

Amelioration of permeable soil with green liquor dregs for the construction of sealing layers for mine waste storage facilities

Ida Kronsell¹, Susanne Nigéus¹, Anna Virolainen², Yu Jia³, Thomas Pabst⁴, and Christian Marurice¹

¹Luleå University of Technology, 87187 Luleå, Sweden

²Boliden Mineral, Boliden, Sweden

³Department of Environment and Mineral Resources, Greenland Institute of Natural Resources, 3900 Nuuk, Greenland

⁴Research Institute on Mines and the Environment (RIME), Polytechnique Montreal, Canada

Abstract. Mining of sulfidic ore generates acidic waste which often leads to the generation of acid rock drainage (ARD) having an adverse impact on aquatic flora and fauna. Engineered multilayer cover is one of the commonly used methods to prevent oxygen from being transported to mine waste. The sealing layer has a key function in the cover, thanks to its high water retention capacity and a low hydraulic conductivity, which enable the layer to remain near water saturation, effectively hindering oxygen diffusion. When adequate material is not available improvement of local soil material with a sealing agent is principally the only option to produce sealing layer material. Apart from bentonite clay, Green Liquor Dregs (GLD), a waste from the pulp and paper industry, have the ability to reduce the oxygen diffusion when mixed to granular soil, improving the water retention capacity of the blend. Experience from the development of Till/GLD-blends for the construction of sealing layers is compiled to address issue related to soil amelioration using waste with focus on material variation and quality control. The presentation focuses on the opportunities and challenges for the establishment of a circular system based on reutilization of a waste, herein GLD for improvement of soil's geotechnical property.

1 Introduction

The global annual generation of mine wastes has been estimated to 15.000 – 20.000 million tons of solid waste [1], the majority of which containing residual sulfidic minerals (primarily pyrite, FeS₂). Similar waste can also be found in many small abandoned mines. Weathering of minerals is a natural process which increases in intensity as new surfaces are created during the mining process. In contact with air and water, sulfide containing waste often leads to the generation of acid rock drainage (ARD), an acidic leachate with high concentrations of metals and metalloids [2], which is a threat to environment [3-4].

Engineered cover is one of the common mine remediation methods used to control oxygen transport to mine waste and prevent oxidation at the source [5]. Engineered covers are typically multi-layer constructions which design depends on waste properties, cover material availability, water table position and climatic conditions [6-7]. In Sweden, over 90% of the active mines plan to use engineered covers for the remediation of the waste rock piles [8].

The sealing layer has a key function and consists of compacted fine-grained material. The sealing material should have a high water retention capacity (WRC) to avoid dehydration (cracking) and a low hydraulic conductivity (HC), to maintain the sealing layer to remain near water saturation, effectively hindering

oxygen diffusion. On top of the sealing layer, an uncompacted protection layer can be placed to protect the sealing layer from frost, dehydration (evaporation), root penetration and it can act as a water storage buffer. A nutrient-rich vegetation layer is placed on the top to prevent erosion (Fig. 1).

In boreal area, till is the most common type of soil available. Till is a glacial sedimentary soil type containing particles of various sizes from boulders to clay. In the most favorable scenarios, clayey till is available close to the mine and can be used to construct the sealing layer. When adequate material is not available, improvement of local soil material with a sealing agent can be used produce the necessary amount of sealing layer, commonly 1 ton/m². Bentonite clay is one possible amendment to reduce the hydraulic conductivity of soil. However, bentonite production is costly both economically and environmentally.

Recycling of industrial wastes as a sealing layer material is another option, which could be beneficial for both the mining industry and the industry providing the leading to less waste disposal and enhancing a circular economy approach. Mine wastes are for example often used for reclamation: non acid-generating or desulfurized tailings can be used as moisture retaining layers [9-11], and waste rock can be used for capillary break layers [12-13].

