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Effect of mixing method on soil water retention and shrinkage properties of polymer-amended bentonite

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ABSTRACT

Polymers have been shown to be able to enhance the resistance of clays to desiccation cracking. This is an important capability in engineering applications such as waste barriers. However, little is known about the reasons for this improvement (e.g., higher water retention, less shrinkage, lower hydraulic conductivity, higher tensile strength) and the most important variables affecting it, in particular the effects of different mixing methods. While dry mixing is easier to implement onsite, wet mixing produces better intercalation between clay and polymer and the two methods will yield different stabilizing outcomes. In this paper, the effects of two mixing methods on water retention and shrinkage properties are investigated. An anionic polymer of sodium carboxymethyl cellulose (Na-CMC) is added to Na-bentonite by dry and wet mixing. A series of laboratory experiments are conducted on amended and unamended bentonite to determine a) soil water retention characteristic (SWCC) using filter paper and dewpoint methods and b) shrinkage potential using the linear shrinkage test. Findings from this study will contribute to a better understanding of the mechanisms underlying the stabilizing effects of polymers on swelling clay and their sensitivity to mixing method.

Keywords: sodium bentonite, polymer, shrinkage, SWCC, Na-CMC

1 INTRODUCTION

It is a common practice to utilize clay materials as a hydraulic barrier in landfill covers or as bottom liners in ponds, reservoirs, and canals because of their low hydraulic conductivity and hence their ability to inhibit water penetration and migration (He et al., 2015; Petrov et al., 1997). However, climatic variations, such as fluctuation in precipitation levels and temperature, can lead to drought or flood events, inducing volumetric deviation (i.e., swelling upon hydration and shrinkage upon dehydration) and as such increasing the likelihood of desiccation cracks (Chaduvula et al., 2017). Typically, under restrained shrinkage conditions, tensile stresses develop and, when they exceed the soil's tensile strength, cracks are initiated (Al-Dakheeli et al., 2017; Corte and Higashi, 1964; Morris et al., 1992; Tang et al., 2012). Desiccation cracks may cause a loss in clay's hydro-mechanical properties. Thus, soil stabilization measurements are often recognized as a potential solution in preventing or mitigating the functional failure of the earthen facilities in landfill cover or lining systems.

Polymer soil stabilization is a method for enhancing clay hydro-mechanical behavior and strength properties in different applications (Lieske et al., 2020). In the early 1980s, hydrogel-forming polymers were first used in agriculture to improve soil water retention and reduce irrigation frequency (El-Hady and Abo-Sedera, 2006; Osman, 2018). Later, these polymers were used in environmental containment facilities such as landfills and tailings impoundments to modify bentonite clay and maintain its low hydraulic conductivity in contact with leachate or aggressive salt solution (Di Emidio et al., 2015; Kolstad et al., 2004; Razakamanantsoa and Djeran-Maigre, 2016; Scalia IV et al., 2018).

In recent years, various polymers, such as polyacrylamide (PAM), Polyurethane (PU), polydimethylsiloxane (PDMS), and sodium carboxymethyl cellulose (Na-CMC), have been empirically investigated and found to be effective in enhancing clay resistance against desiccation cracking (De Camillis et al., 2017; Qi et al., 2020; Tabassum and Bheemasetti, 2020; Yu et al., 2020; Taheri and El-Zein., 2022). However, little is known about the mechanisms underlying this improvement (e.g., higher water retention, less shrinkage potential, lower hydraulic conductivity, higher tensile strength).

Furthermore, the mixing process, which highly impacts the interaction between polymer and clay and the subsequent stabilizing outcomes, has received less attention than it deserves (Norris, 2021). Wet mixing and dry mixing are two common methods of introducing polymer to clay. Wet mixing involves dissolving the polymer in water and then adding the clay to the polymeric solution (De Camillis et al., 2017). Dry mixing, on the other hand, involves the simple blending of the clay and polymer in a dry state (Razakamanantsoa and Djeran-Maigre, 2016). Compared to dry mixing, wet mixing is more likely to yield intercalation of polymers between the clay sheets. However, the later steps of the wet mixing process (oven-drying and grinding) might alter the material properties and make this approach challenging and costly to implement on-site.

This study aims to investigate the mechanisms driving the gains in desiccation resistance of polymer-amended bentonite relative to unamended bentonite. The effects of two mixing methods on water retention characteristics and shrinkage properties have been investigated to understand the mechanisms behind this improvement.

2 EXPERIMENTAL PROGRAM

2.1 Material

The reference soil is a commercial Na-Bentonite provided by Global Synthetics Pty Ltd, Australia, which is used in their Bentofix Geosynthetics Clay Liners (GCL) manufacturing process. The soil's physical and chemical properties are listed in Table 1. An anionic polymer, sodium carboxymethyl cellulose (Na-CMC), with a degree of substitution (DS) 0.9 and a default molecular weight of 250,000, was purchased from Sigma- Aldrich and used to prepare the bentonite-treated specimens.

