

Geotechnical characterisation of Mg-hydroxy-interlayered smectite: A potential sustainable solution for soil stabilisation

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Clayey soil is detrimental for many geotechnical applications. Especially smectite, a widespread clay mineral in soils, presents a challenge for engineers, as it is highly plastic, has low shear strength, long consolidation times, and large swelling and shrinkage strains. As a solution, Portland cement, or any other CaO-based cementitious binder (CB), is commonly used to stabilise smectite. Unfortunately, the production of such stabilizers is energy-intensive and contributes heavily to global CO₂-emissions. A promising alternative is the use of Mg-based cementitious binders (MB), which can be produced with a potentially low-to-negative CO₂-emissions. MB alter the crystal structure of smectite minerals during hydration, forming Mg-hydroxy-interlayered smectites (HIS). HIS exhibit strongly reduced cation exchange capacity, reduced specific surface area, and, most importantly, reduced swelling capacity, compared to untreated smectites. However, the impact of HIS formation on geotechnical properties remains unclear. The aim of this study was to produce HIS out of smectite and MgO and to characterize its geotechnical behaviour. Preliminary results show that, compared to common smectite, the plasticity index of HIS was reduced by ~80%. Likewise, swelling and shrinkage are reduced by ~75-80%. Consolidation tests reveal a significantly higher coefficient of consolidation (c_v) for HIS, attributed to increased hydraulic conductivity. Granulometry and ¹H-NMR relaxometry analyses indicate greater particle aggregation and enlarged pores, even at the nanoscale. Overall, HIS exhibits geotechnical properties closer to silty materials, making it a promising material for sustainable soil stabilisation.

KEYWORDS: Soil Stabilisation, Mg-hydroxy-interlayered smectite.

1 INTRODUCTION

In civil engineering, clayey soils present a great challenge for engineers to build on. Clayey soils, or short “clay”, are fine-grained soils with difficult geotechnical behaviour as they exhibit high plasticity, large swelling and shrinkage strains. The detrimental effect of a clay depends largely on the type of clay mineral and its abundance. There are three common clay minerals in subsoils: kaolinite, illite, and smectite. Of these three minerals, smectite is by far the most detrimental for geotechnical properties.

The common solution to stabilise smectite-rich soils is the usage of quicklime, which is composed of lime (CaO). While this solution is an approved method known for decades, there is a crucial disadvantage: the lime is obtained by the calcination of limestone. The calcination is a highly energetic process that directly emits CO₂ in the atmosphere. In 2022, the USA alone produced 1’890’000 metric tons of lime for soil stabilisation, which is 11% of the total US lime production (U.S. Geological Survey, 2024). This is approximately 2’226’000 tons CO₂ emitted, including calcination and heating energy (Miller et al., 2016). In the context of reducing greenhouse gas emissions, calcination is viewed negatively, and there are ongoing alternatives to replace lime with more environmentally friendly materials.

A potential environmentally friendly solution is the usage of a MgO-based cementitious binder (MB). Unlike CaO-based binders, MB can potentially be produced carbon neutral or even negative (Bernard et al., 2023). Its stabilising effect on soils was also shown in previous studies (Bernard et al., 2024). In the case of smectite-rich soils, however, an additional effect occurs: Smectite is structurally transformed into Mg-hydroxy interlayered smectite (Mg-HIS) (Zucha et al., 2025).

Smectite is, in fact, a mineral group, and the most prominent mineral of the smectite group is montmorillonite. For the sake of simplicity, the term smectite is used throughout this

text. Smectite is structurally composed along the c-axis of two SiO₂ tetrahedral sheets (T), one octahedral Al(OH)₃ or Mg(OH)₂ sheet (O). On top of the TOT layer is an interlayer of variable thickness. This interlayer is generally composed of cations (i.e. Na⁺, Ca²⁺, and Mg²⁺) and a variable amount of water, which hydrates the interlayer cation. The Mg-HIS, on the other hand, has a different interlayer. The hydrated cations in the interlayer are exchanged by mono- and oligomeric Mg-hydroxy (Figure 1).

