

Mechanism of clay swelling in Villarlod molasse, a Swiss sandstone

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Abstract

The characterization of swelling clays is important for diverse fields, including the field of conservation of built cultural heritage. Villarlod molasse, a building stone utilized frequently across Switzerland, is known to be damaged by swelling clays embedded in its matrix. In this study, the mechanism of how the clays lead to swelling in the stone itself is examined, and similar to previous studies, crystalline swelling is noted as the most likely source. A scaling factor linking X-ray diffraction (XRD) and dilatometric swelling experiments is calculated, and evidence for the existence of an initial monolayer of moisture in the embedded clays at ambient relative humidities is presented. A qualitative micromechanical model describing how the nonswelling stone matrix exerts a pressure on the clay layers, affecting their swelling behavior, is presented.

Keywords: swelling, crystalline, sandstone, conservation

1. Introduction

Swelling clay minerals are of interest for diverse areas of geotechnical engineering, in particular stability of slopes, foundations, tunnels, and boreholes (O'Neill and Poormoayed, 1980; Anagnostou et al., 2010; Anderson et al., 2010; Khan et al., 2017). They are of particular interest as well in the field of conservation of built cultural heritage, as they often appear in the matrix of sandstones and other stones used in historic buildings and monuments, where wetting and drying cycles can lead to damage (Sebastián et al., 2008; Ruedrich et al., 2011; Wedekind et al., 2013; Berthonneau et al., 2016; Pötzl et al., 2018). This damage was proposed to come from differential expansion and contraction of the whole stone leading to strain gradients, with damage from wetting cycles leading to buckling failure

(also called spalling, contour scaling), and damage from drying cycles leading to mud cracking failure (Gonzalez and Scherer, 2004; Jiménez-González et al., 2008). Buckling failure, a type of surface delamination more often seen in the field than mud cracking, was shown to require a flaw propagation mechanism in order to reach a critical failure point (Wangler et al., 2011). A more recent study has shown the importance of anisotropy with respect to observed failure mechanisms (Tiennot et al., 2017), and, more critically, the microstructural role that the clays play in the actual fracture process (Tiennot et al., 2019).

The mechanism of swelling in the swelling clay minerals, or smectites, has its origin in the structure of the smectites. Two tetrahedral silica sheets sandwich an octahedral alumina sheet in a 2:1 configuration, and random substitutions of atoms of lower valence, primarily in the alumina sheet, lead to a net negative charge in the 2:1 structure (Olphen, 1977). This net negative charge is balanced by counterbalancing cations that exist between each of these sheets, and subsequent hydration and dehydration of these ions and the negatively charged silica sheets leads to an overall expansion and contraction of the structure. This occurs over two distinct ranges as measured by X-ray diffraction (XRD): 1) crystalline swelling, where layers of water hydrate the cations leading to discrete increases in the d -spacing, and 2) osmotic swelling, a more continuous expansion where the driving force for swelling is a concentration gradient between the clay sheets and the bulk pore solution (Norrish, 1954; Olphen, 1977; Madsen and Müller-Vonmoos, 1989). In the case of heritage stones, other authors have suggested the possibility of disjoining pressures also leading to swelling (Ruedrich et al., 2011) in certain stones which do not contain swelling clays. Previous results suggest that even upon exposure to ambient relative humidities, sandstones with clays are swelling primarily through crystalline swelling (Wangler and Scherer, 2008; Colas et al., 2011; Wangler, 2016). More specifically, in the first of these studies it was demonstrated that

in three American sandstones, including Portland brownstone, the swelling strain could be directly related to the d -spacing as measured by XRD, as depicted in Figure 1.