* Corresponding author: ida.kronsell@ltu.se

During the last decade, research on green liquor dregs (GLD), a waste from the pulp and paper industry, indicate they could efficiently reduce the oxygen diffusion in soil that are too permeable. The practice of using GLD is still rather novel and previous studies have focused on the chemical, physical and mineralogical characterization of GLD [14-16], various mixtures containing GLD [16-20] and construction and evaluation of engineered covers containing GLD in field tests and pilot scale studies [22-23].

GLDs have a low HC and high WRC but also have a paste like consistency and low shear strength. Mixed in a limited amount with granular soil such as till, an increasing addition of GLDs have the capacity to improve the WRC of the mixture, while keeping relatively good geotechnical properties. The success of the methods relies on an efficient mixing of the GLD in the matrix and a sufficient degree of compaction of the blend. Both mixing and packing are therefore essential to obtain a functioning barrier at full scale.

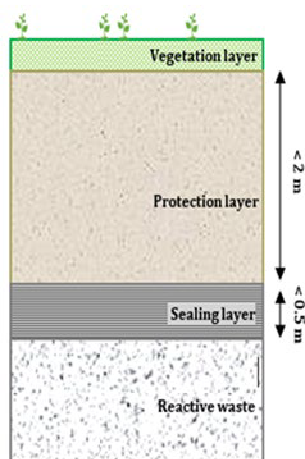


Fig. 1. Schematic illustration of a multilayer engineered cover consisting of a sealing layer, a protective layer and a surficial vegetation layer.

Even if GLD have attractive properties for the construction of sealing layers, GLD cannot be used solely in a 50-cm thick layer because of limited shear strength [17]. Therefore, R&D work has focused on mixing till and GLD in order to produce a material that can be used in slopes and has a low hydraulic conductivity, is able to withstand drying and remain humid to reduce oxygen transport.

Laboratory results indicate that high amendment levels are beneficial but the construction of sealing layer is complicated at amendment levels above 15-20%, as the mixture becomes difficult to handle and compact. Demonstration projects performed in 2014 and 2016 showed that Till/GLD mixture can be produced, placed, and compacted with conventional equipment, at a mine scale i.e. several hectares up to square kilometer. The experiments were done at Boden waste facility and Näsliden former copper mine, Sweden. The main challenges using Till/GLD were associated with the variability of the GLD between different paper mills, but also with time for the same paper mill [14]. Also till

varies regarding water content and particle size distribution.

The key factors for construction of sealing layer are:

- Quality control of the incoming materials (GLD and till)
- The mixing quality that could be achieved and the production rate
- Compaction degree that can be achieved in slopes and flat areas.

2 GLD as an amendment for sealing layer construction

Previous studies have shown that GLD has properties suitable for a sealing layer i.e. it is fine-grained ($< 63\mu\text{m}$), commonly has an HC in the range of 10^{-8} and 10^{-9} m/s and a higher WRC compared to materials with similar particle size, such as clayey/sandy silt [14].

GLDs are classified as a non-hazardous chemical waste by the Swedish EPA [24] and the main solid compounds of GLD consist of CaCO_3 , $\text{Mg}(\text{OH})_2$, C and metal sulfides [18, 25]. The liquid phase of the GLD consists of Na_2CO_3 and NaOH, which generates its characteristic high pH (10-11). Other characteristics of GLD are high porosity (73 - 82 %), a bulk density of $0.44\text{-}0.67 \text{ g/m}^3$, a specific density of 2.47 to 2.60 g/cm^3 [14].

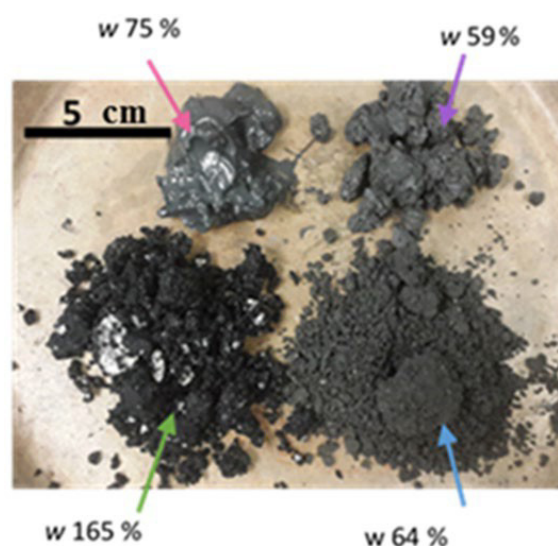


Fig. 2. Green liquor dregs samples from four paper mills having different water content, colour and texture (after [26]).