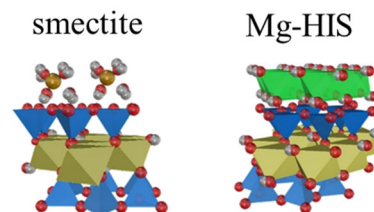


Figure 1. Crystallographic structure of smectite and Mg-HIS. The hydrated cations in the interlayer (left) are replaced by Mg-hydroxy (right).

This exchange fundamentally alters the physicochemical properties of the smectite. The specific surface area (SSA) is reduced by more than 50%, depending on the clay used, and the cation exchange capacity CEC is almost entirely lost. In terms of geotechnical behaviour, previous work has shown that the shrinkage of Mg-HIS is reduced by ~50% compared to smectite (Zucha et al., 2025). However, other microstructural and geotechnical properties have not yet been quantitatively assessed. The objective of this study was, therefore, to measure and quantify the geotechnical properties of Mg-HIS. To achieve this, a bentonite with high smectite content was transformed into Mg-HIS. The Atterberg limits, consolidation behaviour, and swelling characteristics of the bentonite used and Mg-HIS were analysed. Additionally, the aggregate size and pore sizes were analysed.

2 METHODS

2.1 Material used

A bentonite (labelled ES) from the supplier Argiles du Bassin Méditerranéen was used. ES is composed mostly of smectite (~90 wt.%) and traces of illite, quartz, and feldspars. A detailed characterisation can be found in Zucha *et al.*, (2025). The MgO was obtained by heating Mg(OH)₂ (Merck) at 600 °C in a muffle oven for 3 h, followed by unforced cooling to room temperature.

2.2 Mg-HIS formation

Formation of Mg-HIS was achieved after mixing bentonite with MgO (weight ratio 5:1) in a suspension with a liquid/solid ratio of 10. The suspension was constantly shaken for 48 h. Afterwards, the liquid was decanted and the material was dried at 65 °C. The successful transformation was verified by X-ray diffraction analysis (XRD) and thermogravimetric analysis (TGA).

2.3 Geotechnical and microstructural characterisation

The liquid and plastic limit were measured according to the DIN EN 17982 norm. One-dimensional consolidation properties of the material were determined by using an oedometer with incremental loading according to the ASTM D2435 norm. The evaluation was done using the *square root* method. Swelling was investigated under incremental loading in an oedometer cell according to the ASTM D4546 norm. Particle size distribution was measured with a particle size analyser LA-960 from Horiba using a 0.1% calgon solution. ¹H NMR T2 relaxometry was carried out on samples conditioned at 75%RH using a 20 MHz benchtop magnet with a CPMG pulse sequence. For details see (Nagel, Strangfeld and Kruschwitz, 2021).

3 RESULT AND DISCUSSION

3.1 Mg-HIS

X-ray diffraction (XRD) and thermogravimetric analysis (TGA) of the material showed that Mg-HIS was formed. The clearly resolved 002 and 003 XRD reflections are characteristic for Mg-HIS. Likewise, the two characteristic “humps” on TGA pattern are visible. These humps emerge from the dehydroxylation of the Mg-hydroxides. However, both patterns also revealed the presence of brucite. The measured cation exchange capacity (CEC) of Mg-HIS was still ~20 cmol(+)/kg. Hence, approximately 75% of the smectite in the bentonite was transformed into Mg-HIS.

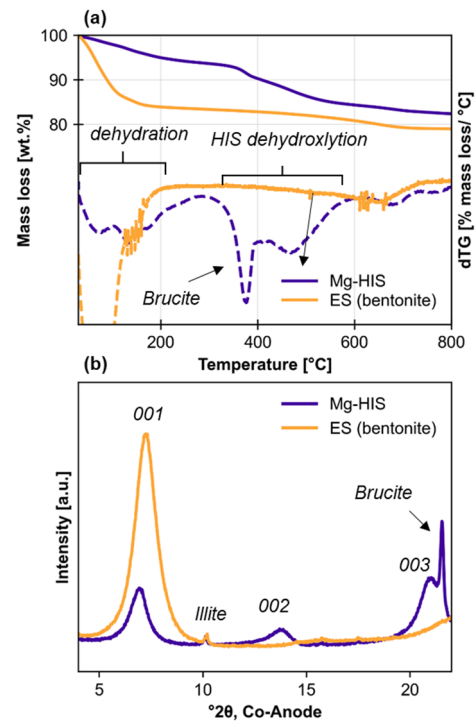


Figure 2. TGA pattern (a) and XRD pattern (b) of ES and Mg-HIS.

