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Moisture-induced elastic weakening and wave propagation in a clay-bearing sandstone

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Abstract

Long-term behaviour of a clay-bearing sandstone is investigated using elastic waves propagation coupled with strain measurements during moisture cycles. Adsorption and desorption are followed in a continuous way during several variations of relative humidity. Moisture adsorption is related to the solvation pressure and the measured elastic weakening during humidification is correlated with the clay minerals sensitivity.

1 Introduction

Monumental stone suffers from environmental conditions, especially from water saturation and moisture content variations [Doehne and Price, 2010, Siegesmund and Snethlage, 2011]. As a result of these natural solicitations, some specific alteration patterns can be related to the swelling ability and the moisture sensitivity, both governed by the microstructural distribution and mineralogical composition of the material [Franzen and Mirwald, 2004, ICOMOS, 2008]. Repeated over time, adsorption and desorption under natural environmental induce swelling strains, internal stresses and can modify the mechanical properties.

Indeed, several researches underlined the impact of water, liquid or vapour, on the compressive or tensile strength [Burshtein, 1969, Sarout, 2006, Van Eeckhout, 1976, Erguler and Ulusay, 2009] and the fracture toughness of various limestones, sandstones and granites [Utagawa et al., 1999, Nara et al., 2011, Kataoka et al., 2014]. Elastic properties evolution with saturation and moisture content has also been highlighted [Sarout, 2006, Vasarhelyi, 2003, Valès, 2008, Erguler and Ulusay, 2009, Pimienta et al., 2014, Tiennot et al., 2019]. The relationship between the evolution of the elastic moduli during adsorption and the deformation of the stone is of main interest to better understand elastic weakening [Yurikov et al., 2018]. Moreover, this phenomenon depends on the amount and nature of clay minerals within the stone, since they are more reactive to water [Nara et al., 2011, Nara et al., 2012, Funatsu et al., 2004].

To investigate the moisture-dependence of elastic behaviour, a non-destructive acoustic P- and S- wave measurement can be carried out. Thus, [Pimienta et al., 2014] and [Nakao et al., 2016] recently studied the influence of adsorption on carbonates, sandstones and granites behaviour thanks to ultrasonic waves propagation velocities measurements. Using this methodology, elastic properties of stones can be characterized as a function of moisture content.

In some rocks elastic weakening can be observed [Clark et al., 1980, Pimienta et al., 2014, Yurikov et al., 2018] and elastic properties variations can be explained by moisture adsorption [Tittmann et al., 1980, Murphy, 1982, Murphy III et al., 1984, Tutuncu and Sharma, 1992, Goertz and Knight, 1998]. To model the elastic weakening, [Yurikov et al., 2018] assumes that this weakening is the consequence of an additional term due to the pressure of the adsorbed fluid, called adsorption stress or solvation pressure. Based on the theory and the experimental protocol they describe, we verify whether this hypothesis is relevant for the Thüringer sandstone, and we estimate the impact of clay minerals on this phenomenon.

This research focuses on the monitoring of P- and S-wave propagation velocities in a clay-bearing sandstone, coupled to swelling strains measurement during moisture variations,