The aspect of GLDs varies greatly among sources (Fig. 2) but also with time for the same paper mill [14].

The natural water content is in the range 80-160 % (total solids content, TSC, 38-55 %), often close to the liquid limit (wL), i.e. the water content at which the material texture transforms from plastic into liquid, which makes them difficult to handle. The clay content varies between 3 and 28 %, the plastic limit (wP) between 46 and 170 %, wL between 60 and 330 %. Intensive mixing of GLD results in transformation from plastic to liquid consistency. The TSC of the GLD, the

wP and the wL are factors influencing the compaction properties, HC, WRC and finally also the oxygen deterring function of a sealing layer [27] (Mácsik et al., 2017).

The HC of GLD decreases with increasing compaction degree [19-20] (Mäkitalo et al., 2015a and b).

2.1 Key properties of GLD in sealing layers

Key properties in a sealing layer are; Low oxygen diffusion, low HC and high WRC.

Oxygen transport in soil occurs through a combination of advective and diffusive processes in both the gas and aqueous phase of the porosity. At a relatively low degree of saturation, most oxygen transport occurs through partially air-filled pores [28], because the diffusion coefficient (D_e) in air is 10 000 times greater than in water [29]. In a layer that is kept close to saturation, the D_e in the soil can be comparable to the D_e in water, reducing the oxygen flux to a level comparable to that of a water cover [28-29].

HC is used as criteria for the effectiveness of a sealing layer, the requirements being for example commonly set to $\leq 10^{-8}$ m/s in Sweden. Adding GLD to till changes the particle size distribution of the soil, increasing the amount of fines compared to the original material. A higher amount of fines together with high compaction reduces the macro porosity and the HC. Results show that the D_e for different till/GLD-mixtures varied greatly for the blends ($10^{-6} > D_e > 10^{-11}$ m² s⁻¹ 26) depending on the degree of saturation. Even though

the GLD contain substantial amounts of water, a high water content of the till was still required to reach a low D_e .

Water retention is a key feature to be considered as high water retention capacity usually corresponds to a high water-saturation (S_r) and low D_e . The typical soil-water characteristic curve (SWCC) consist of three stages: capillary saturation, desaturation and residual saturation (Fig. 3:a; [30]). The matric suction required to remove water from the largest pores corresponds to the air-entry value (ψ_a ; Fig. 3a). The water content corresponding to the asymptote of the SWCC at low degrees of saturation is called the residual water content (θ_r ; Fig. 3:a). The SWCC is hysteretic, which means that matric suction for desorption (drying) is higher than for sorption (wetting), for a given water content (Fig. 3:b; [31]). The shape of the SWCC varies between soil types [32]. Soils with smaller pores and finer particles usually have a higher ψ_a (Fig. 3:c; [30-31]). When fine-grained soils and found that if they are compacted dry of optimum water content, steeper SWCC are generated compared to those compacted wet of optimum water content [33].

The molding water content of the material [35-36], i.e. the water content after compaction, affects the HC. Laboratory tests on different till/GLD-mixtures showed that the initial water content of the materials is controlling the final HC of the mixtures [21]. When using material drier than 1-2% wet of the optimum molding water content more GLD should be added to the mixture to reach optimum HC, as the optimum water content is reached at a higher wt. ([21]; Fig. 4).

