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Effects of pore water chemistry on the unloading-reloading behaviour of reconstituted clays

Effets de la chimie de l'eau interstitielle sur le comportement de déchargement-rechargement des argiles reconstituées

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ABSTRACT: Both clay mineralogy and pore fluid chemistry play an important role in the assessment of the compression characteristics of clays. Based on a series of 1D oedometer tests, a study was carried out to investigate the influence of pore water chemistry and smectite content on the compression, swelling, reloading and consolidation behaviour of mixtures of kaolin and sodium bentonite powders with saline pore fluid of varying salinities and cations. This paper presents part of the study with special focus on the effects of pore water chemistry on the 1D deformation behaviour during unloading and reloading. The paper presents the general effects of clay mineralogy and pore fluid salinity on deformation characteristics of clays, emphasis being on the overconsolidated stress regime. A strong influence of smectite content, pore fluid salinity and valance of the cations in the pore fluid was observed on volumetric compression in both virgin state and for overconsolidated conditions. The hysteretic behaviour of clays in unloading-reloading is found to become more pronounced with increasing smectite content and decreasing pore fluid salinity. Hence, the findings clearly indicate that the influence of pore water chemistry is strictly governed by the presence of smectite minerals and their diffuse double layer.

RÉSUMÉ: La minéralogie de l'argile et la chimie des fluides interstitiels jouent un rôle important dans l'évaluation des caractéristiques de compression des argiles. Sur la base d'une série d'essais 1D oedometer, une étude a été réalisée pour déterminer l'influence de la teneur en chlore et en smectite de l'eau interstitielle sur le comportement en compression, gonflement, rechargement et consolidation. de mélanges de poudres de kenton et de bentonite de sodium avec un fluide interstitiel salin de salinités et de cations variables. Cet article présente une partie de l'étude en mettant l'accent sur les effets du traitement par l'eau interstitielle sur le comportement en déformation 1D lors du déchargement et du rechargement. Le présente les effets généraux de la minéralogie de l'argile et de la salinité des fluides interstitiels sur les caractéristiques de déformation des argiles, en insistant sur le régime de contraintes surconsolidées. Une forte influence de la concentration de smectite, de la salinité du fluide interstitiel et de la variance des cations dans le fluide interstitiel a été observée sur la compression volumétrique à l'état vierge et dans des conditions de surconsolidation. Le comportement hystérétique des argiles lors du déchargement-rechargement devient généralement plus prononcé avec l'augmentation de la teneur en smectite et la diminution de la salinité du fluide interstitiel. Par conséquent, les résultats indiquent clairement que l'influence de la chimie de l'eau interstitielle est strictement régie par la présence de minéraux de smectite et leur double couche diffuse.

Keywords: Pore fluid chemistry; compressibility; unloading-reloading behaviour; clay minerals

1 INTRODUCTION

The physical properties and engineering behaviour of clays are governed by several factors including particle size distribution, mineralogical composition (Lodahl, 2017), and chemistry of pore fluid (Terzaghi et al. 1996). Several previous studies have shown that the engineering properties of clays can be significantly affected by changes in pore fluid chemistry. The effects of pore fluid chemistry are most pronounced in clays characterised by a high content of clay-sized particles and smectite minerals. The diffuse double layer theory is typically used to explain this behaviour (Mitchell and Soga, 2005).

The influence of pore water chemistry must be considered when assessing the engineering behaviour and properties of natural clays of marine origin. If not considered, the obtained engineering properties may not represent the in situ state and/or behaviour to a satisfactory degree. Often, deionised water or tap water is used for e.g. Atterberg limit tests or when preparing reconstituted specimens for soil element testing. This may result in a dilution of the salinity of the natural pore fluid, which in turn leads to an alteration of the physical properties and intrinsic behaviour of the soil, which renders the results unsuitable as reference for the natural state.

