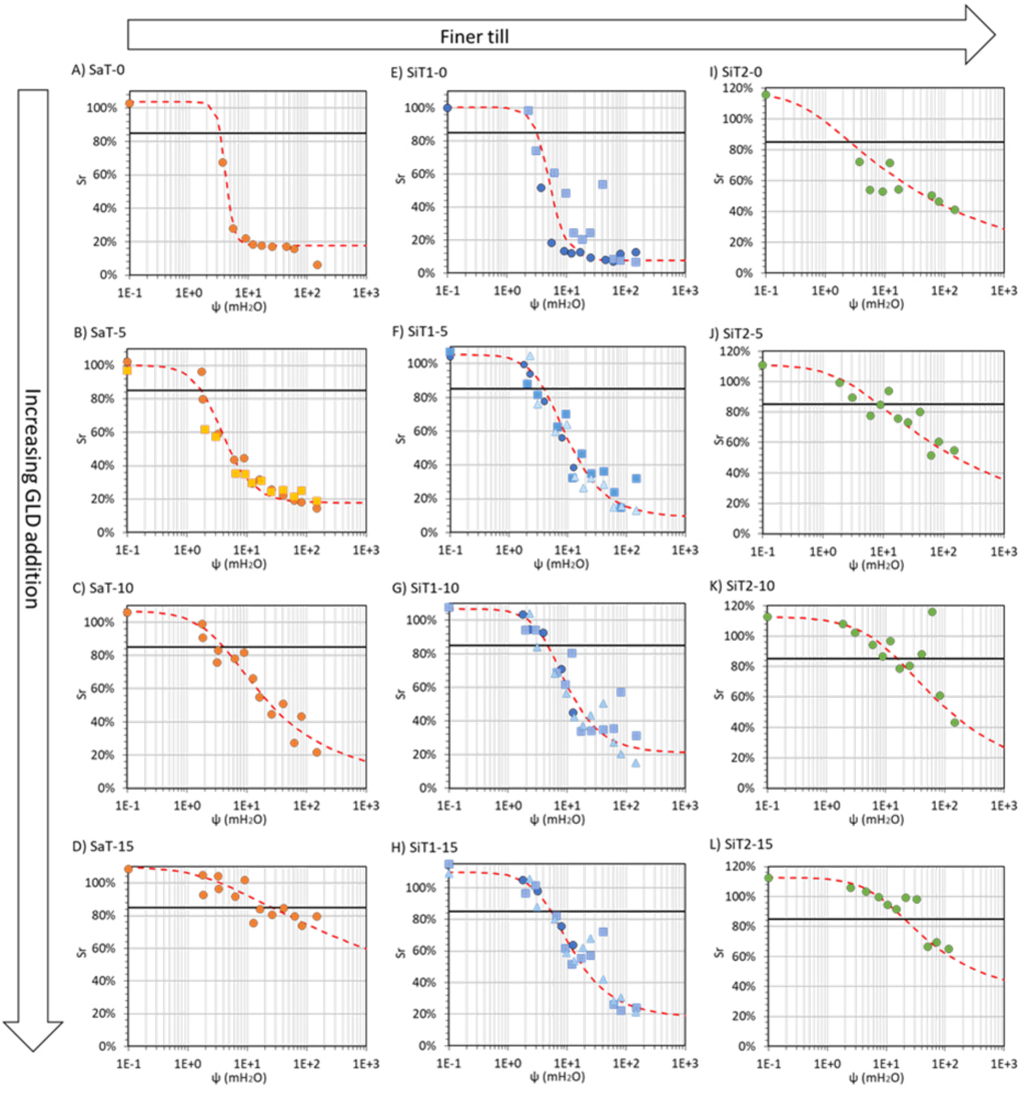

Figure 2 WRC in the mixtures of GLD and sandy till (SaT; A-D), silty till 1 (SiT1; E-H), and silty till 2 (SiT2; I-L). On the x-axis is the matric suction ψ (mH$_2$O) on the y-axis the water saturation. Each mixture has been run 1-3 times and each run is marked with a different geometric figure in the graph. The red line is from the van Genuchten equation, best fit, and the black line represent 85% water saturation. 0 m suction is in the graphs 0.1 m suction due to the logarithmic scale

According to a study conducted by Jia *et al.* (2019), GLD could be dried without affecting its physical properties after rewetting, regarding hydraulic conductivity and WRC. However, this needs further investigations because it is difficult to achieve in field and should be done before transportation to the mine site. This is however worth consideration as the residue becomes more attractive to the mining industry both in view of transportation costs, due to the high water content of the GLD leading to a higher weight than when the material is dried, but also due to the increased effectiveness of the sealing layer with higher amounts of GLD in the mixtures. A previous study by Mäkitalo *et al.* (2016) has shown that GLD in its own lacks long term stability in slopes due to its low shear strength, indicating that the GLD amendment should not be too high. The

