

# Microstructural changes leading to chemically enhanced drainage

## Modifications de microstructure entraînant un drainage chimiquement amélioré

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**ABSTRACT:** The hydration state of clay mineral surfaces is a key influence factor on the mechanical and hydraulic behaviour of clays. Small changes of the cation occupancy of these surfaces can lead to pronounced changes in macroscopic material parameters. The sensitivity of the material response allows for designing chemical soil improvement by selectively exchanging the cations. In this study we explore the effect of a targeted cation exchange in smectite clays to modify soil properties in situ. The highly selective and strongly exchanging organic cation guanidinium was used to stabilise the interlayer distance between clay platelets.

On the particle scale the cation exchange led to the formation of stable aggregates. Mercury intrusion porosimetry and oedometer tests confirmed the stability of these aggregates and of an opened pore structure also under high stresses. Macroscopically, the modification resulted in a permanently increased permeability. The magnitude of the improvement is such, that after infiltration of the chemical into a clayey soil around an injection pipe, the modified soil zone could act as a drainage conduit. Potential application of such flexible drainage systems are in creeping landslides, where continuing displacements cause failure of conventional drains.

**RÉSUMÉ :** Un facteur d'influence prédominant pour le comportement mécanique et hydraulique des argiles est l'état d'hydratation des surfaces des minéraux argileux. La sensibilité accentuée de la réponse du matériau permet de concevoir des méthodes d'amélioration chimiques pour sols au moyen d'un échange ciblé de cations. Dans cette étude, nous explorons les effets d'un tel échange dans le contexte d'un argile smectique pour modifier les caractéristiques d'un sol in situ. Le cation organique guanidinium a été utilisé pour stabiliser l'espace entre les couches des lamelles d'argile. Au niveau particulaire, l'échange de cations a engendré des agrégats stables. Les essais oedométriques et de porosimétrie par intrusion de mercure ont confirmé la stabilité de ces agrégats et une structure de pore ouverte, même sous forte sollicitation. Au niveau macroscopique, la modification se manifesta par une augmentation permanente de la conductivité hydraulique. L'amélioration est telle qu'après l'infiltration du produit chimique dans un sol argileux entourant une conduite d'injection, le sol affecté peut être utilisé comme drain. Les applications de tels drainages se trouvent par exemple dans les glissements de terrains en état de fluage, où les déplacements continus entravent le fonctionnement de drainages conventionnels.

**KEYWORDS:** Soil improvement, chemical modification, hydraulic conductivity, clay minerals, drainage.

## 1 INTRODUCTION

Seepage of groundwater in creeping landslides is a key parameter for the creep velocity. A reduction of the water table by means of drainage could increase the overall safety of critical slopes. Conventional drainage systems based on rigid drainage pipes are prone to failure due to the on-going deformation in such unstable areas. The development of alternative drainage techniques with increased operating life in creeping landslides has therefore a high potential in commercial application.

For fine grained soils, both theoretical and experimental studies have pointed out a high dependency of the permeability on the pore fluid. Major influence factors are the type of liquid (mainly via its dielectric constant, Fernandez and Quigley 1985), type of dissolved salts and ionic strength of the solutes (e.g. Madsen and Mitchell 1989, Lagaly et al. 2006). These studies were often carried out in the context of hydraulic barrier design and containment of nuclear wastes, where the increase was an unintended and dangerous effect caused by contaminants and leachates.

With the goal of purposely creating zones of higher permeability in-situ as part of a drainage system, this paper focuses on the fundamental aspects required in the development of an innovative soil improvement technique. Enhancing soil permeability with chemicals may be accompanied with unexpected side effects such as deterioration of stiffness or strength. In field applications such mechanical consequences could result in excessive deformation or failure. The effects on

other geotechnical parameters than hydraulic conductivity therefore need to be addressed as well.

## 2 MATERIALS AND METHODS

Mineralogical investigations indicated that guanidinium cations affect directly the interlayer distance of the stacked sheet-silicate structure of montmorillonite (Plötze and Kahr 2008). This strongly binding cation is therefore chosen as a possible chemical agent to increase permeability by inhibition of interlayer swelling.