The transformation from smectite to Mg-HIS was only partly successful, because the CEC would have been otherwise close to zero. The reason is that not all dissolved MgO precipitated as Mg-hydroxy as an interlayer but precipitated as brucite, as the XRD and TGA measurements confirm. This is a known phenomenon and likely linked to the suspension pH (Zucha *et al.*, 2025). However, at the time of this study, no viable solution was at hand to inhibit brucite formation.

3.1.1 Aggregate size and pore size distribution of Mg-HIS

Figure 3a shows the particle size distribution (PSD) of ES and Mg-HIS. ES had a clay fraction of ~20% and a silt fraction of 80%. Mg-HIS is almost completely composed of silt particles. Likewise, the ¹H-NMR shows shorter relaxation times, which suggests that the sizes of water-filled pores were reduced (Figure 3b).

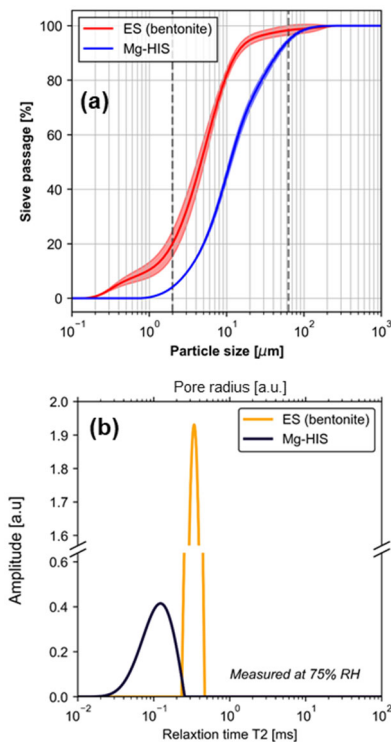


Figure 3. Particle size distribution (a) and NMR relaxation times distribution (b) of ES and Mg-HIS. Dashed lines at 2 and 63 μm mark the clay and silt fraction.

The smectite to Mg-HIS transformation caused an aggregation of the clay particles. However, the reason for the aggregation is unknown. The structural change of the crystal does not enlarge the unit cell. It is also not an effect of the brucite precipitation: (i) Mg-HIS samples that do not contain brucite are also aggregated, and (ii) single crystals of brucite in the scanning electron microscope (not shown). This further indicates that brucite is not a binder to enhance aggregate formation.

3.2 Atterberg limits

The original bentonite sample ES has a plasticity index (I_p) of 92 with a liquid limit of 153% and a plastic limit of 61%. The Mg-HIS sample had a liquid limit of 60% and a plastic limit of 46%, which results in a plasticity index of just 14. This is a reduction of $\sim 85\%$. Likewise, it changes the classification of the bentonite from “very high plastic” to low-medium plastic”.

3.3 Consolidation

Figure 3 shows the void ratio as a function of the incremental load from the oedometer test. ES starts with a higher initial void ratio (~ 1.28) and compresses more sharply to a final value of ~ 0.8 at 400 kPa. Mg-HIS begins with a lower void ratio (~ 0.9) and compresses more gradually. The estimated compression indices C_c are ~ 0.51 for ES and 0.24 for Mg-HIS.

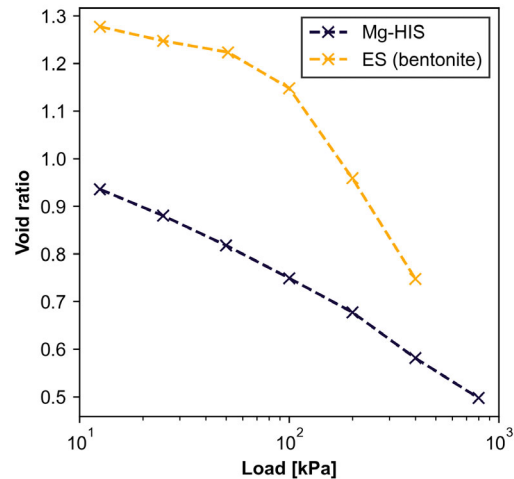


Figure 4. Void ratio vs load of ES and Mg-HIS.