Multiple authors have investigated the effects of pore fluid salinity on the Atterberg limits, e.g. Tiwari and Ajmera (2014), Spagnoli and Sridharan (2012) and Sivapulliah and Savitha (1999). In general, increasing pore fluid salinity or cation valence in the pore fluid results in a reduction in the Atterberg limits, provided that the tested specimens contain smectite minerals. Illite rich specimens are, however, found to show the opposite effect (Bjerrum and Rosenqvist, 1956) due to rather stable edge-face bonds in saline conditions. Figure 1 illustrates the magnitude of suppression of the Atterberg limits when comparing saline and freshwater conditions for a clay, based on a comparison of data available from literature. In the figure a strong depression of the ratio of the liquid limit in saline conditions, $w_{L,s}$ and in deionised conditions $w_{L,DW}$ is observed above $w_{L,DW} < 100\%$, as also presented by Yukselen-Aksoy et al. (2008). However, also for series below this threshold, significant influce of pore fluid salinity can be observed from the data – even for
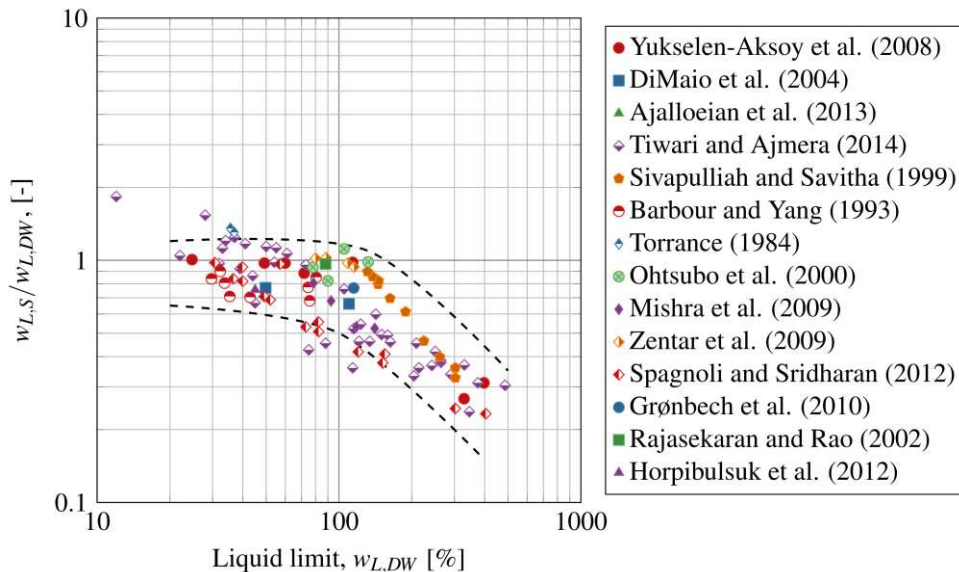

Figure 1: Relative change in w_L when comparing deionised water (subscript DW) and saline water (subscript S) for artificial and natural clays from literature. Salinity for all data is above 2.9 %.

$w_{L,DW}$ as low as 30 %. This suggests that the content of smectite minerals governs the behaviour.

The radical change in w_L with increasing salinity, as observed for extremely high-plasticity clays and some intermediate and high-plasticity clays, is also reflected in studies on compression index, C_C. The magnitude of reduction in C_C when comparing specimens tested using deionised water and saline water is found to be of approximately equal magnitude to the reduction in w_L for extremely high-plasticity clays (Tiwari and Ajmera, 2014, Di Maio et al., 2004).

Previous studies have primarily focused on the effects of pore water chemistry (salinity and cation valence) on the 1D deformation behaviour in normal compression, while the influence on the swelling and associated recompression behaviour in the overconsolidated region has received little attention.

This paper presents results from parts of a larger study into the influence of pore water salinity and cation valence on the 1D deformation behaviour of kaolin-sodium bentonite mixtures. The presented results focus especially on the observed effects of pore water chemistry on the 1D deformation behaviour during unloading and reloading. Throughout the study, a systematic and uniform approach to sample preparation and testing was followed.

2 MATERIALS AND METHODS

A number of artificial specimens, mixed from clay powders, were tested. Dry, commercially available clay powders were used to achieve clays of high plasticity (50 % < w_L < 70 %), very high plasticity (70 % < w_L < 90 %) and extremely high plasticity (w_L > 90 %) in combination with a range of different pore fluids. The test specimens are named after the convention KXXBYY, where the XX signifies the percentage of Kaolin and YY the percentage of Bentonite, determined from dry mass. The mineral distribution and the clay fraction, CF (diameter, $d \leq 2$ μm) of the clay powders are presented in Table 1. The mineral distributi-

Table 1. Clay fraction (CF) and clay mineral distribution in the clay fraction (d ≤ 2 μm) of the artificial specimens. S = Smectite, I = Illite and K = Kaolinite.