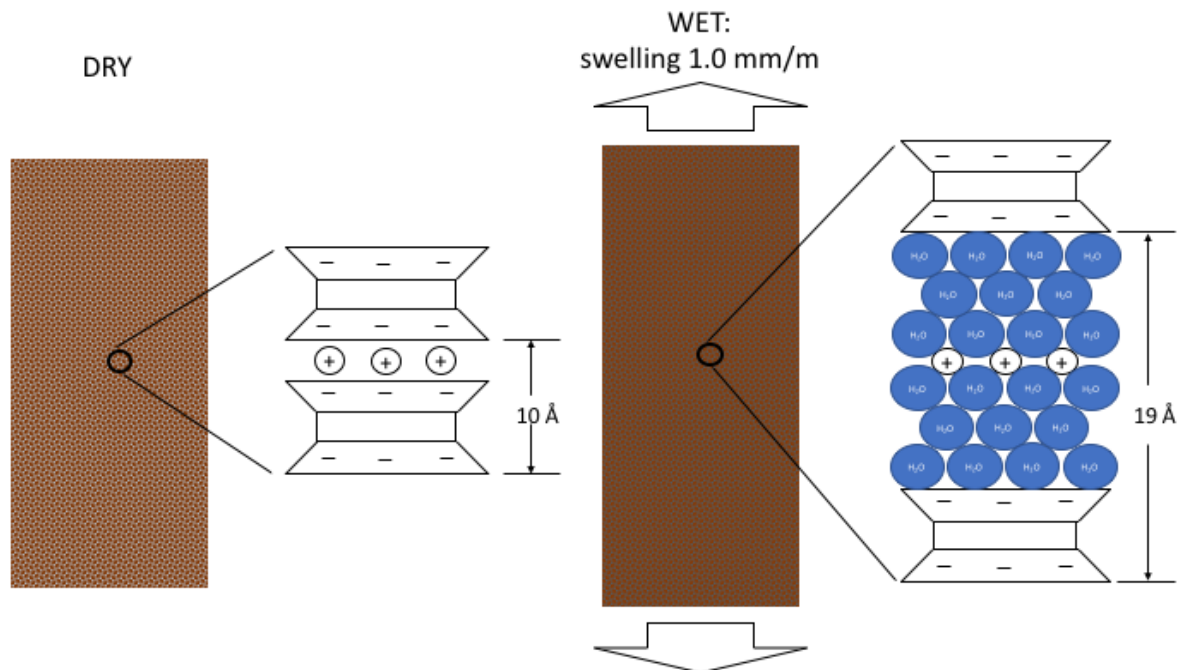


Figure 1. Relationship between swelling strain and clay moisture content in Portland Brownstone and Pennsylvania Bluestone, based on Wangler and Scherer (2008).

Of particular note is the role of the microstructure in swelling of stones. Clays that exist between grains are of far more consequence than clays found within pockets, and this has large consequences for the mechanical behavior of the stones when either wet or dry (Gonzalez and Scherer, 2004; Ruedrich et al., 2011; Tiennot et al., 2019).

Villarlod molasse is a stone of high importance to the cultural heritage of Switzerland, and variants of the molasses in the Swiss plateau are widespread throughout Switzerland and southwest Germany. The molasse has been noted for poor weathering performance, mostly attributed to the generally high swelling clay content (Félix, 1994; Gonzalez and Scherer, 2004). Recent studies on molasse have focused on the details of its fracture mechanism (Tiennot and Bourguès, 2016) and on-site performance (Demoulin et al., 2016). The swelling

clays have also been pointed out as problematic in application of consolidation strategies for this stone, rendering treatments ineffective after just one or two wetting cycles. Thus understanding the swelling behavior and how it leads to damage is of prime importance for the built cultural heritage of Switzerland. In this study, the focus is primarily on the details of the swelling mechanism, and a similar approach is taken as the study of Wangler and Scherer (2008) to uncover the relationship between d -spacing and swelling strain, as seen in Figure 1, for Villarlod molasse.

2. Materials & Methods

2.1 Materials

2.1.1. Stone

The stone used in this study was Villarlod molasse, a clay-rich subarkose sandstone quarried from Carrière de Molasse de Villarlod, Farvagny, Switzerland. Stone is cut from the quarry in large blocks, then samples are cut for swelling experiments using diamond saws down to prisms 5 cm in height and 5 x 5 mm in cross section. The swelling strain in water of the Villarlod molasse used in this study was 1.8 mm/m. The anisotropy of the stone was taken into consideration, and for all samples the bedding planes were oriented perpendicular to the swelling direction.

2.1.2. Cation pretreatments

For cation pretreatments, solutions of 2.0 M were prepared of various chloride salts (Na^+ , K^+ , Mg^{2+} , and Ca^{2+}). The stone samples were pretreated by immersion in the solutions for 12 hours, followed by washing with stirred deionized water baths for 18-24 hours.

2.1.3. Clay fraction

The clay fraction from the Villarlod molasse was separated according to USGS OFR 01-041 (Poppe et al., 2000). The procedure includes first a manual grinding, followed by an acetic acid digestion of the carbonate phases. The remaining solid is then put through a series of

centrifugation steps until only the clay sized fraction (<2 mm) remains suspended. This suspension is then flocculated with a 1.0 M CaCl₂ solution and centrifuged, with the solid fraction dried in an oven.

2.2. Experimental Methods

2.2.1. X-ray Diffraction (XRD)

XRD experiments were all performed on a Bruker AXS D8 ADVANCE diffractometer (Bruker Corporation, Karlsruhe, Germany) with CoK α ($\lambda = 1.7902 \text{ \AA}$) radiation. Two types of experiments were performed: quantitative Rietveld analysis of the whole stone, and *d*-spacing measurements on the oriented clay fraction under various conditions.

Rietveld analysis, reported elsewhere (Wangler, 2016), was performed on a randomly oriented sample of stone powder, ground and sieved to <20 μm and placed in a front-loading sample holder, using a razor blade to reduce preferred orientation. Scanning was performed between 2° and 80° 2θ at step sizes of 0.02° and 4 second per step. Qualitative analysis to identify peaks was performed using DIFFRACPlus EVA software by matching peak locations and intensities to the database of the International Centre for Diffraction Data (ICDD). AutoQuan software (GE Inspection Technologies) was performed for quantitative mineralogical phase analysis, using Rietveld refinement to adapt phase contents and phase-specific parameters to minimize the difference between a calculated and the measured diffractogram.