The consolidation coefficient c_v was determined using the square-root method. In case of ES, the determined c_v remains very much stable in the range of 0.1-0.25 m^2/s at all loads. For Mg-HIS, on the other hand, c_v increases steadily from 0.5 m^2/s at 8 kPa to 3.5 m^2/s at 400 kPa.

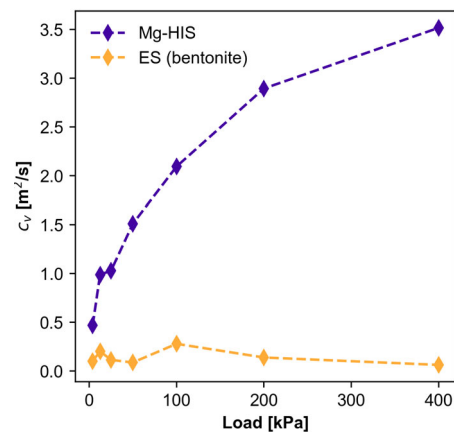


Figure 5. Consolidation coefficient c_v versus load

As expected, ES is a more compressible sample than Mg-HIS. It consolidates also magnitudes lower and exhibits more creep (secondary compression) than Mg-HIS (not shown). The unchanging consolidation coefficient of ES indicates that the hydraulic conductivity and the compressibility of the skeleton decrease at the same rate. Mg-HIS, on the other hand, has a much higher hydraulic conductivity than its parent material. This is a result of the larger aggregate and pore size, as previously mentioned.

3.4 Swelling

Figure 6 shows the swelling strain of ES and Mg-HIS as a function of the load. At the start of tests at 8 MPa, ES had a swelling strain of $\sim 7\%$. Its swelling strain exponentially increased with the step-wise unloading to a final value of $\sim 25\%$ at 1 MPa. Mg-HIS had an initial swelling strain of 2% at 8 MPa,

which increased over to final value ~3% at the end of the test.

Zucha, W. J. *et al.* (2025) 'Effects of MgO-based cementitious binder on smectites', *Applied Clay Science*, 265, p. 107677. doi: 10.1016/j.clay.2024.107677.

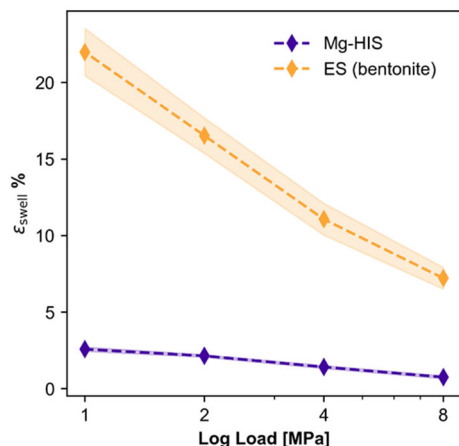


Figure 6. Swelling strain ϵ_{swell} of Mg-HIS and bentonite

The water uptake (and its associated swelling) is several times lower for Mg-HIS than for ES. This demonstrates once again the limited interaction of Mg-HIS with water. The intercalated Mg-hydroxy seal of the interlayer of the smectite. This, and the drastically reduced CEC, limit the interaction with water. The reduction of the pores limit it even further.

4 CONCLUSIONS

The preliminary results presented here demonstrate that MgO-based cementitious binder is a promising candidate for sustainable stabilisation of smectite-rich soils. Compared to conventional smectite (bentonite), Mg-HIS has favourable properties for geotechnical construction. Its plasticity is significantly lower, the consolidation occurs faster, and its swelling is reduced, even in samples that still contain ~25 wt.% of unaltered bentonite. This improvement is the result of two effects: (i) the Mg-hydroxy intercalation, which inhibits the swelling of the otherwise reactive smectite. (ii) an aggregation of the clay particles and a reduction in pore size and amount of pores. The causality between (i) and (ii), however, remains unclear, and the mechanisms need further investigation.

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