In order to assess the mechanisms behind the increase in permeability caused by guanidinium, experiments on different scales were performed. To quantify the changes in soil fabric and structure due to the cation exchange, a closer look on the resulting changes on particle and aggregate scale was taken. The macroscopic stability of the new features was subsequently determined in standard geotechnical tests.

### 2.1 Materials

The laboratory tests were carried out on soil samples reconstituted with commercially available, standardised constituents. A commercial Ca-bentonite (Calcigel, Südchemie, Germany) with a total montmorillonite content of 65% was used as fine grained component. Other occurring mineral phases in this bentonite were quartz, feldspar, kaolinite and mica. Where

appropriate the bentonite was stretched with rounded inert quartz grains (washed Perth sand,  $d_{50} = 0.24$  mm, Cook Industrial Minerals, Australia).

The pore water of the reconstituted samples was prepared with a 0.01 mol/L  $\text{CaCl}_2$  solution as standardised groundwater equivalent.

Guanidinium solutions were prepared from analytical grade guanidinium hydrochloride salt ( $\text{C}(\text{NH}_2)_3\text{Cl}$ ,  $\geq 99\%$ , Fluka Analytical, Switzerland) and demineralised water.

## 2.2 Investigations on particle scale

Homo-ionic bentonites were produced by saturation of Calcigel-clay with 1.0 mol/L calcium and guanidinium solutions, respectively. Images of the particles and aggregates were taken with a scanning electron microscope. The grain size distribution of the suspended material was measured with a Laser scattering analyser (Partica LA-950, Horiba, Germany).

After saturation, the modified soils were washed in suspension with demineralised water to remove excess ions and to avoid salt precipitation in the inter-particle pore space. Compacted samples were reconstituted from the washed material at water contents slightly above to their liquid limit. Mercury intrusion porosimetry (MIP, PASCAL 240/440, Porotec, Germany) on dry samples was used to quantify the alterations of the pores system due to guanidine treatment. Crack-free pieces of the slowly dried clays were subjected to vacuum evacuation for 2 h prior to mercury intrusion.

## 2.3 Macroscopic tests

The primary and intended soil improvement of chemically enhanced drainage is the increase in hydraulic conductivity. This increase was measured based on the evaluation of time settlement curves on bentonite samples. For this purpose, compacted samples of unmodified bentonite were reconstituted and mounted into a standard oedometer cell. The chemical agent was then delivered to the soil in the pore water by diffusion from the top and bottom filter plates. Subsequent loading up to 800 kPa allowed deriving hydraulic conductivities at different void ratios. Untreated samples served as reference for comparison.

Oedometric tests on compacted mixtures of bentonite (40 %) and quartz sand (60 %) were carried out to assess both the increase in permeability and the effects on stiffness due to the chemical treatment. Samples were reconstituted with different chemical composition of the pore water - either artificial groundwater or guanidinium solutions - and subsequently tested according to the procedure given in ASTM D2435-04.

The same mixtures were used in constant head permeameter tests, where the permeability was measured on both modified and unmodified soil. Additionally, the temporal evolution of the permeability during flow-through treatment with guanidinium solutions was recorded for a sample with initially unmodified soil.

Since chemically enhanced drainage is planned to be applied in the context of stabilisation measures, the effects of the chemical modification on the strength parameters of the soil should not be neglected. Therefore, we assessed the influence on the residual shear resistance of pure bentonite samples with a ring shear apparatus. The samples were directly reconstituted from homo-ionic calcium and guanidinium bentonites, as the device used in this study did not allow for chemical modification of the soil within the sample cell.

## 3 RESULTS AND DISCUSSION

### 3.1 Modification of particles and pores

During sample preparation of modified bentonites, a granular, non-plastic behaviour was observed. Images taken with a

scanning electron microscope (Figure 1) revealed that in suspension the clay fraction aggregated upon addition of guanidinium.

The measurement of the particle size distribution supported this observation. Both calcium and guanidinium bentonites feature a bimodal distribution. However, figure 2 shows that the total volume fraction of the larger mode – containing the aggregates – is almost doubled for the guanidinium samples (42.4 %) compared to the calcium clay (22.4 %).