Oriented clay fraction XRD measurements were performed by first applying a thin slurry of the clay-sized fraction on a glass slide and then allowing it to dry. Glass slides were then treated using various solvents: water and methanol, applied by an eyedropper directly before scanning, and ethylene glycol and 1,3 propanediol, applied by placing the slides in a dessicator with each of these solvents, overnight, in a 60 degree oven. Diffractograms were performed from 1° to 30° 2θ at 0.02° steps, with 1 second dwell time. The scan time of about

25 minutes was too long to allow measurement of the methanol spacing due to its volatility, and thus this is excluded from the results.

2.2.2. Exchangeable cations

These experiments were carried out using a procedure adapted from (Meier, 1999). A copper pentaethylenehexamine (Cu pent(en)) solution of 0.01M was prepared from copper sulfate and pentaethylenehexamine. The solution was then contacted with a measured amount of the stone that had been crushed by a mortar and pestle and allowed to equilibrate in an agitated suspension for 1 h. The suspension was then centrifuged and decanted. The supernatant was diluted 1:100 with a 2wt% HNO₃ solution before analysis by inductively coupled plasma optical emission spectroscopy (ICP-OES) for the following ions, expected to be the dominant exchangeable species: K⁺, Na⁺, Ca²⁺, and Mg²⁺.

2.2.3. Swelling experiments

Swelling experiments were carried out with a Linear Variable Differential Transducer (LVDT) obtained from TESAGroup (France) with a resolution of $\pm 0.2 \mu\text{m}$. The LVDT tip was contacted to the surface of the sample in a small stainless steel cup, which was then filled with water or an organic solvent. The swelling strain was measured after approximately 60-90 minutes by dividing the measured linear deformation by the length of the sample. In one experiment, the water or solution surrounding the sample was suctioned out and either replaced with another solution, or the sample was dried with a compressed air gun.

3. Results

The Rietveld analysis of Villarlod molasse is seen in Table 1. The swelling clay fraction is on the order of 5-6%, and also noteworthy is the carbonate fraction of the rock, with calcite at 17% and dolomite at 3%.

Phase	% by mass	Error
Quartz	43.55	0.72
Calcite	17.10	0.36
Plagioclase albite	14.64	0.69
Microcline	6.63	0.42
Dolomite	3.19	0.45
Hematite	0.20	0.11
Muscovite	4.13	0.45
Chlorite	4.82	0.57
Smectite	5.75	1.17

Table 1. Rietveld analysis of Villarod molasse, from (Wangler, 2016).

The exchangeable cations as measured by the release of Ca^{2+} , Mg^{2+} , K^{+} , and Na^{+} are seen in Table 2. The dominant exchangeable cations of these four are Ca^{2+} and K^{+} . The CEC that can be estimated from these results is 7.44 meq / 100 g.

Ion	Exchangeable cation content (mmol/100 g stone)
Ca^{2+}	1.39
Mg^{2+}	0.27
Na^{+}	0.04
K^{+}	4.07

Table 2. Exchangeable cations of Villarod molasse. A CEC of 7.44 meq / 100 g stone can be estimated by multiplying the charge of the ion and the total ion content, and summing across all ions. Error of ± 0.02 for all values.

Table 3 shows the results of the swelling experiments with cation pretreatments. Noteworthy results from this include the relatively lower amounts of swelling observed with K^+ and Mg^{2+} . A higher swelling amount is observed with Na^+ , while Ca^{2+} swelling strain is more in line with that observed for the untreated stone.

Ion pretreatment	Swelling strain (mm/m)
None	1.8
Ca^{2+}	1.85
Mg^{2+}	1.15
Na^+	2.15
K^+	0.65

Table 3. Swelling experiments with cation pretreatments, all values subject to an error value of ± 0.02 mm/m.

Figure 2 shows the swelling strains for various organic solvents, while Figure 3 shows the corresponding XRD results. The drying rate of methanol was too high to allow for the accurate measurement of the d -spacing of the clay fraction with methanol, therefore a reference value of $14.1 \text{ \AA}$ taken from German and Harding (1971) is taken. One can see that ethylene glycol's layer spacing is $16.5 \text{ \AA}$, a bit lower than the usual layer spacings observed in swelling clays ($16.9\text{-}17.1 \text{ \AA}$), but lower ethylene glycol layer spacings have been noted in higher layer charge swelling clay minerals (Brindley, 1966). Difficulties in timing the drying of the water saturated clay fraction made a good scan difficult to procure, but a diffuse peak towards $19 \text{ \AA}$ (indicative of the upper limit of Ca-montmorillonite swelling) is observed.