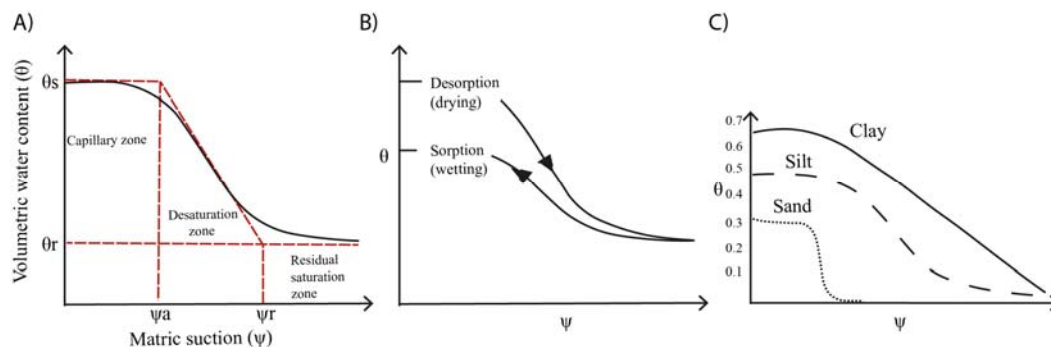


Fig.3. A) A typical SWCC, where ψ_a is the air-entry suction, ψ_r the water entry value, θ_s the nearly saturated water content and θ_r the residual water content (modified from [31]). B) A typical SWCC for desorption (drying) and sorption (wetting; modified from [34]). C) A typical SWCC for clay, silt and sand (modified from [30]).

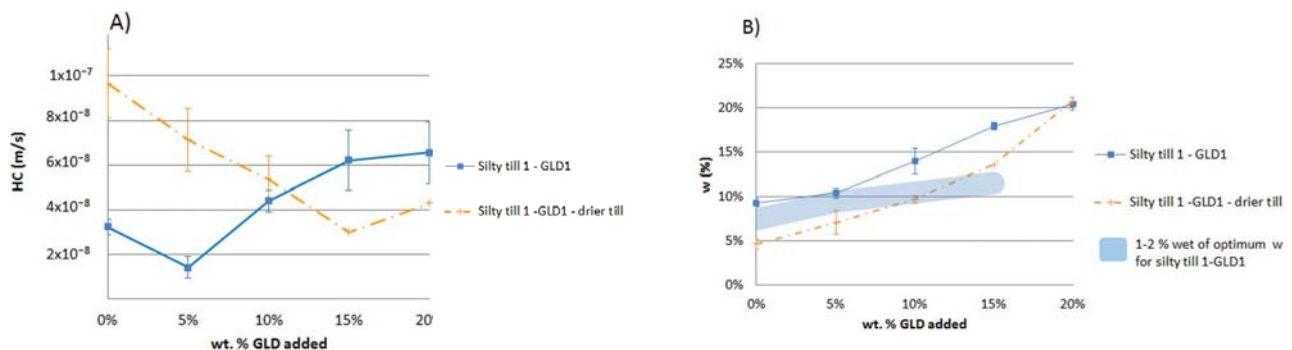


Fig. 4. A) Hydraulic conductivity (HC) and B) molding water content (w) in different till/GLD mixtures.

2.2 Construction of Mine Waste Cover - Experience from pilot tests

The production of the gld-blend should be based on a site-specific mixing ratio defined in laboratory tests before applying at large scale. commonly, the gld amendment varied between 5-10 % based on dry weight basis and a tolerable span has to be defined. Quality control of the material as well as of the mixture is necessary during the whole production.

While till is extracted close to the site GLD are transported from the pulp mill. For mine remediation, a major part of a mill annual generation is needed, during the few months available for the remediation work. GLD are generated evenly all year around. Transport and storage are therefore necessary to provide enough material when the remediation starts. GLD should be controlled at the source regarding water content. It is important to ensure that GLD do not deteriorate during transport and storage i.e. the GLD are not exposed to humidity.