Table 2 *Key parameters in the SWCC; Van Genuchten parameters (m; n; α), matric suction for 85% S_r (ψ85), air entry suction (ψ_a), residual suction (ψ_r), residual water content (θ_r), and saturated water content (θ_s).*

	ψ_{85}	ψ_a	ψ_r	θ_r	θ_s	n	m	α
SaT-0	3.4	2.8	6.3	0.06	0.35	6.5	0.85	0.25
SaT-5	1.7	1.2	14	0.06	0.338	1.9	0.80	0.3
SaT-10	4.1	1.5		0.02	0.36	1.4	0.29	0.3
SaT-15	25	0.9		0.005	0.37	1.1	0.09	0.5
SiT1-0	3.2	2.5	10	0.025	0.34	3.5	0.71	0.22
SiT1-5	4	2	45	0.03	0.36	1.9	0.47	0.2
SiT1-10	4.7	2.3	40	0.07	0.36	2	0.50	0.19
SiT1-15	5.3	2.5	50	0.06	0.37	1.8	0.44	0.19
SiT2-0	2.6			0.09	0.34	1.5	0.33	0.9
SiT2-5	8.4			0.01	0.34	1.28	0.22	0.1
SiT2-10	15.7			0.00001	0.34	1.37	0.27	0.04
SiT2-15	20			0.005	0.34	3	0.67	0.012

hysteretic effect is also a parameter that is worth taking in consideration, as it might impact the interpretation of the SWCC and the actual WRC of the mixtures studied. The hysteretic effect will cause greater matric suction for drying than for wetting for the same water content (Tinjum *et al.* 1997), and the ψ_{85} will thus be higher in field than the laboratory experiments indicates, i.e. the required 85% S_r will be held for greater suction giving a higher WRC.

Conclusion

An amendment of GLD to a till that alone would not meet the commonly used requirements for a sealing layer in Sweden, i.e., hydraulic conductivity $<10^{-8}$ m/s, was shown to increase the WRC of the material and can keep a high saturation (>80%) even with high matric suction (up to 25 mH_2O; SaT-20, Table 2). This suggest that GLD can beneficially be used in a sealing layer in mine site remediation. Even if the hydraulic conductivity does not reach the requirements for a sealing layer with GLD addition (results from a previous study by Nigéus *et al.* 2018), the water retention capacity increases the more GLD is added to the till. Usually hydraulic conductivity is used as a parameter to determine the quality of the sealing layer in Sweden, however this study shows that the water retention capacity is an important factor preventing oxygen diffusion to the mine waste by keeping the sealing layer saturated. It can therefore be said to have potential to be a more accurate method to evaluate the effectiveness of a sealing layer. In future studies it would be of interest to connect WRC with actual results from oxygen diffusion, to predict oxygen diffusion through a sealing layer and also to be able to retrieve a regulation value for WRC, i.e. ψ_{85}, in a sealing layer material, as there is for hydraulic conductivity in Sweden. Recycling of GLD in mine remediation is a procedure to strive for as it is beneficial for not only the mining company, but also the pulp and paper industry as it minimizes the waste landfilled. The use of an industrial waste as a product is however challenging as there is a great variability in its chemical compounds and physical properties. GLD varies both between industries, but also between batches, within the same industry (Nigéus *et al.* 2018). This creates a challenge when the waste is to be used as a product, as it is difficult to predict its chemical and physical properties which in turn affects its effectiveness.

Acknowledgements

The study is financed by Boliden Mineral AB, Vinnova, the Swedish Energy Agency, the Swedish Research Council Formas and Mistra's program "Closing the loop" (project GLAD) and the European Union's Horizon 2020 research and innovation program under agreement № 730305 (project Paperchain). SP Processum, Ragn-Sells AB, Ramböll Sverige AB, Ecoloop AB and the Centre of Advanced Mining and Metallurgy (CAMM) at Luleå University of Technology are gratefully acknowledged. Smurfit Kappa Kraftliner paper mill is acknowledged for providing Green liquor dregs. Special appreciations are dedicated to Dr. Thomas Pabst at Polytechnique Montréal, Canada for knowledge and support regarding WRC measurements. Together with Johan Sandberg, Jenny Palmenäs, and Ida Kronsell former students at the Luleå University of Technology, for help with some of the laboratory work.

References

Aachib, M., Mbonimpa, M., Aubertin, M. (2004). "Measurement and Prediction of the Oxygen Diffusion Coefficient in Unsaturated Media, with Applications to Soil Covers." Water Air Soil Pollut., 156(1), 163-193.

Aubertin, M., and Mbonimpa, M. (2001). "Diffusion of Oxygen through a Pulp and Paper Residue Barrier: Discussion." Canadian Geotechnical Journal, 38(3), 658-660.

Avfall Sverige. (2020). Swedish Waste Management 2020. Avfall Sverige.