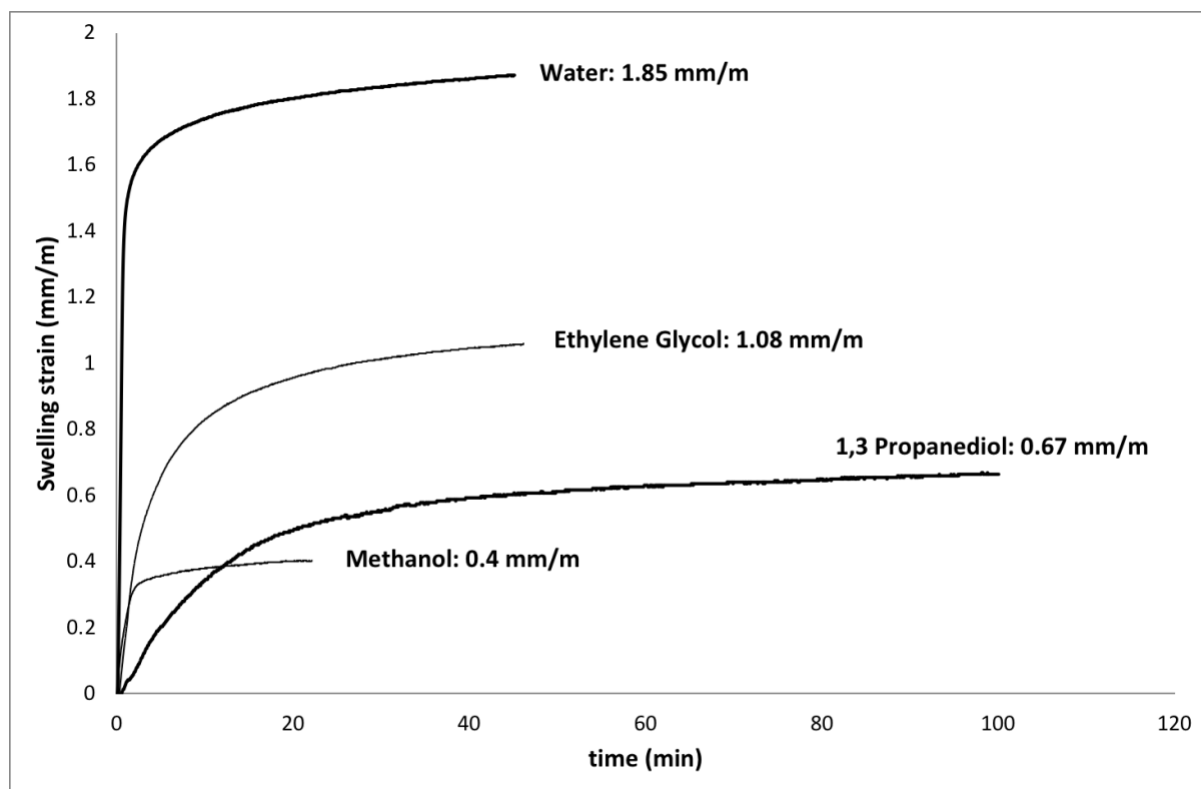


Figure 2. Swelling strain of Villarlod molasse with various organic solvents and water.

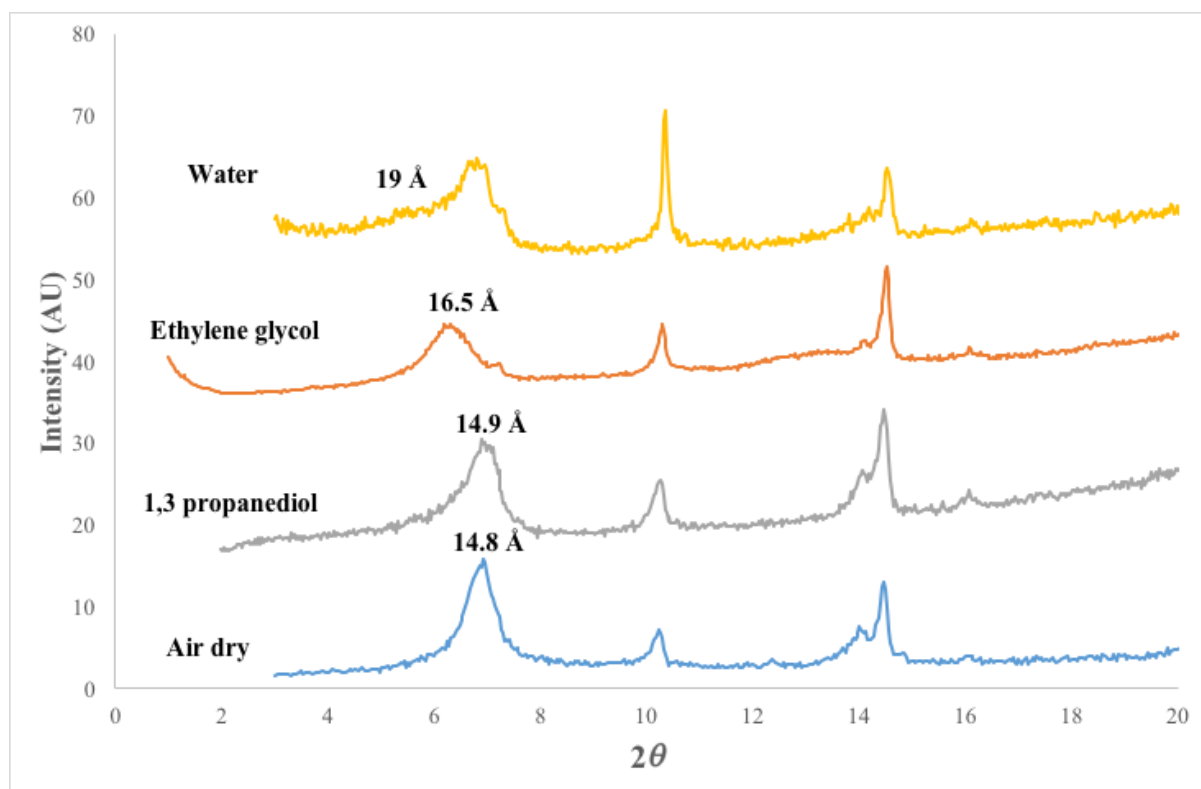


Figure 3. Oriented XRD with various organic solvents, with relevant d -spacing marked.