Levelling of the surface: The Waste rock is a coarse-grained fraction containing boulders resulting in large pores in the pile. To prevent material migration into the pores (suffusion), the surface of the waste rock pile must be levelled with an adequate grain sized material and the slopes are shaped between at maximum 1:3 to ensure stability and 1:10 to divert precipitation [37]. Compaction of the levelling layer surface is performed with a roller to reach a dry density above 1.9 t/m^3 .

Production of Till/GLD mixture: Different types of mixing equipment exist. For the pilot experiment, an ALLU shovel DH 4-17-X75 was used with success (Fig. 5A). Important factors when selecting equipment are the ability to produce a homogeneous mixture and to have sufficient mixing capacity. Pre-mixing using wheel loader is needed in order to dose the amendment and obtain a homogeneous mixture quality using front shovel- and bucket mixer.

Construction of the sealing layer: In accordance with the control program, the surface should be controlled regarding levels, compaction and water ratio. The sealing layer is commonly at least 30-50 cm thick, and the pilot test showed that application and compaction in one single layer is sufficient to reach the criteria. A 60-70 cm layer of Till/GLD layer was leveled with a bulldozer and compacted with a roller (Fig. 5B). A project specific lowest acceptable compaction degree should be determined in advance (e.g. at least six overpasses by vibrating compaction roller in the pilot in Näsliden, Västerbotten, Sweden) and the compaction degree is regularly controlled by e.g. water volumeter (surface), nuclear density gauge (e.g. Troxler) or electrical impedance spectroscopy (e.g. SDG). Compaction resulted in an increased dry density from $1.5\text{-}1.6 \text{ tons/m}^3$ up to approximately $1.7\text{-}2 \text{ tons/m}^3$. The

number of overpasses by roller might need to be adjusted after control of the dry density.