Table 1. *Soil physical and chemical properties*

Soil properties	Value	Method
Specific gravity	2.89	ASTM D854
Free Swelling Index	30 mL/2g	ASTM D5890
Atterberg Limits		ASTM D4318
Liquid Limit	398%	
Plastic Limit	46%	
Plasticity Index	352%	
USCS classification	CH	ASTM D2487
Compaction Study		ASTM D698
Optimum Moisture Content	31.80%	
Maximum Dry Density	1.38 g/cm3	
Cation Exchange Capacity	≥70 cmol/kg	Methylene Blue

2.2 Bentonite-polymer mixture

To evaluate the impact of mixing method on the mechanism affecting soil behavior, two different methods were followed in preparing the polymer-modified samples: wet mixing (WM) (also known as solution intercalation), and dry mixing (DM). For WM, the method described by De Camillis et al. (2017) was followed: the powder of Na-CMC polymer was first dissolved in water (polymer/water ratio: 1:400), then clay was added to the polymeric solution (soil/solution ratio: 1:7). After 30 minutes of mechanical stirring, the mixture was allowed to equilibrate in sealed buckets for two days. The slurry was then oven-dried for 16 h under 105 °C. After drying, samples were crushed with a Tema Mill machine and passed through sieve No. 200.

For DM mixing, the polymer powder was added directly to the bentonite powder and mixed manually for 5 minutes to help achieve an even distribution of polymer (Haase, 2017; Razakamanantsoa and Djeran-Maigre, 2016). The default polymer dosage was set to 2% in all experiments.

2.3 Soil water retention characteristic (SWRC)

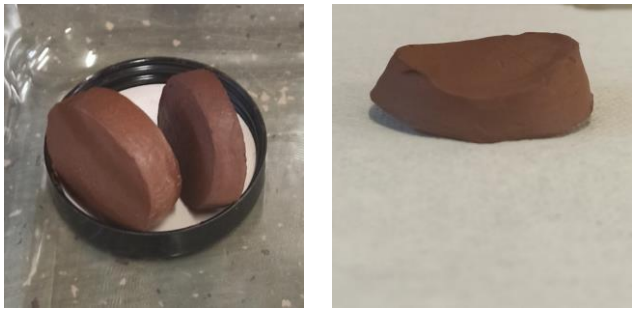
The water retention behavior in bentonite and bentonite-polymer mixtures was measured along the drying path. Low suction ranges (0.01 to 6 MPa) were measured via contact Filter Paper Method (FPM) as per the ASTM D5298 whereas high suction ranges (6 to 550 MPa) were measured by the chilled mirror dew-point device (WP4C).

For FPM, treated and untreated bentonite were mixed with 1.35 times of the liquid limit (w=540%) and consolidated under 45 kPa for 25 days. After completing the primary consolidation stage, the specimens were carefully extruded from the molds and trimmed into 15 mm height disks using a thin wire saw. Every two soil disks were then allowed to be air-dried until they achieved the desired moisture content. Filter papers were then sandwiched between two of the soil disks, sealed, and left for four weeks to reach moisture equilibrium. The gravimetric moisture content of the filter papers and the corresponding matric suctions were then calculated as per the calibration curves for Whatman 42 filter paper proposed by Leong et al. (2002) (Equation 1). The average degree of saturation was also calculated based on the dimensions and moisture content of the top and bottom discs.

$$\log \psi = 2.909 - 0.0299\, w_f; \quad w_f \geq 47$$
$$\log \psi = 4.905 - 0.0673\, w_f; \quad w_f < 47 \qquad (1)$$

where ψ is the matric suction (kPa) , and w_f is gravimetric water content of the specimen disks.

Although FPM can determine the suction in a wide range of 1- 10^6 kPa, it may not be accurate to be used when the soil disks experience extensive shrinkage and deformation during the dehydration process (Figure 1). Because of the poor contact between the uneven surface of the soil samples with filter paper at low water contents, WP4C was used to measure the suction at the dry end of SWCC curve.



(a) (b)

Figure 1*. a) soil discs dehydration for FPM, and b) the curvature form of soil disks*

2.4 Linear Shrinkage (LS)

Linear shrinkage experiments were performed on specimens with and without polymer additive as per the British Standard (BS 1377:90). The soil samples were mixed with water (about 1.1 times the liquid limit, w =450 %), placed in shrinkage troughs, and exposed to air-dehydration. The mass of the specimens was monitored on a daily basis for 15 days until no further change in mass was identified. The length of the samples after shrinking was then measured to calculate the linear shrinkage (LS) in percentage (Equation 2).

$$LS\ (\%) = (L_1 - L_0)/L_0 = \Delta L \times 100 \qquad (2)$$

where L_0 is the initial length of the sample, and L_1 is final length of the dry clay specimen.