4. Discussion

In the earlier study of Wangler and Scherer (2008) a scaling factor was developed relating swelling strain measured by dilatometry, and d -spacing obtained from XRD:

$$\frac{\epsilon_{s,2} - \epsilon_{s,1}}{d_{001,2} - d_{001,1}} \quad (1)$$

Where ϵ_s and d_{001} are the measured swelling strains and d -spacings at different conditions.

With this relationship, one could easily determine the d -spacing of the clays confined within the stone with a simple dilatometric experiment. This relationship can be modified to

$$\frac{\epsilon_s}{d_{001,final} - d_{001,initial}} \quad (2)$$

where $d_{001,initial}$ corresponds to the initial d -spacing before a swelling experiment, and ϵ_s and $d_{001,final}$ correspond to the measured swelling strain and final d -spacing, respectively, for a given swelling experiment.

This relationship arose due to the direct comparison of XRD and dilatometric experiments, which showed an approximate doubling of the swelling strain when comparing methanol to ethylene glycol swelling. This corresponded to an approximate doubling of the interlayer space from an increase of 4.1 Å (methanol) to 7.1 Å (ethylene glycol). A key conclusion from this study was that in addition to the swelling range apparently being entirely crystalline, the initial d -spacing before any kind of wetting in the stones of the study ($\epsilon_{s,1}=0$) was 10 Å, which corresponds to a completely dehydrated interlayer. This result was unexpected, as usually, there are one or more layers of water in a clay interlayer at ambient relative humidities; for example, most studies show a bilayer at ambient relative humidities in Ca-montmorillonite (Méring, 1949; Tamura et al., 2000). The dehydrated interlayer was hypothesized to be due to the fact that the rock matrix exerts a pressure on the clay interlayers at grain interstices, forcing the water out.

The swelling behavior described in the previous paragraph is not observed in this study with Villarlod molasse, however. The ethylene glycol swelling strain is almost triple that of the methanol swelling strain, which does not correspond proportionally to an increase in interlayer spacing from an initially dry clay interlayer. However, when one assumes that there is a monolayer of water in the clay interlayer, then the proportional increases that are subsequently calculated make sense. From Figures 2 and 3, if one assumes an initial layer spacing of approximately 12.5 Å, then one can see that the increase to 14.1 Å for methanol, and 16.5 Å for ethylene glycol (as well as the spacing of 14.8 Å for 1,3 propanediol) can then be related well to their respective swelling strains through (2). For example, the relative increase for methanol would be 1.6 Å, corresponding to 0.42 mm/m in swelling, giving approximately 0.26 mm/m-Å as a scaling factor relating the two. A similar analysis for ethylene glycol gives 0.27 mm/m-Å. This is summed up in Table 4, and leads to a similar conclusion to the study of Wangler & Scherer (2008), that swelling in this particular stone is predominantly crystalline in nature. It is clear to see in Table 4 that assuming a completely dry interlayer leads to a systematic error, which is immediately corrected when one assumes that there is a single monolayer of water in the interlayer. The agreement among the four solvents that this correction makes gives very compelling evidence for the accuracy of the scaling factor, as well as the accuracy of the assumption of the initial *d*-spacing. Considering the difficulty in measuring the actual initial *d*-spacing within the stone, this method could be quite useful. However, this rests on the assumption that the swelling behavior of the clay within the stone while saturated with each solvent is the same as the saturated swelling behavior outside of the stone during an XRD experiment.

Solvent	swelling strain (mm/m)	$d_{001, \text{final}}$ (Å)	scaling factor	
			$d_{001, \text{initial}}=10 \text{ Å}$	$d_{001, \text{initial}}=12.5 \text{ Å}$
Water	1.85	19	0.2	0.28
Ethylene glycol	1.08	16.5	0.17	0.27
1,3 propanediol	0.67	14.9	0.14	0.28
Methanol	0.41	14.1*	0.10	0.26
Average				0.27

Table 4. Swelling layers / mm calculation for Villarlod molasse, relating swelling strains measured via dilatometry with d -spacings measured via XRD ($d_{001, \text{final}}$). Scaling factor shows two columns, one calculated with $d_{001, \text{initial}}$ at 10 Å vs 12.5 Å. *data obtained from (German, 1971).