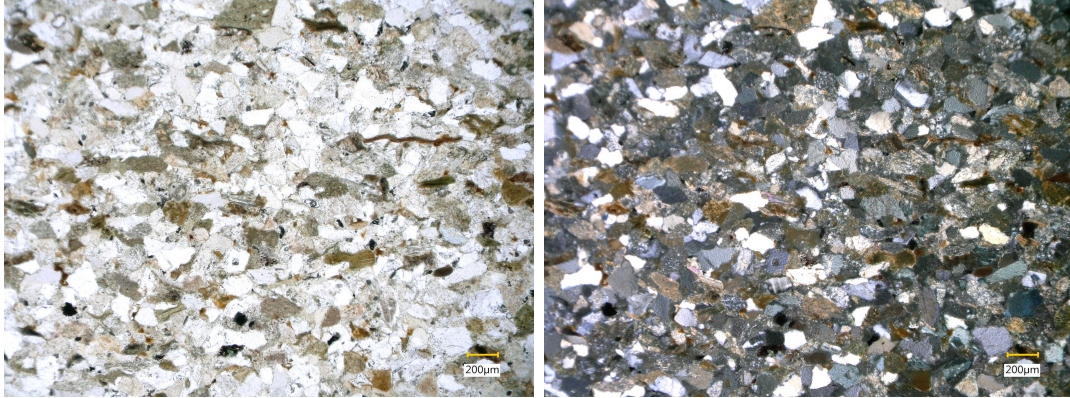


Figure 1: Observation of thin sections of Thüringer sandstone in natural light (top) and polarized light (bottom)

over repeated cycles. Identification and consequences of the adsorption and desorption processes in close correlation to the moisture sensitivity of the phases, especially the clay minerals is proposed.

2 Sandstone properties

2.1 Sampling

The samples were cored from a single Thüringer sandstone block. This sandstone is a clay-bearing sandstone from the Superior Trias. The block was obtained from a working quarry located in Germany. Plugs of 40 mm in diameter and 80 mm in length were cored in the direction perpendicular to the natural bedding plane of the rock, corresponding to the preferred orientation of the clay minerals. After coring, the faces were ground to ensure a good parallelism.

2.2 Mineralogical and structural properties

Rock composition is investigated and clay minerals identification is performed by X-Ray Diffraction (XRD) using a Bruker D8 Advance[®] Bragg-Brentano diffractometer on the extracted clay fraction. Optical microscopy and observation under FESEM on fresh fracture surfaces are proposed to confirm the minerals' identification. Finally, the total open porosity and the pore size distribution are quantified using Mercury Injection Porosimetry (MIP) [ASTM, 2010].

Mineralogical phases identified are coarse-grains of quartz and feldspars associated with muscovite and biotites (Fig. 1). The clay phases in this rock are chlorite, illite and mixed-layered chlorite-smectite (Fig. 2). The estimated mineral compositions for this sandstone are 55 % of quartz, 25 % of feldspars and 20 % of clay minerals. The connected porosity is measured at 13.4 (0.80) % with a bimodal pore size distribution. The first set of pore access radius centered at 0.02 μm is associated with the clay porosity while the second set at 1.5 μm is associated with the inherent cracks porosity (Fig. 3). The bimodal porosity is also visible in Figure 2.

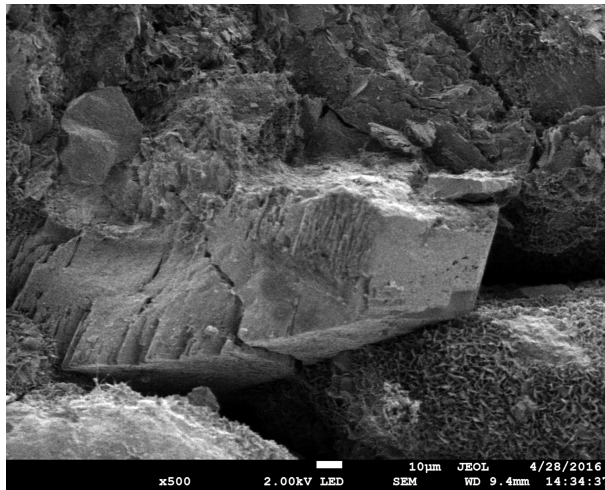


Figure 2: FESEM observation of the Thüringer sandstone: feldspars and chlorite distribution creates a bimodal porosity