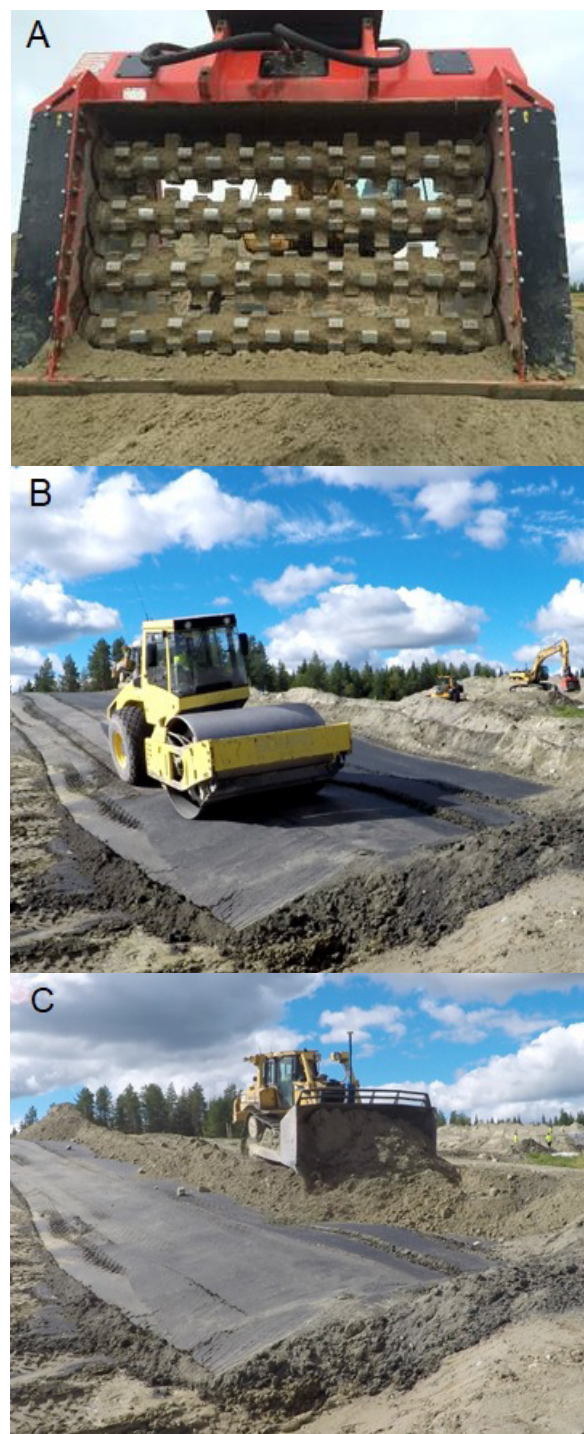


Fig. 5. A) ALLU shovel DH 4-17-X75. B) Compaction of the sealing layer with roller. C) Application of the protective layer with a bulldozer. Näsliden remediation site, Västerbotten, Sweden. Photo: Josef Mácsik.

3 Conclusion

GLD can be used as amendment in coarse soil such as till in order to increase the WRC and decrease the HC of the sealing layer in engineered covers for mine waste deposits. GLD are waste and consequently present a

* Corresponding author: ida.kronsell@ltu.se

large variability regarding physical and chemical properties e.g. water content and particle size distribution, which affects the properties of the GLD/till-blends. Site-specific recipe taking into account the GLD and the till to be used is necessary. The recipe includes the proportions of respective material, the number of mixing steps and overpasses by vibrating roller.

Quality control all along the production process is necessary to ensure the sealing layer meets the requirements for an oxygen barrier. The key factors for the quality of sealing layer are the control of: The incoming materials, both GLD and till, regarding e.g. water content; The homogeneity of the blend obtained from the mixer; The compaction degree in both slopes and the plateau.

Acknowledgements

The research presented was financed by Processum, Mistra's program "Closing the loop" (project GLAD), the European Union's Horizon 2020 research and innovation program under grant agreement N° 730305 (project Paperchain) and GEORES, a Marie Skłodowska-Curie Research and Innovation Staff Exchange (RISE) project funded from the H2020 Programme (H2020-MSCA-RISE-2017) of the EC under grant agreement n. 778120.

