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### ▶ To cite this version:

Fabien Cherblanc, Jérémie Berthonneau, Philippe Bromblet. Role of hydro-mechanical coupling in the damage process of limestones used in historical buildings. 13th International Congress on the Deterioration and Conservation of Stone, Sep 2016, Glasgow, United Kingdom. hal-01364003

HAL Id: hal-01364003

https://hal.science/hal-01364003

Submitted on 12 Sep 2016

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# ROLE OF HYDRO-MECHANICAL COUPLING IN THE DAMAGE PROCESS OF LIMESTONES USED IN HISTORICAL BUILDINGS

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#### **Abstract**

Historical monuments of the Provence region display different sensitivities to spalling decay. Even if some phenomenological scenario have been proposed, the physical processes that govern this kind of damage have not been clearly identified and quantified. The more reliable hypothesis involves the hydro-mechanical behaviour of such limestones triggered by naturally occurring expandable clay minerals (smectite). In order to sense the internal factors involved, a characterization campaign has been carried out. Clay mineral quantification was performed based on a recent methodology coupling transmission electron microscopy, X-ray spectrometry, X-ray diffraction. The relevant mechanical properties were characterised with a particular attention paid on their dependencies on the water content. Then, permeability, capillary pressure, vapour diffusion, sorption isotherm, and hydric dilation of these stones were measured. This set of experimental characteristics was introduced in a numerical model of a block inside a masonry structure to simulate water transport and mechanical behaviour. Simple climatic scenarios have been imposed to simulate alternating wetting and drying boundary conditions. Numerical simulations showed that the mechanical stresses generated by hydric dilation or shrinking are not large enough to lead to a direct damage. Nevertheless, in a case study, numerical simulations highlighted some tendencies in agreement with observations.

**Keywords**: limestone, spalling decay, clay minerals, hydro-mechanical behaviour, numerical simulation

#### 1. Introduction

Roman and medieval historical monuments of the Provence region (South East of France) erected with limestones are affected by particular stone deterioration patterns called "spalling" and "splintering" (Vergès-Belmin 2008). Spalling corresponds to the progressive loss of plates from surfaces exposed to rain and/or runoff whatever their position in the building. The plane of detachment is located near the stone surface and develops perpendicularly to the bedding so that it cannot result from mechanical weaknesses linked to the anisotropy of these sedimentary stones (Berthonneau *et al.*, 2014, 2015). Splintering

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corresponds to the detachment of sharp and slender pieces of stone broken off from the main body.

Even if several phenomenological scenario have been proposed, the physical processes that govern this kind of damage have not been clearly identified. One reliable hypothesis involves the hydro-mechanical behaviour of such limestones triggered by naturally occurring expandable clay minerals (smectite). Indeed, among clay minerals, the smectite group is very sensitive to water and is subject to a significant tendency to swell. Under cyclic conditions of wetting and drying, swelling and shrinkage phenomena can play a major role in the deterioration processes (Jiménez-González *et al.*, 2008; Ruedrich *et al.*, 2011; Berthonneau *et al.*, 2014). Numerous studies on stone conservation point out the swelling clay content to be a key factor in durability evaluation of sedimentary stones (Weiss *et al.*, 2004; Franzini *et al.*, 2007; Sebastián *et al.*, 2008; Berthonneau *et al.*, 2014).

While this description of stone damage seems to be well accepted, the relative roles of underlying physical phenomena need to be quantified. In particular, several studies refer to mechanical interpretation such as "differential stress" (Doehne and Price 2010) or "buckling of wetted surface" (Jiménez-González *et al.*, 2008; Wangler *et al.*, 2011) without being supported by unambiguous results. At this point, the objectives are to investigate this hypothetical process of degradation from a physical approach by proposing a numerical modelling. In the framework of this work, the focus has been put on two historical monuments: the Roman amphitheatre in Nîmes and the church St Maurice in Caromb.