Mixture	S	I	K	CF
K100B0	0.8	14.2	85.0	53
K90B10	10.6	12.9	76.5	56
K80B20	23.7	11.9	64.3	60

ons were obtained based on XRD analyses. The rest of the grains for all mixtures are expected to be in the silt region.

All specimens were tested with pore fluids created from deionised water, mixed with laboratory-grade salts to achieve the desired solutions. In total, 11 different pore fluids were used; comprising deionised water, tap water (approx. 1.2×10^{-4} % salinity, primarily Ca^{++} (Aarhus vand, 2015)), NaCl solutions (2 %, 4 % and 6 %), $CaCl_2$ solutions (0.5 %, 1 %, 2 %, 4 % and 6 %) and PC02 (a synthetic pore fluid used to represent typical pore fluid conditions for natural high-plasticity clays (Femern A/S, 2011), with a total salinity of approx. 1.7 %).

In all tests reported, the liquid limit, w_L was determined by use of the fall cone device (20 mm penetration of 80 g cone with 30° apex) and the plastic limit, w_p by rolling the clay into 3 mm threads until crumbling (BS 1377-2, 1990). Both liquid limit and plastic limit tests were carried out for each of the different pore fluids. The reported water contents were corrected for salt content, according to BS 1377-2 (1990). Generally, w_L-testing was performed prior to partial drying of the clay for w_p-testing.

A total of 24 oedometer tests were carried out on reconstituted, pre-consolidated specimens, prepared by mixing the clay powders with the different pore fluids to obtain slurries with a water content, $w = 1.25 w_L$ prior to pre-consolidation. Pre-consolidation was carried out by step-loading the slurry in a tall acrylic floating ring consolidometer (70 mm inner diameter) to a final nominal vertical pressure $\sigma'_v = 163$ kPa. This procedure facilitates creation of high-quality, homogenous specimens without air bubbles. After pre-consolidation, the specimens were extruded and hand-

trimmed using a cutting ring (D × H = 60 × 30 mm), and transferred to the oedometer ring. The initial void ratio, e_0 of the tested specimen, was calculated based on the initial mass and volume of the specimen (G_s = 2.65) in the oedometer ring for all tested specimens.

The oedometer tests were carried out using manually operated load frames with a 1:10 load exchange ratio. The displacement of the upper pressure head was measured using a potentiometric displacement transducer, and each new load step was initiated when the displacement log indicated complete primary consolidation and some secondary consolidation under the current load. In all tests, a pseudo-fixed ring setup was applied as the ring was supported, limiting downwards movements, whereas upwards lifting was not hindered. However, lifting was only observed in a single load step for two specimens (during final unloading from 1925 kPa to 9 kPa in tests on K90B10 and K80B20, using deionised water). Compliance of the load frames and cells was accounted for by obtaining the false compression in the equipment using a dummy steel disc, tested with the planned loading programme.

All specimens were tested using a load increment ratio of approx. 1, following the same programme: a) A loading phase from 17 kPa to 486 kPa, b) unloading to 35 kPa (every other load increment was skipped due to time constraints), and c) reloading to 1925 kPa. Finally, the specimens were d) unloaded to 9 kPa and allowed to completely swell prior to removal from the oedometer cell. In all tests, the specimens were submerged under testing in the same pore fluid as was used during slurry mixing to avoid leaching and osmosis effects. Measures were taken to minimize evaporation during testing. After testing, the obtained time curves were interpreted using the method presented by Casagrande (1938) to obtain the time and void ratio for 100 % consolidation.