3 RESULTS AND DISCUSSION

3.1 Soil water retention characteristic (SWRC)

The SWCC test results for bentonite and polymer-modified specimens, prepared by wet mixing and dry mixing, are shown in Figure 2. Figure 2a and 2b, plots the equilibrium values of matric potential versus gravimetric water content and degree of saturation, respectively. Generally, in all cases, higher matric suction was seen after a decrease in soil moisture content. When comparing samples B (unmodified bentonite) and WM (produced by wet mixing method), not much difference in water retention can be observed.
Following the approach proposed by Pasha et al. (2016), the air-entry value (AEV) for sample B was about 1.7 MPa which seems to be only slightly higher than polymer-modified samples (about 1.55 MPa) irrespective of their mixing method. According to Benson et al. (2014), the AEV is sensitive to particle size, and the finer the particles the higher the water retention at a given suction. It is hence possible that polymer additives can bind smaller soil particles together, potentially resulting in larger soil particles or aggregates (Lieske et al., 2019; Jiang et al., 2022; Taheri and El-Zein, 2023).

However, the effect of polymer additives on soil particle and pore size can vary depending on the type and dosage of polymer used, as well as the properties of the soil being treated. All in all, it appears therefore the polymer-amended bentonite produced by wet mixing, at 2% Na-CMC, achieves only slightly higher water retention compared to both unamended and dry-mixed samples.

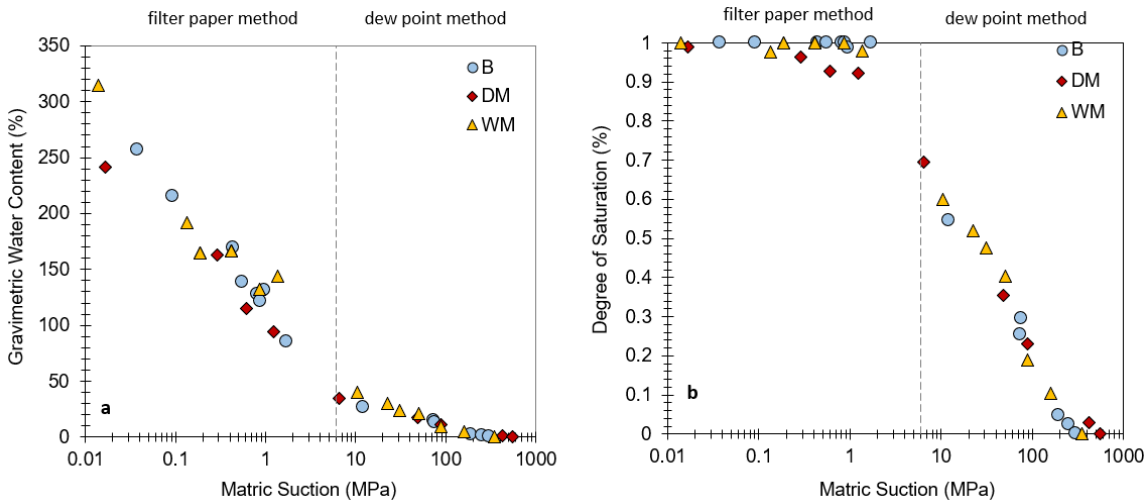


Figure 2. *SWCC-drying path of unamended and amended bentonite, prepared with wet mixing (WM) and dry mixing (DM).*

3.2 Linear Shrinkage (LS)

Linear shrinkage tests were carried out on reference (sample B), dry-mixed (DM) and wet-mixed (WM) specimens. The photos, taken after 3, 10 and 15 days are shown in Figure 3 and the results of the linear shrinkage potential in percentages are presented in Table 2. The test was repeated for
every single specimen to ensure data reliability and, as seen in Table 2, results from repeats confirm patterns from original tests. During the drying process, the soil experienced bending and curling. This happens due to the uneven drying rate, which varies with the distance to the surface and differential shrinkage of the top and bottom sides of the soil layer. At early stages of air-dehydration when the soil is soft (low modulus of elasticity), there is a small curling deformation (concave surface due to upward curling of edges) because the top surface undergoes more shrinkage compared to the bottom surface. Nevertheless, in pure bentonite and dry-mixed specimens, from days 3 to 10, most of the deformation is attributed to the axial shrinkage. However, as the drying process proceeds further, the bottom surface starts shrinking more than the top surface, developing compressive stresses at the bottom and tensile stresses at the top (Lakshmikantha, 2009). Hence, in response to the resulting bending moment, the middle section tends to lift up, giving rise to the downward curling deformation and creating a convex structure (Kodikara et al., 2004). At later stages of drying, when the soil stiffness is high, curling deformation becomes dominant over axial shrinkage. Due to the curling issue, the length measurement

was done from two sides of the sample, using a tailor craft flexible ruler tape, and the average is reported in Table 2. The results showed that the shrinkage potential of sample B (about 47%) was significantly higher than that of the dry-mixed (26%) sample.