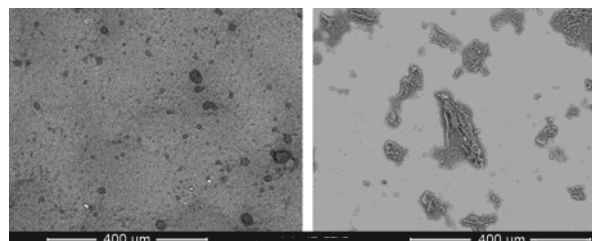


Figure 1. SEM-images of bentonite grains after washing in suspension with demineralised water. The calcium form remains finely dispersed (left), whereas the exposure to guanidinium ions (right) leads to the formation aggregates.

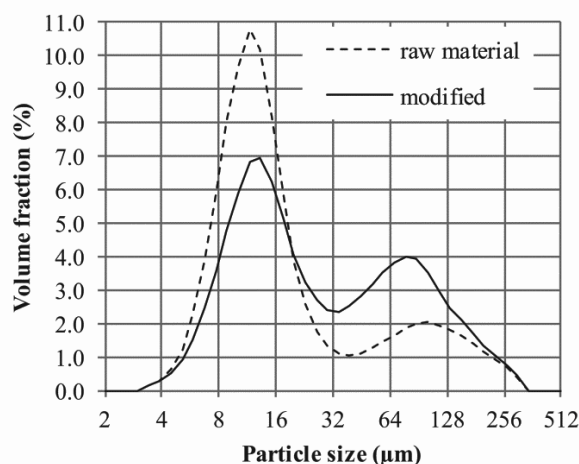


Figure 2. Bimodal particle size distribution measured with laser diffraction. The volume fraction of the larger mode (aggregates) is significantly increased by the chemical modification.

Compacted samples were analysed with MIP in order to examine whether these aggregates were capable of maintaining an open pore structure. Considerable changes in the pore system were detected. Even though the unmodified soil was prepared at a higher water content ( $w_{L,Ca} = 102\%$ ) the modified bentonite ( $w_{L,Gnd} = 64\%$ ) features a larger accessible pore volume (Figure 3). The largest contribution to the additional pore volume stems from pores with average radii of 2  $\mu\text{m}$ . The total pore volume of these larger pores had increased, the volume in the smaller pore fraction (radius  $< 0.1 \mu\text{m}$ ) however was slightly reduced compared to the reference material.

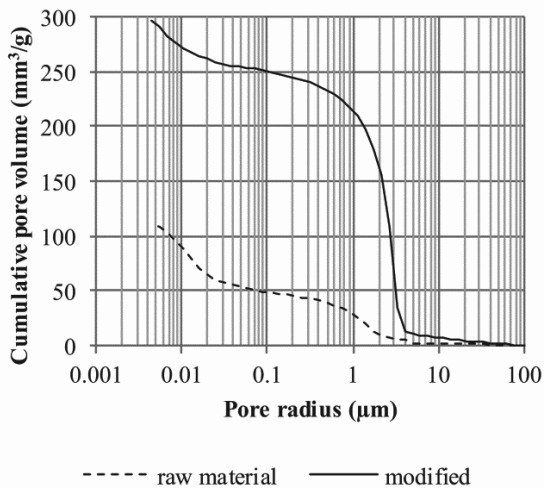


Figure 3. Cumulative pore size distribution of two bentonite samples measured with mercury intrusion porosimetry. The modified material features a much larger pore volume fraction in pores with radii of about 2  $\mu\text{m}$ .

### 3.2 Geotechnical parameters

The tests on pure Calcigel samples, where the chemical was delivered without mixing but diffusively, showed a constantly improved hydraulic conductivity even with this gentle treatment method. The evaluation of the time-settlement curves based on the theory for one dimensional consolidation showed that – although the absolute permeability decreased with increasing compaction – the relative improvement compared to the raw material was preserved and ranging in the order of one magnitude (Figure 4). The aggregated clay minerals are apparently able to keep the newly created flow paths open also without the presence of a rigid sand skeleton.