For each test, the compression index, C_C was extracted from the normally consolidated region of the stress-void ratio curves as the best fit in the stress interval of 243-1925 kPa, while the swelling index, C_S was determined from the unloading
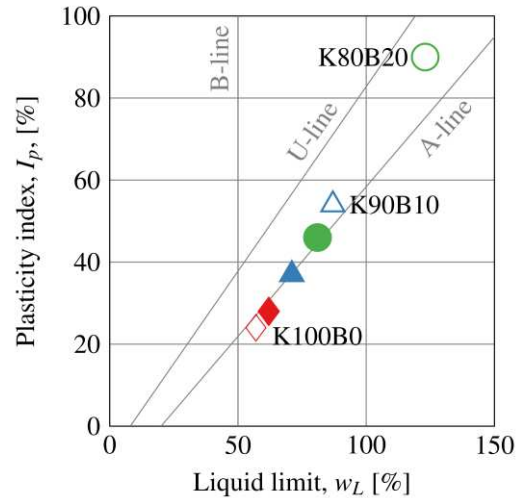

Figure 2: Casagrande chart with colour-coded results for artificial specimens. Open markers denote values obtained using deionised water and filled markers the values obtained using saline water (~1.7 % salinity).

curves as secant values in the interval 486-35 kPa.

3   ATTERBERG LIMITS

The change in the Atterberg limits caused by the change in pore fluid salinity is shown in the Casagrande plasticity chart, presented in Figure 2 It can be seen from Figure 2 that the liquid limit as expected, reduces significantly with increasing pore fluid salinity for artificial clays with $w_{L,dW} >$ 90 %. Thus, it is clear from the results that the observed behaviour is mainly governed by the smectite content. Increasing smectite content results in increasing sensitivity of the liquid limit towards changes in pore fluid salinity. While the small content of illite in the kaolin powder can explain the small increase in liquid limit observed with increasing salinity for specimen K100B0. In contrast to the sensitivity of the liquid limit, the plastic limit, w_p, of all the tested clays was generally seen to be little affected by changes in pore fluid salinity. Hence, the plasticity index for the clays showed a change approx. equal to the observed change in the liquid limit.

4   1D DEFORMATION BEHAVIOUR

The effect of the pore fluid salinity on the compression and swelling behaviour is illustrated in Figure 3. The figure shows compression curves for K80B20, tested with different pore fluid chemistries. The curves for the other mixtures were similar, but the strongest effect of salinity was seen for the K80B20 specimens due the high content of smectite minerals. For K80B20 mixtures tested with deionised water, the initial void ratio is seen to be significantly higher than the initial void ratios obtained for saline mixtures. This illustrates the significant effect of salinity on the compression behaviour and state for smectite-rich clays. Moreover, it was observed that the void ratios at given stress levels for $CaCl_2$ specimens were slightly lower compared to NaCl specimens.

Figure 4 compares the unloading-reloading behaviour of the K80B20 specimens tested with deionised water and NaCl solutions. The plot depicts the change in void ratio from the start of unloading against vertical effective stress. It can be seen from the figure that the swelling response reduces significantly as the salinity of the pore fluid increases. Similar results, but reduced effect, was obtained for specimen K90B10, while the effect of pore fluid salinity was insignificant
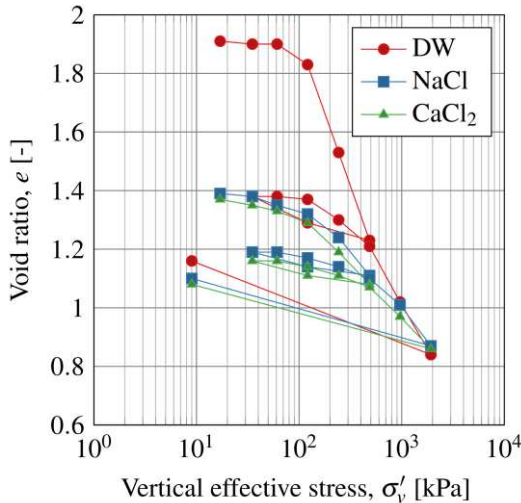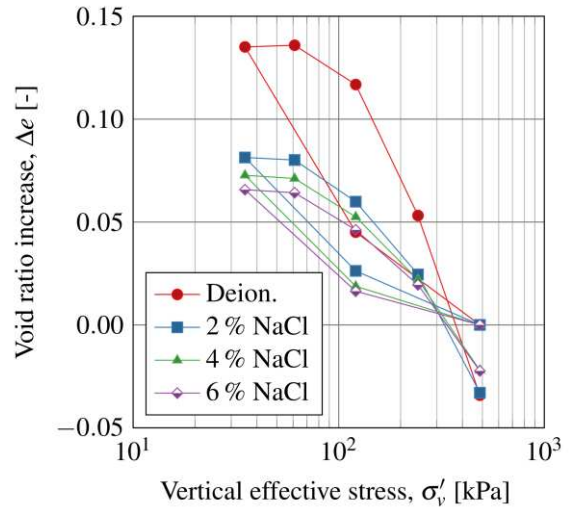