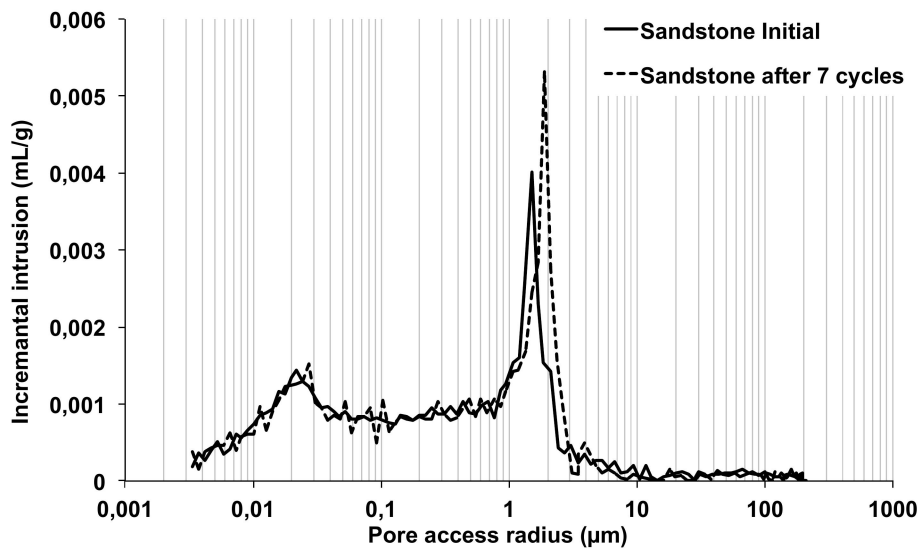


Figure 3: Pore size bimodal distribution of the Thüringer sandstone at the initial state and after 7 cycles of hygric sollicitations

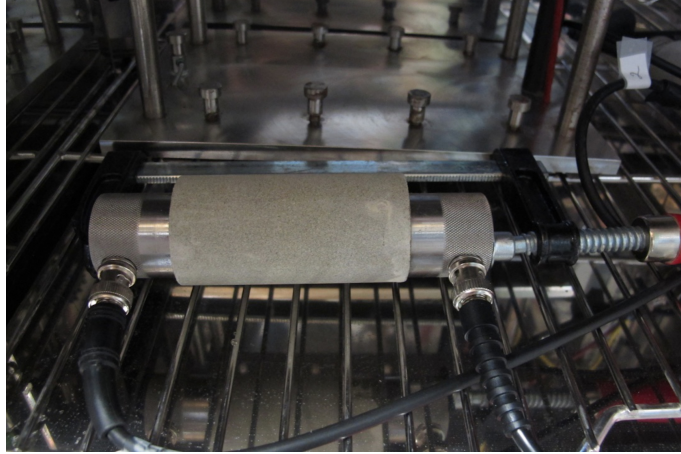


Figure 4: Core sample and wave velocity sensors in the dilatometric device

3 Methodologies

3.1 Strain measurement and exposing conditions

The hygric swelling ability of the stone on the direction perpendicular to the natural bedding plane is measured using the dilatometric system (Fig. 4) adapted at the laboratory [Mertz et al., 2012]. LVDT sensors TWK (accuracy $0.3 \mu\text{m}$) recorded the free swelling strains under isothermal condition ($T=20^\circ\text{C}$) from 25% to 97% of relative humidity (RH). RH variations cycles are then performed, after stabilization of RH at 25%. Humidification phase is defined when moisture content reaches and maintains 97% of RH during 96 hours. Drying phase corresponds to the decrease and stabilization of RH at 25% during 48 hours. Samples are submitted to 7 cycles of such moisture variations.

3.2 P- and S- wave propagation

P-wave and S-wave velocities are measured at ultrasonic frequencies using a pulse transmission technique. Compressional (Sofranel CHRFO18) and shear (Sofranel SS018) piezoelectric sensors with a 1 MHz eigen frequency are used. They are applied at the surface of the samples (Fig. 5) and a vise ensures the contact. Honey is applied to improve the coupling between the sensors and the rock. For each velocity measurement, 50 waveforms are stacked in order to increase the signal/noise ratio. The reference first-arrival peak time and amplitude are measured for samples oven-dried until constant mass. The evolution with moisture content is then evaluated. The assembly samples and sensors are placed in the dilatometric system (Fig. 4) and are submitted to cycles of relative humidity changes. P- and S- wave measurements are performed during the moisture variations over the cycles and are coupled with strains measurement to evaluate moisture adsorption and desorption consequences on stone behaviour.