Oedometer tests on a triplicates series of the quartz/bentonite mixtures were additionally analysed with respect to changes in stiffness. The difference between unmodified and natural soil was small and in the order of the standard deviation for both initial loading and unloading/reloading (Table 1). The evaluation of the time-settlement curve again provided hydraulic conductivities at different stress levels. Figure 5 clearly shows, that despite vertical stresses of up to 800 kPa (corresponding to the smallest void ratios), the increased permeability is maintained during compaction.

Table 1. Stiffness parameters averaged over three samples and three load steps each. Standard deviation is given in parenthesis.

	Raw Material	Modified Material
Compression index $C_c$	0.38 (0.04)	0.35 (0.02)
Swelling index $C_s$	0.09 (0.03)	0.11 (0.03)

By means of permeameter tests the magnitude of possible improvement under flow-through conditions was determined. By mixing soils with guanidinium solutions an increase of the permeability by the factor 30 was achieved (Figure 6). Even when unmodified samples were simply permeated with guanidinium solutions – instead of water – the average hydraulic conductivity increased by one order of magnitude. In potential field application this method of treatment could facilitate the delivery of the chemical in-situ without mechanical disturbance.

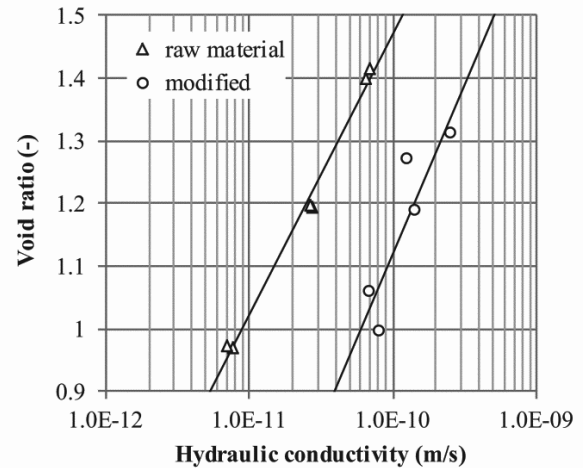


Figure 4. Decrease of hydraulic conductivity during sample compaction (including log-linear regression) of bentonite samples. The relative improvement is not deteriorated during compaction.

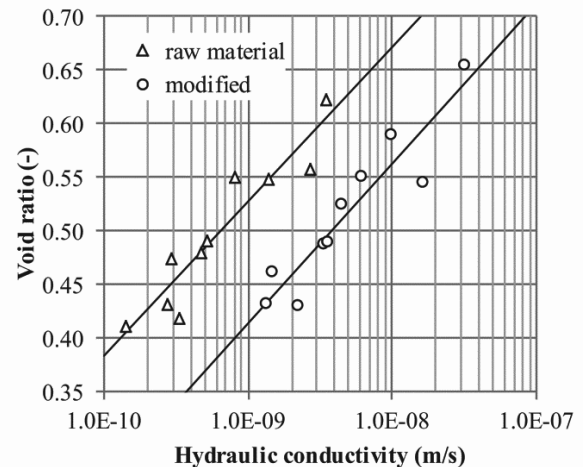


Figure 5. Decrease of hydraulic conductivity during sample compaction (including log-linear regression) of quartz-bentonite mixtures. For identical void ratio the modified soil is constantly about one order of magnitude more permeable.

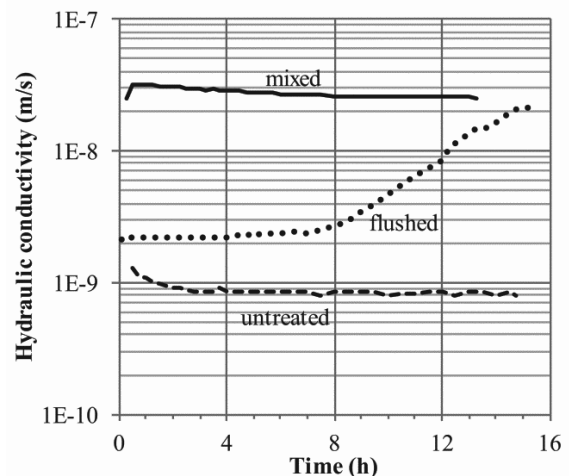


Figure 6. Evolution of hydraulic conductivity during flow-through treatment (dotted line) with guanidinium solution. For comparison the data for raw material (dashed line) and reconstituted modified soil samples (full line) permeated with water are plotted as well.