#### 2. Experimental characterization of limestone

Five different types of limestone are under investigation (ES: Estaillades, BM: Modern Barutel, BA: Antique Barutel, CY: Yellow Caromb, CG: Grey Caromb) all coming from the south region of France. While Barutel stone is a fine grained limestone of Barremian age (129 -125 Ma), Caromb and Estaillades stones belong to the "Pierre du Midi" type, a Burdigalian (20-16 Ma) deposit extensively used for building since antiquity. Materials (BA, CG, CY) were sampled on deposits of ancient stones coming from dismantling of old buildings (Nîmes amphitheatre, Caromb church) and BM, ES were taken from active quarries. Additional details can be found in Berthonneau *et al.* (2014, 2015), Cherblanc *et al.* (2016).

#### 2.1. Clay content quantification

Mineralogical characterization was conducted by calcimetry (NF X 31-105) and X-ray diffraction (XRD) on total and acid (acetic acid 0.2 N) insoluble powders. Diffractograms were recorded on a Bruker D8 Focus diffractometer (CoK $\alpha$  radiation) equipped with a Lynx'Eye detector operating with an aperture of 1°2 $\theta$ .

Clay mineral identification and quantification were done on the clay fraction (<4 µm) through the combination of transmission electron microscopy (TEM, JEOL JEM 2011) coupled with an energy-dispersive X-ray spectrometer (EDS, X-Flash Silicon Drift Detector 5030, Bruker) and profile modelling of XRD patterns of oriented preparations. Details on the elemental quantification and structural formula calculation from TEM microanalysis can be found in Berthonneau *et al.* (2014). XRD patterns were collected on oriented preparations saturated with calcium (Ca) at 0.01426°20 step intervals from 2.5 to 60°20, using a 4 seconds counting time per step. A first set of XRD patterns was acquired after drying at room temperature (Ca + AD), then the same preparations were exposed to

ethylene glycol (Ca + EG) vapour for at least 24 hours and re-analysed. All the experimental XRD patterns were fitted over the 4.5 -  $60^{\circ}2\theta$  CoK $\alpha$  range using the ASN program.

Eventually, the quality of the simulation was estimated using the unweighted goodness of fit parameter  $R_p$ . This parameter was calculated over the 5 - 60°20 range, excluding the hkl reflections, and was used as an estimation of the error associated to the final quantitative values. The quality of the fits was mainly influenced by the low crystallinity of the phyllosilicates structures and the presence of strong hkl reflections of quartz. These mismatches explained the relatively high values of the goodness of the fit parameters ( $R_p$ ), especially for the Barutel stones (BA, BM). In order to account for it within the proposed quantitative models, the evolution of the diffracted intensity of the 101 reflections of quartz was studied as a function of its weight fraction in a mixture with pure Na-Montmorillonite (Swy-2). The results showed a linear trend which slope was used to provide a precise estimation of the quartz content of the analysed clay fractions. These results varied from 0.21 (CY) to 6.02 wt.% (BA) with an error of about 5%.

The total contents of each clay mineral can be found in Cherblanc *et al.* (2016). ES is a pure calcite rock, while the clay content varied from 1.7% to 2.6% in Caromb stones. One can note the significant amount of quartz in the clay fraction of Barutel stones. Since the smectite group (Beidellite and Montmorillonite) presents the larger specific area, it will have the more significant impact on the sorption behaviour this work will focus on it.

#### 2.2. Hygric/hydric properties

Water transport inside block occurs in both gaseous and liquid phases. Thus, the characterization of stone/water interactions includes the measurement of water permeability at saturation, vapour diffusivity, sorption isotherm curve, water retention curve.

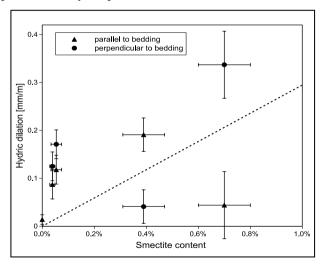


Fig. 1: Hydric dilation as a function of smectite content.

One major issue concerns the determination of hydric dilation coefficient. The principle relies on the measurement of the length variation during a capillary imbibition experiment. Since this property was only measured at saturation, it is supposed to depend linearly on the

water content. Regarding the 5 limestones under investigation, the hydric dilation measured at saturation is represented as a function of the smectite content in Fig. 1. Despite the weak correlation shown in this case, smectite content seems to be a key factor that governs swelling mechanisms.