Irrespective of the saturated swelling behavior, it is clear, however, that the d -spacing of the clay interlayers at ambient RH when measured outside of the stone mass (14.8 Å, Figure 3) and the calculated d -spacing at ambient RH when within the stone mass (~12.5 Å) do not match. This supports the earlier evidence from Wangler & Scherer (2008) that there is an external pressure applied from the rock mass on the clay interlayers, acting to keep water out at ambient RH. Evidently, in the case of Villarlod molasse, this external pressure is just high enough to keep out a bilayer of water, but a monolayer is possible. Ambient RH is generally in the range of 40-60%, and experimental results show that Ca-montmorillonite converts from a monolayer to a bilayer at RH no higher than 10% (Tamura et al., 2000), so the external pressure is lowering the effective RH to <10%.

Building upon the idea that the stone mass creates an external pressure on the clays, a microstructural model of the stone can be imagined in which the sandstone consists of grains where some have swelling clays, and some do not, and as described earlier, the nonswelling grain junctions exert a stress on the grain junctions that swell, as seen in Figure 4a. The

nonswelling junctions can be imagined as a spring, as depicted in Figure 4b for the case of Villarlod molasse, and this spring exerts the opposing stress for the entry of water into the clay interlayer. This stress is apparently not enough to remove the final interlayer of water in Villarlod molasse at ambient relative humidities. Additionally, this stress also does not seem to play a large role in the saturated state using various solvents, as described earlier, although further work will need to confirm this.

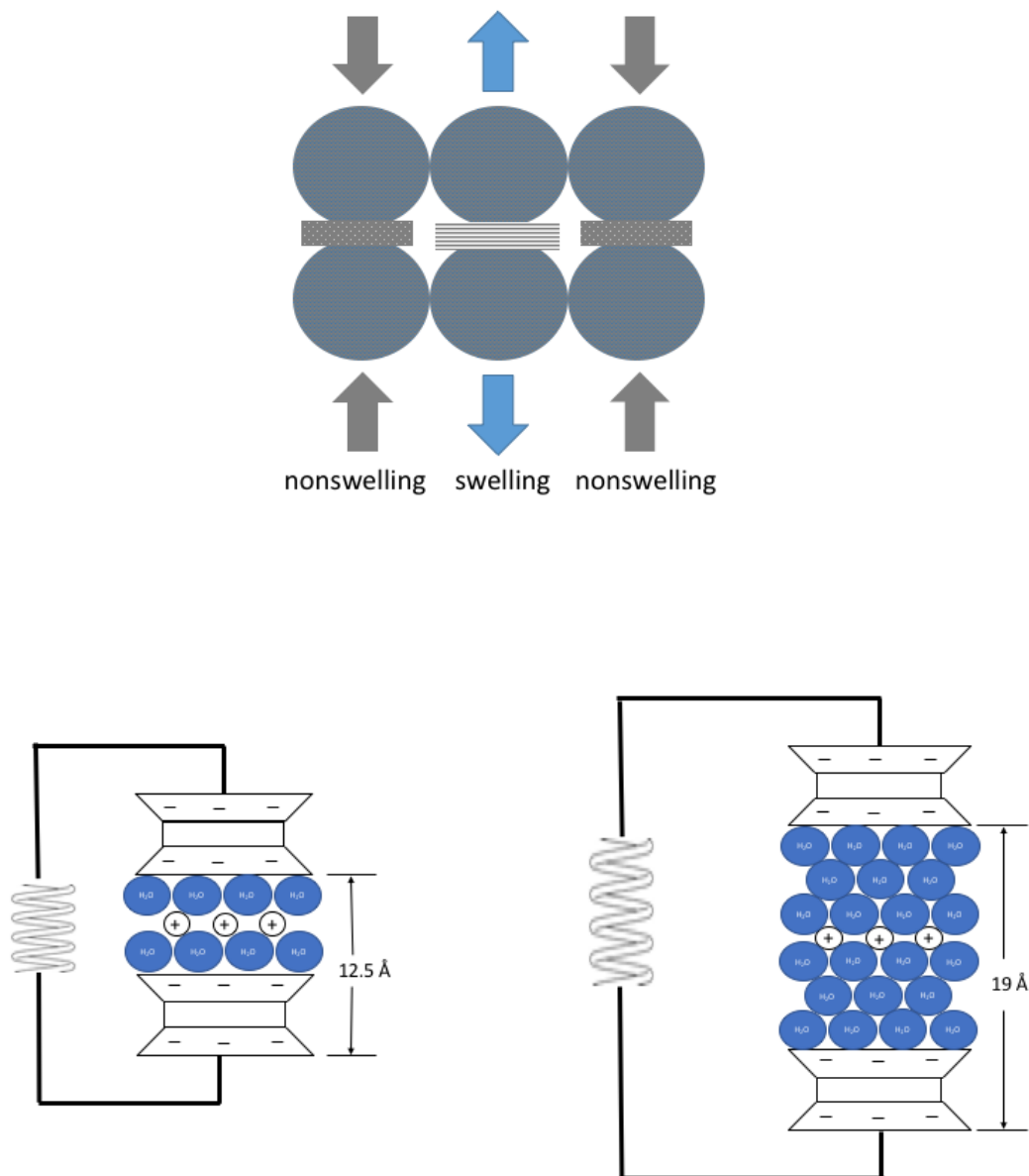


Figure 4a-b. (top) Microstructural model of sandstone in which nonswelling parts of the stone resist the swelling of the junctions with swelling clays. The nonswelling parts can be imagined as a spring which exerts a stress on the swelling clays (bottom).