Bussière, B., Aubertin, M., Chapuis, R. P. (2003). "The Behavior of Inclined Covers used as Oxygen Barriers." Canadian Geotechnical Journal, 40(3), 512-535.

Collin M., Rasmuson A. (1990). "Mathematical modeling of water and oxygen transport in layered soil covers for deposits of pyritic mine tailings." In Proceeding of the GAC-MAC Annual Meeting: Acid Mine Drainage Designing for Closure, Vancouver, BC, Canada, pp 311-333.

Corey, A. T. (1957). "Measurement of Water and Air Permeability in Unsaturated Soil 1." Soil Sci. Soc. Am. J., 21(1), 7-10.

Dagenais, A., Aubertin, M., Bussière, B., Cyr, J. (2006). "Performance of the Lorraine Mine Site Cover to Limit Oxygen Migration." Transactions-Society for Mining Metallurgy and Exploration Incorporated, 318, 190.

Fredlund, D. G., and Rahardjo, H. (1993). Soil Mechanics for Unsaturated Soils, John Wiley & Sons.

Hargelius, K. (2008). In Swedish: "Pilotyta med tätskikt på Ätrans deponi, Fältförsök-Värö-FAVRAB-Hylte." Ramböll Sverige AB, Region Väst, Gothenburg.

Jia, Y., Hamberg, R., Quereshi, A., Mäkitalo, M., and Maurice, C. (2019). "Variation of green liquor dregs from different pulp and paper mills for use in mine waste remediation". Environmental Science and Pollution Research, 26: 31284-31300.

Mäkitalo, M., Maurice, C., Jia, Y., Öhlander, B. (2014). "Characterization of Green Liquor Dregs, Potentially Useful for Prevention of the Formation of Acid Rock Drainage." Minerals, 4(2), 330-344.

Mäkitalo, M., Stenman, D., Ikumapayi, F., Maurice, C., Öhlander, B. (2015a). "An Evaluation of using various Admixtures of Green Liquor Dregs, a Residual Product, as a Sealing Layer on Reactive Mine Tailings." Mine Water and the Environment.

Mäkitalo, M., Macsik, J., Maurice, C., Öhlander, B. (2015b). "Improving Properties of Sealing Layer made of Till by Adding Green Liquor Dregs to Reduce Oxidation of Sulfidic Mine Waste. " Geotech. Geol. Eng., 33(4), 1047-1054.

Mäkitalo, M., Lu, J., Maurice, C., Öhlander, B. (2016). "Prediction of the Long-Term Performance of Green Liquor Dregs as a Sealing Layer to Prevent the Formation of Acid Mine Drainage." J. Environ. Chem. Eng., 4(2), 2121-2127.

Nigéus, S., Maurice, C. (2018). "Green liquor Dregs-amended till to cover sulfidic mine waste". Lic. Thesis: Luleå University of Technology.

Saria, L., Shimaoka, T., Miyawaki, K. (2006). "Leaching of Heavy Metals in Acid Mine Drainage." Waste Manag. Res., 24(2), 134-140.

SGI. (2003). In Swedish: Inventering Av Restprodukter Som Kan Utgöra Ersättningsmaterial För Naturgrus Och Bergkross i Anläggningsbyggande, SGI.

SGU, and Swedish EPA. (2017). Förslag Till Strategi För Hantering Av Gruvavfall. Redovisning Av Ett Regeringsuppdrag.

Sillers, W. S., Fredlund, D. G., Zakerzadeh, N. (2001). "Mathematical Attributes of some Soil—water Characteristic Curve Models." Unsaturated Soil Concepts and their Application in Geotechnical Practice, Springer, 243-283.

Tinjum, J. M., Benson, C. H., Blotz, L. R. (1997). "Soil-Water Characteristic Curves for Compacted Clays." J. Geotech. Geoenviron. Eng., 123(11), 1060-1069.

Van Genuchten, M. T. (1980). "A Closed-Form Equation for Predicting the Hydraulic Conductivity of Unsaturated Soils 1." Soil Sci. Soc. Am. J., 44(5), 892-898.

Vanapalli, S. K., Fredlund, D. G., Pufahl, D. E. (1996). "The Relationship between the Soil-Water Characteristic Curve and the Unsaturated Shear Strength of a Compacted Glacial Till." Geotech Test J, 19(3), 259-268.

Vanapalli, S., Fredlund, D., Pufahl, D. (1999). "The Influence of Soil Structure and Stress History on the Soil-Water Characteristics of a Compacted Till.". Geotechnique, 49 (2), 143-159.

Virolainen, A., Maurice, C., and Pabst, T. (2020). Effective Oxygen Diffusion coefficient of Till and Green Liquor Dregs (GLD) Mixes Used in Sealing Layer in Mine Waste Covers. Water Air Soil Pollut, 231:60.