#### 2.3. Mechanical properties

Main mechanical characteristics of samples were identified experimentally, including Young's modulus, Poisson's ratio, tensile and compressive strength. Each property was measured on several samples prepared at imposed water content to cover the full range of hydric state from dryness to water saturation. While plotted for CY limestone in Fig. 2, further data can be found in Cherblanc *et al.* (2016).

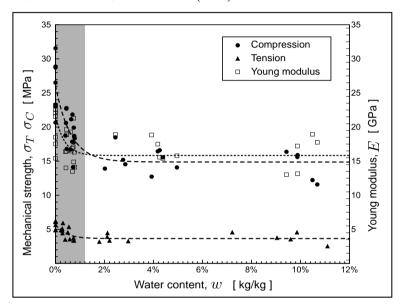


Fig. 2: Mechanical characteristics of CY limestone as functions of the water content.

As already evidenced, the mechanical properties of sedimentary rocks highly depend on the water content. It usually shows a strong decrease from dry condition to low water content and almost constant values for larger water content that can be described by a simple exponential representation based on 3 material parameters (Vásárhelyi and Ván 2006). It is worth noting that the range of water content where the decrease of mechanical strengths is observed (grey zone in Fig. 2) corresponds to the hygroscopic domain defined in sorption isotherm curves (Cherblanc *et al.*, 2016). It means that it is not necessary to undergo rain or flooding to observe the softening behaviour, a 97% *RH* atmosphere is sufficient to lead to a significant loss of mechanical properties.

#### 3. Numerical modelling

The state variables chosen to describe the phenomena are the water content and the solid phase strains. To describe water transport inside stone block in both gaseous and liquid phases, local equilibrium is assumed at liquid/gas interface (Ouedraogo *et al.*, 2013). Modelling is based on classical diffusive equations involving capillary phenomena in liquid

phase and vapour diffusion in gas phase. Using the small perturbations framework, the mechanical behaviour is considered elastic relying on Young modulus and Poisson ratio. A linear swelling term is added to represent hydric dilation phenomena. This theoretical description is fully coupled through the hydric dilation term that describes the influence of water transport on stress, and by means of the dependence of stone mechanical properties on water content (Fig. 2).

Numerical modelling is developed using the open source software LMGC90. Based on finite elements method, it allows simulating multi-physics processes. For physical consistency purposes, 2<sup>nd</sup> order discretization is used for mechanical phenomena and 2<sup>nd</sup> order for water transport. Temporal integration is performed using a fully implicit scheme to get numerical stability. To handle with non-linearities, a Newton–Raphson method ensures an accurate convergence for a moderate time step. Since we focus on the external part of stone blocks, a finely discretized mesh is taken in the first 10 cm from the outer surface while a coarser mesh is imposed elsewhere.

Since one block is embedded in a masonry wall, rigid boundary conditions is imposed on 5 faces. The outer face is free and subjected to climatic conditions, wetting or drying. To represent the gravity load from above building, a vertical background stress of about  $\sigma_b = 1$  MPa is imposed over the block. Finally, the damage level is analysed based on the failure criterion proposed by Hoek&Brown (Eberhardt 2012).

#### 4. Case study

Two case studies have been carried out, the 1<sup>st</sup> one relates to the Roman amphitheatre of Nîmes (Gard, France) while the 2<sup>nd</sup> one deals with the church St Maurice of Caromb (Vaucluse, France). Only this last case is presented below.

#### 4.1. The Caromb church

The church Saint Maurice of Caromb displays a good example of the different degrees of spalling. In particular, the grey Caromb limestone CG is much more sensitive to this type of decay mechanism than the yellow one CY, independently of any preferential cardinal orientation (Berthonneau *et al.*, 2014). While both materials are subjected to the same climatic conditions, some intrinsic stone properties should play a major role in the decay pattern. As proposed by Jiménez-González *et al.* (2008), hydric dilation can generate overstresses during wetting or drying processes. Thus, a simple climatic scenario is considered comprising 2 successive stages: first, a 2 hour wetting phase by imposing 95% of the water content at saturation over the outer face of the block, second, a 10 days drying phase by imposing a 20% *RH* atmosphere.