Figure 4: Influence of pore fluid salinity on unloading-reloading response of K80B20 specimens.

for specimen K100B0. A similar influence of salinity was found during recompression as seen from Figure 4.

Thus, all unloading-reloading curves are very close to being closed loops. Interestingly, the accumulated plastic deformation after a full unloading-reloading cycle appears not to be influenced by salinity. Looking at the closed loops, the size of the hysteresis, i.e. the energy loss, increases as smectite content increases and salinity

Figure 3: Influence of pore fluid salinity on deformation behaviour of K80B20 specimens (6 % salinity).
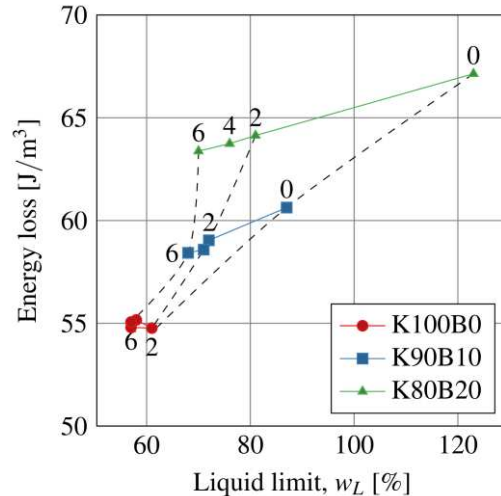

Figure 5: Energy loss during unloading and reloading as a function of liquid limit and clay mineralogy. Salinities presented with numbers next to the points.

decreases (i.e. increasing plasticity of the specimens), as illustrated in Figure 5.

Figure 5 illustrates the energy loss of the unloading-reloading loop, calculated for the mixtures tested with NaCl solutions. The energy loss (the area of the unloading-reloading loops) was found to increase nearly linearly with an increasein liquid limit for each of the mixtures. Although the energy loss is found to be influenced by the salinity of the pore fluid for specimens with high smectite content, it is clear from the plot that the energy loss for specimens with high-salinity pore fluid (above 6%) are mainly governed by the content of smectite minerals, while being rather insensitive to further increases in salinity. Thus, it seems clear that the smectite minerals are heavily influencing the unloading-reloading behaviour. Hysteresis during unloading-reloading may also be a result of internal friction between the oeodometer ring and specimen. However, Lodahl et al. (2016) documented this to be insignificant.

The effect of pore fluid salinity on the swelling index is presented in Figure 6 for specimen K90B10. From the results shown in the figure, it is evident that the swelling behaviour is strongly influenced by pore fluid chemistry. Figure 6 shows a rapid decrease in C_S-value with increasing pore fluid salinity from deionised water to 2
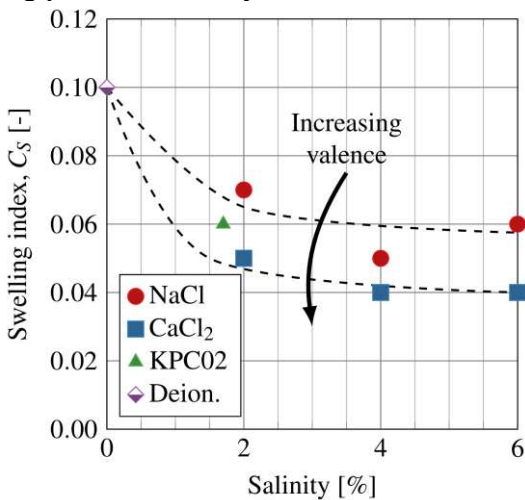