The effects of the chemical modification on strength parameters are presented based on ring shear tests. The residual

shear resistance was measured by a multi-stage procedure with a constant shear velocity of 0.2 mm/min and three different load steps for each sample. Figure 7 shows the residual shear stress after 30 to 40 mm of displacement. Moreover, the first load step was used to measure one value for the peak shear resistance. By assuming zero cohesion this peak value was used to derive the peak friction angle for each material. Table 2 summarises the friction angles obtained from this data set. It shows that both peak and residual resistance are increased after chemical treatment.

Table 2. Strength parameters of bentonite samples. For all values a frictional material with zero cohesion was assumed.

	Raw Material	Modified Material
Peak friction angle	24°	31°
Residual friction angle	7°	11°

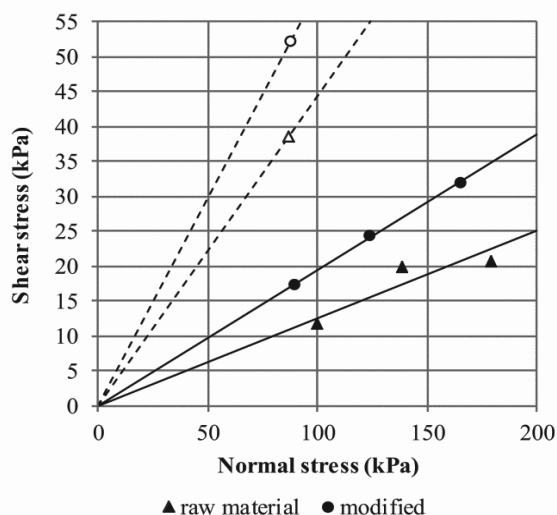


Figure 7. Results of multi stage ring shear tests on bentonite samples. The empty symbols represent peak shear resistance of the initial failure during the first load step loading. Residual state (full symbols) was reached after 30 to 40 mm of displacement in each load step.

#### 4 CONCLUSIONS

Based on the experimental work on different scales the following conclusions could be drawn:

The molecular change of the clay minerals on the level of the surface chemistry results on the particle scale in an aggregation of small clay minerals to larger and stable particles. The aggregated, non-swelling particles provide an open pores system with pores of about 2 μm. Upon wetting these pores are no longer filled with an expanding clay phase but kept open. The pore water is not bound as immobile interlayer water and remains available for circulation.

The compression tests have shown that the new structure is stable also under stresses up to 800 kPa for both pure bentonites and quartz/bentonite mixtures. Despite these drastic changes on the microstructural level, the stiffness is not significantly affected by chemical treatment. The material strength is positively influenced by the chemical treatment as both peak shear resistance and residual shear resistance for the modified bentonite is higher compared to the raw material. A negative influence on these mechanical properties can therefore be excluded.

In summary, the findings are positive indicators for further development of the technique towards purposely creating zones

of higher permeability in-situ as part of a drainage system in creeping landslides.

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#### 6 REFERENCES

ASTM Standard D 2435, 2004. *One-Dimensional Consolidation Properties of Soils Using Incremental Loading*, ASTM International, West Conshohocken, USA.

F. Fernandez, R.M. Quigley, 1985. Hydraulic conductivity of natural clays permeated with simple liquid hydrocarbons. *Can. Geotech. J.*, 22(2), 205-214.

G. Lagaly, M. Ogawa, I. Dékány, 2006. Clay Mineral Organic Interactions. In: F. Bergaya, B.K.G. Theng, G. Lagaly, (eds.) *Handbook of Clay Science*, Elsevier, 309-377.

F.T. Madsen, J.K. Mitchell, 1989. Chemical effects on clay hydraulic conductivity and their determination. *Mitteilungen des IGBM*, ETH Zürich, 135, 67.

M. Plötze, G. Kahr, 2008. Diagnostic intercalation in clay minerals – use of Guanidine carbonate. *Proc. of the 4th MECC, Mineralogia*, 33, 132.