The results of the cation pretreatment experiments support the implication of the previous analysis, in that swelling of Villarlod molasse with K^+ , which forms an “inner sphere” hydrate, has a significantly reduced swelling strain. The behavior of the untreated stone compared to stone pretreated with Ca^{2+} indicates that Ca^{2+} is most likely the predominant cation, which is reasonable to expect considering the calcitic matrix. The results of the exchangeable cation analysis (Table 2), however, show a significant proportion of potassium as the exchangeable cation. This likely means that calcium and potassium are intermixed in the interlayers, with calcium dominating the swelling behavior. The potassium interlayer cation source is possibly altered microcline (Saigal et al., 1988), as both XRD and SEM-EDX show significant amounts of microcline and plagioclase albite (Wangler, 2016; Wangler and Sanchez, 2020). In any case, this underscores the importance of the cation species in understanding the behavior of swelling clay minerals in sandstones.

With the scaling factor of $0.27 \text{ mm/m-}\text{\AA}$, one can then easily calculate relative layer spacing increases for various cations based on their swelling strains in water, summarized in Table 5.

Ion pretreatment	Interlayer spacing change
	$\Delta d \text{ (\AA)}$
None	6.8
Ca^{2+}	6.8
Mg^{2+}	4.1
Na^+	8.0
K^+	2.4

Table 5. Layer spacing increases for various cations, calculated with scaling factor of $0.27 \text{ mm/m-}\text{\AA}$.

One can see that some layer spacing changes are very large, such as sodium's, and some are very small, such as potassium's. Crystalline swelling does not really proceed above a 10 Å layer spacing increase (Madsen and Müller-Vonmoos, 1989), and therefore some of these calculated layer spacing increases result from either a lower initial layer spacing (indicating some water has been forced out) or a certain degree of osmotic swelling, or both. Looking at the case of potassium, the layer spacing increase of about 2.4 Å indicates a monolayer of water is entering the interlayer. However, whether this is happening from an initially dry state (and thus the layer is the first monolayer) or from the monolayer to the bilayer state, cannot be determined from these data. Concerning the higher increase in the sodium treated stone, it is possible that this range corresponds to the full range allowable by a dehydrated interlayer, because a monolayer spacing of water added to the calculated interlayer spacing change is 10.5 Å, which is a bit above the upper limit for crystalline swelling. This would mean that similar to the sandstones studied by Wangler and Scherer (2008), the rock matrix is able to successfully push out the monolayer of water at ambient relative humidities. Considering that the hydration energy of Na^+ is weaker than Ca^{2+} , this is a plausible explanation, but it does not rule out the possibility that there is already a monolayer of water and there is a certain amount of osmotic swelling occurring. It is also interesting to note that apparently Mg^{2+} has initially a bilayer of water in the interlayer at ambient conditions. This contrasts with the behavior of Ca^{2+} , also divalent. This could be because of the stronger hydration energy of Mg^{2+} relative to Ca^{2+} , which would attract a bilayer of moisture at ambient relative humidities (Salles et al., 2013). In any case, further studies with solvents and cation pretreatments should lead to a better understanding, in particular on the initial d -spacing. It bears noting that the degree of exchange in the clay of the various cation pretreatments is unknown. It is therefore quite possible that in any of the experiments, there is an incomplete degree of exchange, and thus the swelling behavior is reflective of mixed interlayers.

However, one can state quite uncategorically that the ion pretreatments have a large effect on the swelling behavior, which again points to crystalline swelling being the primary swelling mode. Additionally, general trends such as those noted above can be determined as in agreement with what is known on cation effect on crystalline swelling in smectites.

An interesting experiment to confirm the existence of interlayer water at ambient conditions was carried out, in which a dilatometric experiment was carried out by swelling initially with water, then replacing the surrounding fluid with a 1.0 M KCl solution, followed by drying and then rewetting. This experiment is seen in Figure 5, in which a Villarlod molasse stone with an Na^+ pretreatment is subjected to this procedure.