#### 4.2. Evolution of water content

Regarding CG material, simulated water content profiles inside a block are plotted in Fig. 3 for different time steps. In the first phase of wetting (0 < t < 7200 s), a water content front penetrates perpendicularly to the outer surface. After 2 hours of rain, a few centimetres are saturated by water. Then, during the second phase of drying  $(7200 < t < 100\ 000\ \text{s})$ , the water content near the outer surface decreases quickly while a slow diffusion process takes place inside the block. At low water content and in particular in the hygroscopic domain (grey zone in Fig. 2), unsaturated permeability lowers drastically and vapour diffusion becomes the dominant transport mechanism. Therefore, the drying condition imposed on the surface takes several days to spread through the block. These alternating wetting and

drying phases maintain an average water content above the hygroscopic state. Indeed, in situ mechanical characteristics are closer to water saturated values rather than dry ones (Fig. 2).

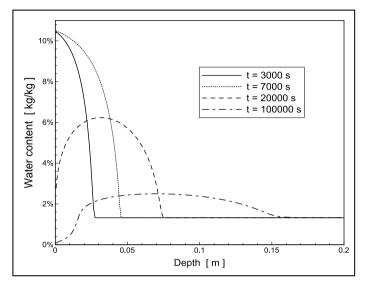


Fig. 3: Water content profiles inside CG block at different time steps.

#### 4.3. Development of mechanical stresses

Based on the climatic scenario simulated above, the most critical situation is encountered during the wetting phase. Since the block under investigation is embedded in the masonry wall, vertical strains are prevented. Thus, the swelling driven by water imbibition generates mechanical stresses perpendicular to the outer face in the thin layer saturated by water. To account for the triaxial stress state, the evolution of Hoek & Brown failure criterion is represented in Fig. 4. The grey spread of these curves represent the variability in mechanical characteristics that can be observed in Fig. 2. Indeed, CG limestone is more heterogeneous and a larger scattering of measures was obtained with CG samples than with CY ones.

Obviously, swelling stresses are not large enough to lead to the direct rupture of blocks and the risk level does not exceed 50% in the worst case (Fig. 4). Thus, the spalling decay should rather be associated with a progressive phenomenon. The variations in climatic conditions generate hydro-mechanical cycles that would lead to a slow damaging process occurring over years. Anyway, the swelling stresses that develops parallel to the outer surface are compatible with the failure mode observed where crack planes are perpendicular to the bedding plane and parallel to the outer face. This case study evidences that CG material is more sensitive to spalling decay than CY in agreement with in situ observations (Berthonneau *et al.*, 2014).

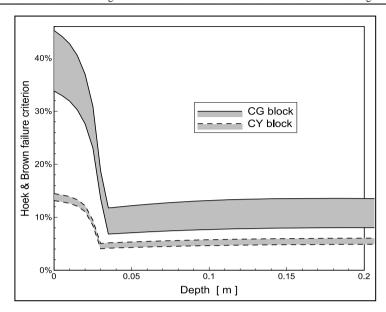


Fig. 4: Hoek & Brown failure criterion along the depth of stone blocks at t = 4000 s.

#### 5. Conclusions

Numerical simulations showed that the mechanical stresses generated by hydric dilation or shrinking are not large enough to directly lead to damage. Obviously, the spalling decay phenomenon takes place over a very long time representing many wetting / drying cycles and should rather be associated with a progressive fatigue process. Furthermore, several factors can act simultaneously (freezing, salt crystallization) and hydro-mechanical effects would be a speed up process. Nevertheless, in a case study on Caromb church where two kinds of stone are used, numerical simulations highlighted some tendencies in agreement with observations. These results constitute an encouraging first step toward a predictive approach for stone replacement campaign aimed at limiting the impact of spalling of historical building facades.

#### Acknowledgements

This work was done within the framework of a collaboration between the CICRP and the LMGC supported by the French Ministry of Culture and Communication through the PNRCC program (Programme National de Recherche sur la Connaissance et la Conservation du patrimoine culturel matériel).

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