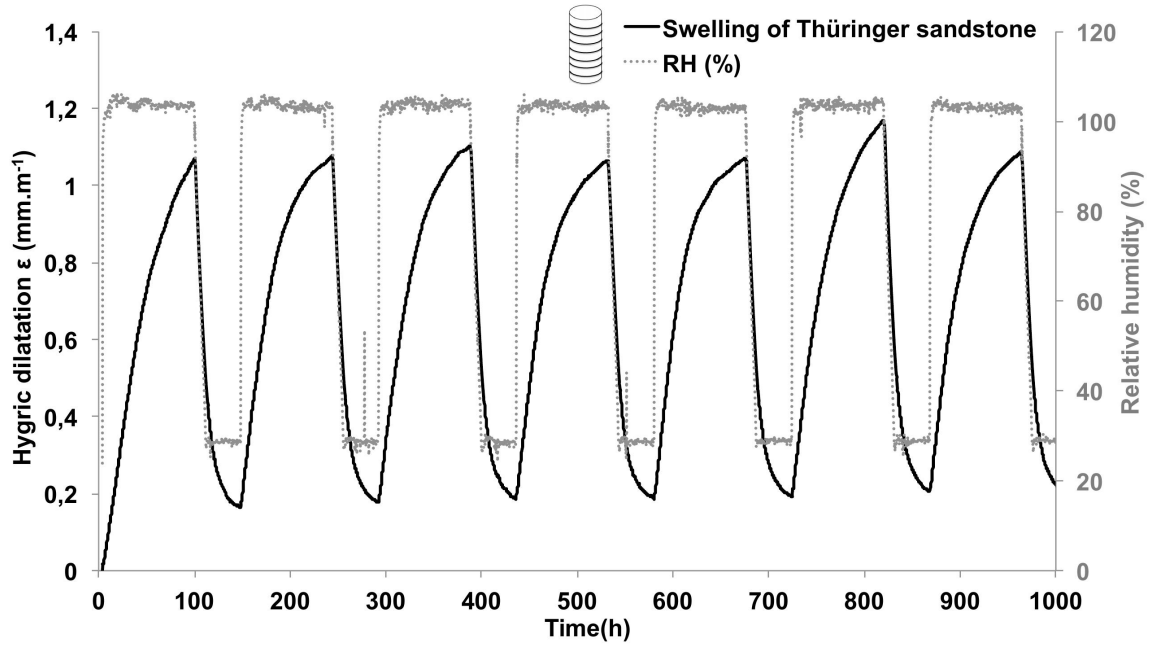


Figure 5: Dimensional behaviour of Thüringer sandstone in the direction perpendicular to the bedding plane during seven cycles of RH variations from 25% to 97%

4 Results and Discussion

4.1 Free swelling behaviour

The free swelling ability of the Thüringer sandstone in the direction perpendicular to the natural bedding plane is presented in Fig. 5. The swelling behaviour is reversible along the 7 cycles and the mean free swelling strains measured for RH variations from 25% to 97% are $\varepsilon = 0.68 \text{ mm.m}^{-1}$ for this clay-bearing sandstone.

4.2 Adsorption and desorption of moisture content over 1 cycle

The P- and S- elastic waves velocities under relative humidity variations are measured. Waveforms are studied at several relative humidity levels during humidification and drying along one cycle. The evolution of P-wave velocity during a complete cycle of RH variations from 25% to 97% is presented in Fig. 6.