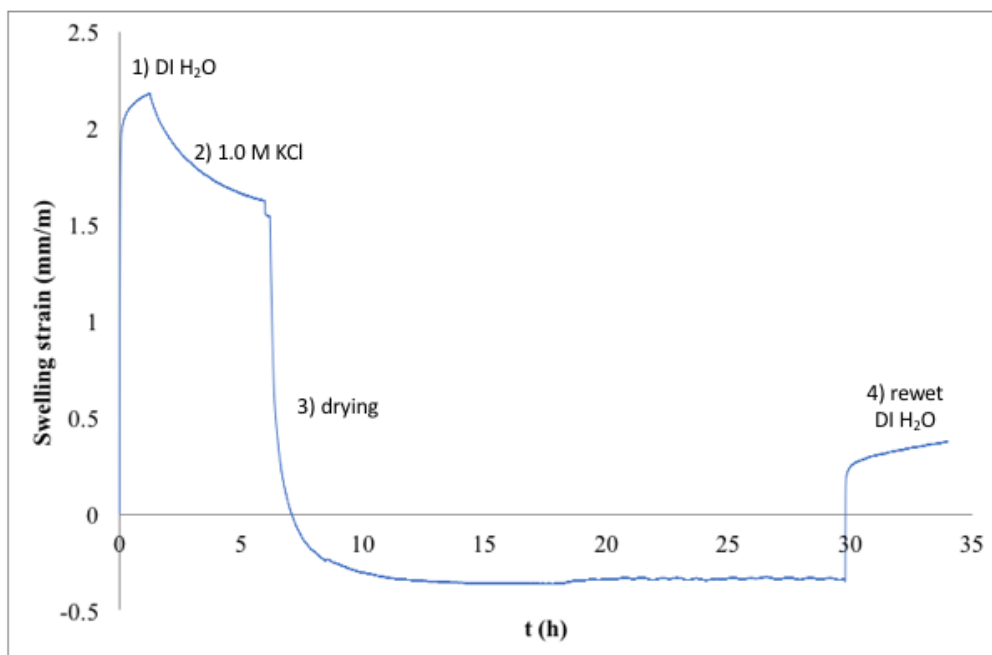


Figure 5. Experiment in which a Na^+ treated sample of Villarlod molasse is first swelled with DI water (1), then the water is suctioned out and replaced by a 1.0 M KCl solution (2) which diffuses into the sample and causes a contraction. Subsequent drying (3) then causes the sample to contract to a length shorter than its initial length, indicating a change in initial interlayer spacing in the clay. Upon subsequent rewetting (4) the sample swells to the swelling strain of a K^+ treated sample.

In this experiment, one can observe the initial swelling with water followed by a contraction as the potassium diffuses into the sample and into the clay interlayers. Upon drying, the sample then contracts, as expected, but it contracts to a dimension less than its initial dimension, indicating a contraction of the clay interlayers relative to the initial *d*-spacing. This experiment provides clear evidence that there is some moisture in the interlayer initially, and that the full exchange of a weaker hydration energy cation (K^+) allows the matrix of the rock to remove this interlayer water when drying occurs. This is noteworthy, as earlier it was suggested that sodium might allow the moisture in the interlayer to dehydrate fully at ambient RH, but this is apparently not the case. Finally, subsequent wetting then matches the measured swelling strain of the potassium treated stone in Table 3 of approximately 0.65 mm/m.

There are some implications of this work for the conservation of built cultural heritage constructed with Villarlod molasse, primarily when it comes to understanding and mitigating the swelling behavior. The fact that the swelling is predominantly crystalline points again to treatment strategies that target this, such as ion exchange with species that induce lower swelling. The most effective anti swelling agents to date have been the α , ω alkyldiamines, first tested by Snethlage and Wendler (1990) and the mechanism elucidated by Wangler and Scherer (2009). The mechanism of swelling reduction in Villarlod molasse, performed similarly to the previously cited study of Wangler and Scherer, will be the subject of another study. However, the possibilities of a species such as potassium, which is much cheaper and appears to be able to reduce swelling significantly, have not yet been explored in the context of cultural heritage. Besides understanding its efficacy, however, the use of potassium must also be examined in terms of the amount required and the durability of the treatment. A treatment such as this, however, or a proven one such as the alkyldiamine treatment, could prove useful if done as a pretreatment before a restoration.

5. Conclusion

The results of this study demonstrate that for Villarlod molasse:

- The clays responsible for moisture expansion are swelling almost exclusively by crystalline swelling, although other swelling modes could contribute in a minor way. This corresponds closely with earlier results suggesting that in many sandstones with swelling clays, crystalline swelling is the dominant mode.
- The stone swelling strain can be linked to the clay swelling behavior on the Å scale, with a scaling factor calculated at 0.27 mm/m-Å. This can be utilized to calculate changes in the clay interlayer spacing via simple swelling experiments. It should be cautioned that clay content may vary within stone taken from the same formation, so the scaling factor calculated here is not necessarily fixed, and experiments performed should serve as a framework for characterizing swelling in Villarlod molasse and other stones.
- At ambient relative humidities, a monolayer of water exists in the clay interlayer within the stone matrix, while a bilayer exists in the separated clay fraction. It is likely that the reason for this discrepancy is the stress imposed on the clay interlayer by the rock matrix, which forces out one layer of water.
- The interlayer cation makeup is primarily Ca^{2+} and K^{+} , with Ca^{2+} dominating the swelling behavior. The interlayer cation has a very large effect on the swelling behavior, as one would expect with crystalline swelling being the dominant mode.

Further points of investigation then can be recommended:

- The role of external pressure on a swelling clay interlayer should be further investigated, especially as it relates to variable moisture contents. The combination of moderate external pressures and lower relative humidities has not been studied a lot,

and this unique environment exists in a stone matrix at relatively dry (ambient) conditions.

- The micromechanical model of clays at interlayer junctions and their impact on global mechanical properties can be further investigated and developed, which will be the topic of further study.
- The common swelling inhibitors used in cultural heritage conservation (and also used in swelling inhibition for drilling borehole stability) can be studied using the techniques and relationships developed in this study. This will be the topic of another study for the specific case of Villarlod molasse and alkyldiamines.

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