References

1. B. Lottermoser, 3rd ed. Springer-Verlag, Berlin Heidelberg (2010)
2. D.W. Blowes, C.J. Ptacek, J.L. Jambor, C.G. Weisener, D. Paktunc, W.D. Gould, D.B. Johnson. In: *Treatise on Geochemistry* (Second Edition), Holland and Turekian (Eds), **11**, 131–190.
3. A. Akcil, S. Koldas. *J Clean Prod.*, **14**,1139–1145 (2006)
4. L. Saria, T. Shimaoka, K. Miyawaki. *Waste Manag. Res.* **24**,134-140 (2006)
5. M. Aubertin, B. Bussière, T. Pabst, M. James, M. Mbonimpa. (2016). *Proc. Geo-Chicago: Sust. Energy and the Geoenviron.*, Chicago, August 14-18 (2016)
6. M. Aubertin, B. Bussière, L. Bernier. *Envir. et Gestion des Rejets Miniers*. Published by Presses Polytechnique Internationales (2002)
7. T. Pabst, T. B. Bussière, A. Aubertin, J. J. of Cont. Hydro. **214**, 39-53 (2018)
8. Statens offentliga utredningar. SOU 2018:59 (2018)
9. M. Aubertin, B. Bussière, M. Monzon, A.M. Joanes, D. Gagnon, J.M. Barbera, M. Aachib, C. Bédard, R.P Chapuis, L. Bernier. *Rapport de Recherche, Projet CDT P1899. NEDEM/MEND 2.22.2c* (1999)
10. B. Bussière, M. Benzaazoua, M. Aubertin, M. Mbonimpa. *A laboratory study of covers* (2004)
11. I. Demers, B. Bussière, M. Mbonimpa, M. Benzaazoua, M. Canadian *Geotech. J.* **46**, 454–469. (2009)
12. A. Kalonji Kabambi, B. Bussière, I. Demers. *Geotech. and Geo. Engr. J.* **35** (3): 1199-1220. (2017)
13. C.G. Larochelle, B. Bussière, T. Pabst. *Water Air Soil Pollut*, **230**: 57. <https://doi.org/10.1007/s11270-019-4114-0> (2019)
14. M. Mäkitalo, Y Jia, C. Maurice, B. Öhlander. *Minerals* **4**, 330-344 (2014)
15. M. Mäkitalo, J. Lu, C. Maurice, B. Öhlander. *J. of Envir. Chem. Engr.* **4** (2): 2121–2127 (2016)
16. F.M Martins, J.M Martins, L. Ferracin, C.J. da Cunha. *J. of Haz. Mate.* **147**, 610-617. (2007)
17. Y. Jia, D. Stenman, M. Mäkitalo, C. Maurice, B. Öhlander. *Use of amended tailings as mine waste cover. Waste and Biomass Valori.* **4** (4) :708–718 (2013)
18. Y. Jia, C. Maurice, B. Öhlander. *Envir. Earth Sci.* **72**, 319–334 (2014)
19. M. Mäkitalo, J. Mácsik, C. Maurice, B. Öhlander. (2015a) *J. of Geotech. and Geo. Engr.* **33** (4):1047-1054 (2015a)
20. M. Mäkitalo, D. Stenman, F. Ikumpayi, C. Maurice, B. Öhlander. *Mine Water and the Envir.* **35** (3):283-293 (2015b)
21. S. Nigéus. Lic. thesis. Luleå University of Technology (2018)
22. A. Chtaini, A. Bellaloui, G. Ballivy, and S. Field investigation of controlling acid mine drainage using alkaline paper mill waste. *Water, Air and Soil Pollut.* **125**, 357 – 374 (2001)
23. Y. Jia, N. Stahre, M. Mäkitalo, C. Maurice, B. Öhlander. *Envir. Sci. and Pollut. Research.* **24**, 20372-20389 (2017)
24. SFS. 2001. *Avfallsförordning (SFS 2001:1063)*. S Riksdag - Ministry of Sustainable Development, Stockholm, Sweden (2001)
25. D. Sanchez, H. Tran. *Enginring, Pulping & Envir. Conf.*, Philadelphia, August 25-31 (2005).
26. A.Virolainen. Master thesis. LTU (2018)
27. J. Macsik, C. Maurice, P. Odén, M. Bergknut. (2017)
28. M. Aachib, M. Mbonimpa, M. Aubertin. *Water, Air and Soil Pollut.* **156**,163-193 (2004)
29. E. Yanful. *J. of Geotech. Engr.* **119** (8) :1207-1228 (1993)
30. W.S. Sillers, D.G. Fredlund, N. Zakerzadeh. *Mathematical Attributes of some Soil—water Characteristic Curve Models. Unsaturated Soil Concepts and their Application in Geotechnical Practice*, Springer, 243-283 (2001)
31. J.M. Tinjum, C.H. Benson, L.R. Blotz. *J. Geotech. Geoenviron. Engr.* **123**(11):1060-1069 (1997)
32. D. Fredlund. (2002). "Use of soil-water

characteristic curves in the implementation of unsaturated soilmechanics." Proc., Proceedings of the 3rd International Conference on Unsaturated Soils, Recife, Brazil, 887-902 (2002)

33. S. Vanapalli, D. Fredlund, D. Pufahl. *Geotech.* **49** (2): 143-159 (1999)
34. A. Zhou, D. Sheng, J.P. Carter. *Géotech.* **62**(8): 669 (2012)
35. C.H. Benson, J.M. Trast. *Clays Clay Miner.*, **43**(6):669-681 (1995)
36. S. Leroueil, J. Le Bihan, S. Sebaihi, V. Alicescu. *Canadian Geotech. J.* **39**(5):1039-1049 (2002)
37. SGI, 2007. Swedish geotechnical institute. Deponiers stabilitet, vägledning för beräkning. Linköping. Information 19 (2007)