P-wave velocity V_P decreases from 2900 m.s^{-1} to 2500 m.s^{-1} during the humidification phase and adsorption. The evolution of V_P during the increase of the relative humidity is not instantaneous and a time delay is observed: V_P still decreases during the stage while RH is maintained at 97%. During the drying phase and desorption, V_P increases again and reaches almost the value measured at the beginning of the cycle.

S-wave velocity V_S evolution coupled with the free swelling strains measured over one cycle within the sandstone is presented in Fig. 7 and a similar trend as the one recorded for P-wave velocity is observed. The length variation of the sample is relatively small (Section 4.1) and cannot be responsible for the observed modification of the P-wave and S-wave velocities.

V_P and V_S evolutions both highlight the elastic weakening of the Thüringer sandstone dur-

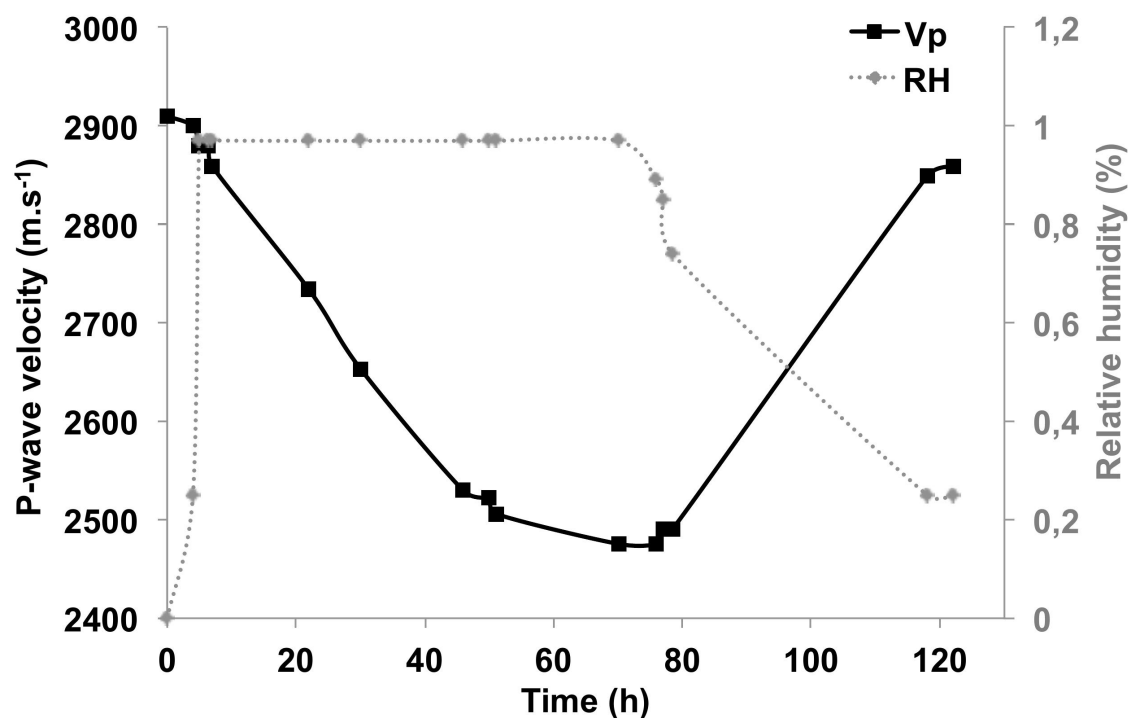


Figure 6: P-wave velocity and relative humidity for the Thüringer sandstone over 1 cycle of adsorption. These measurements illustrate the impact and the magnitude of hydration processes on the elastic behaviour on this clay-bearing sandstone.