The wet-mixed specimen tended to arch and shatter, breaking into pieces (see Fig 3 from day 3 onwards). Although the presence of cracks complicates the comparison with the other two specimens, it is still instructive and the shrinkage potential for the wet-mixing method is reported (based on the combined shrinkage of all pieces of the cracked sample) in Table 2. Results show that the shrinkage potential of the wet-mixed specimen is about 37% which is less than the untreated bentonite sample but higher than the dry-mixed modified specimens. This may be due to structural changes (i.e., decrease in crystallinity) that occur during oven-drying and grinding of specimens in the final stages of wet mixing (Juhasz and Opoczky, 1990; Tole et al., 2018).

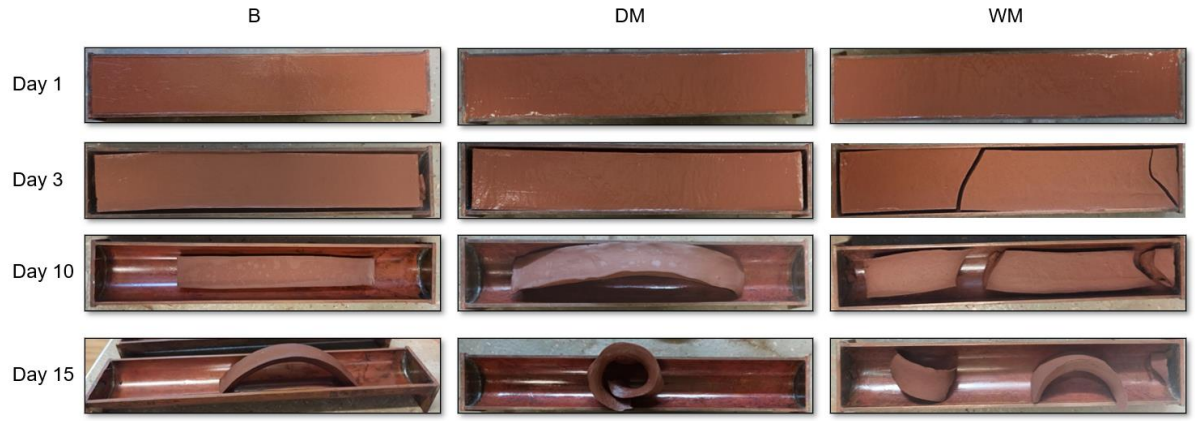


Figure 3. *Linear shrinkage test result for pure bentonite, dry-mixed (DM), and wet-mixed (WM) specimens.*

Table 2. *Sample measurement and linear shrinkage potential in percentage after 15 days.*

Sample	Top Length (cm)	Bottom Length (cm)	Average Length (cm)	Linear Shrinkage (%)
B	13.4	13.2	13.3	47.6
B*	13.1	13.1	13.1	48.4
DM	18.3	18.9	18.6	26.7
DM*	18.6	18.7	18.4	27.5
WM	15.5	16.5	16	37
WM*	14.4	17.1	15.7	37.9

Note: * stands for repeated tests.

4 CONCLUSIONS

The research conducted here has investigated the effect of polymer modification and mixing method, dry mixing (DM) and wet mixing (WM), on soil water retention ability and shrinkage potential of untreated and treated bentonite with sodium carboxymethyl cellulose (Na-CMC). Nevertheless, key findings of this study can be summarized as follows:

- Adding 2% Na-CMC does not make a significant difference in the water retention ability of polymer modified bentonite compared to unamended bentonite. The air entry value (AEV) of bentonite specimen was around 1.7 MPa which is comparable with the AEV of polymer modified samples (1.55 MPa), regardless of mixing method.

- Adding Na-CMC, reduces the shrinkage potential in dry-mixed modified bentonite specimens (26%) compared to unmodified bentonite (47%). Wet-mixed specimens have a shrinkage potential of around 37%, which is less than untreated bentonite samples but more than dry-mixed specimens. The reason might be due to the structural changes (i.e., decrease in crystallinity) that occur during the oven-drying and milling of the soil in the later stages of wet mixing process.

A key motivation of this research is a better understanding of the mechanisms driving observed reduction in desiccation cracking when bentonite is mixed with polymers. Findings of this study seem to indicate that, for 2% Na-CMC addition to the bentonite used here, lower shrinkage potential (evident under dry mixing), rather than any increase in water retention, may help explain the reduction in desiccation cracking. Other possible factors include tensile strength and hydraulic conductivity and research by the authors on the effect of Na-CMC addition on these two material properties is ongoing.

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