Figure 6: Influence of pore fluid salinity on C_S for K90B10. Stress interval: $\sigma_v' = 486 – 35$ kPa.
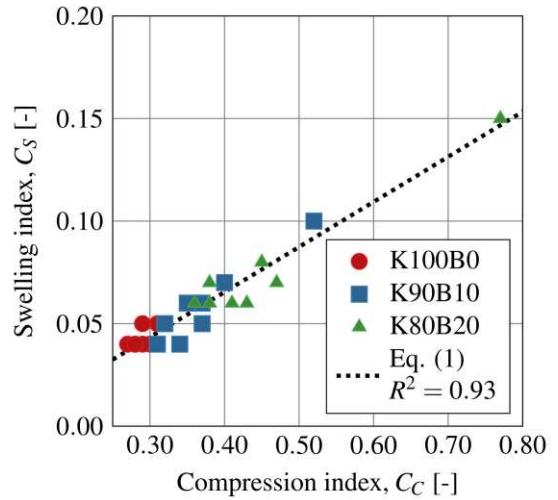

Figure 7: Unique relationship between compression index and swelling index for tested artificial clays mixed with deionised and saline pore water.

% salinity, and a tendency towards a larger effect for divalent ions compared to monovalent ions. Although specimen K80B20 showed the largest effect of pore fluid salinity on the swelling behaviour, the effect of cation valence was not as clear as shown for specimen K90B10 in Figure 6. Similar effects were observed for the compression indices C_C of the tested specimens, cf. Figure 3. Figure 7 shows values of C_C plotted against C_S from all conducted tests with a range of salinities (0-6 %) and cation valence (Na^+ and Ca^{++}). The data is found to plot reasonably close to a straight line, and hence, despite large variations in mineralogy, pore fluid salinities and cation valence, a good correlation between C_C and C_S is found. This indicates that the compression and swelling behaviour are governed by the same factors, and that these parameters correlate well when consistent stress intervals were used for deriving the parameters. The relationship between C_C and C_S is presented in Equation (1) based on a linear best fit ($R^2 = 0.93$) to the data from the present study.

$$C_S = 0.2198\, C_C - 0.0225 \quad (1)$$

5 CONCLUSION

The present study shows that the deformation behaviour and thus the magnitude of deformation parameters are highly sensitive towards pore fluid salinity for clays containing smectite minerals. Based on a range of consistenly performed oedometer tests on artificial clays specimens prepared with different pore fluids, the effects of the ion type and concentration on the deformation behaviour are highlighted, with special focus on the unloading-reloading behaviour. For specimens containing smectite minerals, an increase in the salinity of the pore fluid decreases the liquid limit, the compressibility and the ability to swell. As the smectite content increases, so does the effect of an increase in pore fluid salinity.

In addition, the study shows, that the pore fluid salinity affects the magnitude of void ratio increase during an unloading-reloading cycle. The recompression potential is found to be influenced by salinity and smectite content in similar ways to the swelling behavior. As a result all unloading-reloading loops are found to be essentially closed. Moreover, the results show that the energy loss (area of hysteresis loop) during a full unloading-reloading loop is close to being linearly dependent on the liquid limit for a given mineralogy.

The presented data illustrates that the change in deformation behaviour is most profound between 0 and 2 % salinity. Thus, performing Atterberg limit testing on natural, marine clays containing smectite minerals with deionised water may dillute the in situ pore fluid, and break any relationship with deformation parameters obtained using natural salinity pore fluid. Based on the observations in the tests performed within the current study, it may be concluded that sensitivity of clay behaviour towards pore fluid salinity may not be restricted to liquid limits above a certain threshold. This contradicts the previous study, where a fixed threshold of $w_L =$ 110 % was proposed. On the contrary, large effects of pore fluid salinity were observed at significantly lower liquid limits – both in the tests presented in the current paper and from data presented in literature. It may be concluded that the behaviour is controlled by the presence of smectite. Hence, low plasticity soils will also be influenced by pore water chemistry if smectite minerals are presents.

The data in the present study clearly highlights that the concentration of ions in the pore fluid is a very important parameter. Thus, the authors recommend that more emphasis should be put on performing tests with in situ salinity to enable the development of more consistent relationships between index parameters and deformation behaviour.

6 ACKNOWLEDGEMENTS

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