4.3 Continuous evaluation of wave propagation under cycling of moisture content variations

S-wave velocity within the sandstone cores is investigated continuously during 7 cycles of RH variations. The evolution of V_S along 7 cycles - or 50 days - of humidification and drying is presented over time with respect to moisture content and dashed line for the fourth cycle is an extrapolation in Fig. 8. Two main results are observed: i) adsorption and desorption consequences are illustrated for each individual cycle, ii) the wave velocity V_S in the sandstone at RH=25% increases along the cycles. The temporal change is measured at $0.1 \mu s$ between the third and the seventh cycle of RH variations, corresponding to a $5 m.s^{-1}$ increase in V_S . Such wave velocity increase indicates that the dynamic modulus of elasticity perpendicular to the bedding plane E_{perp} becomes slightly higher after the seven cycles of RH variations. In other word, the repeated hygric solicitations seem to have an irreversible impact on the microstructure or the microstructural organisation of this stone. This irreversible evolution is confirmed by the porous distribution modification, presented in Figure 3. A shift at $2 \mu m$ of the second mode pore access radius illustrates the effect of the repeated RH cycles on the stone microstructure.

4.4 Elastic weakening and solvation pressure

To better understand the observed phenomena, the hypothesis that solvation pressure induced by adsorption of water in the microstructure is responsible for the elastic weakening

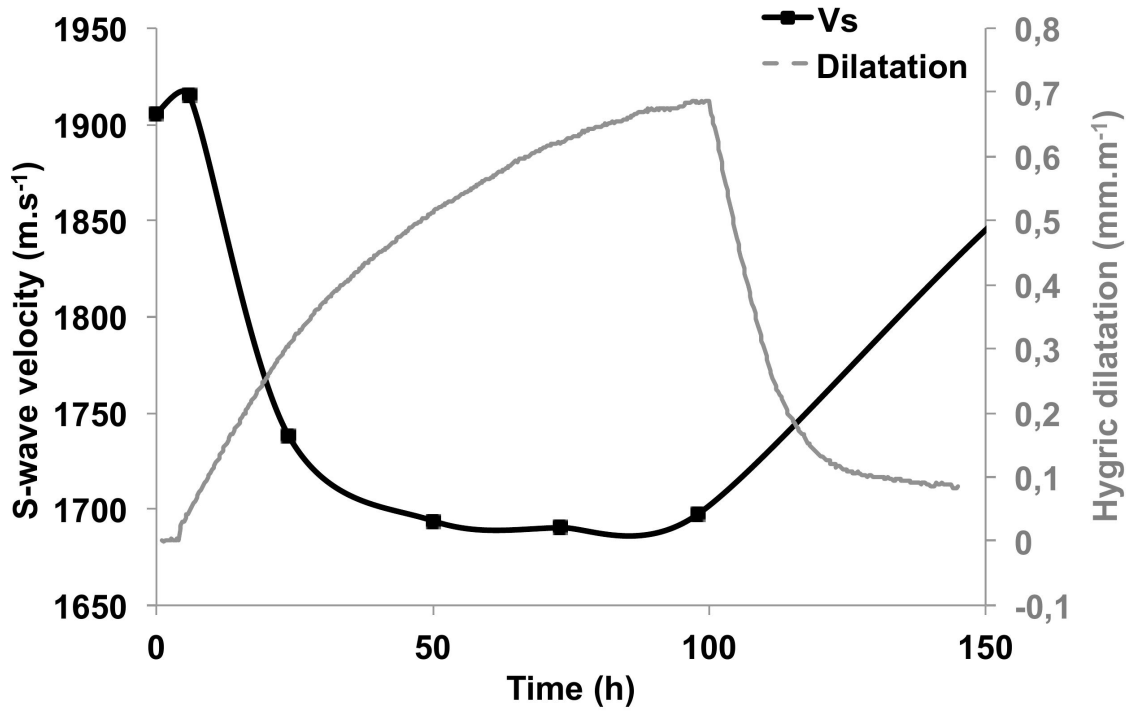


Figure 7: S-wave velocity and swelling strains for the Thüringer sandstone over 1 cycle of RH variations

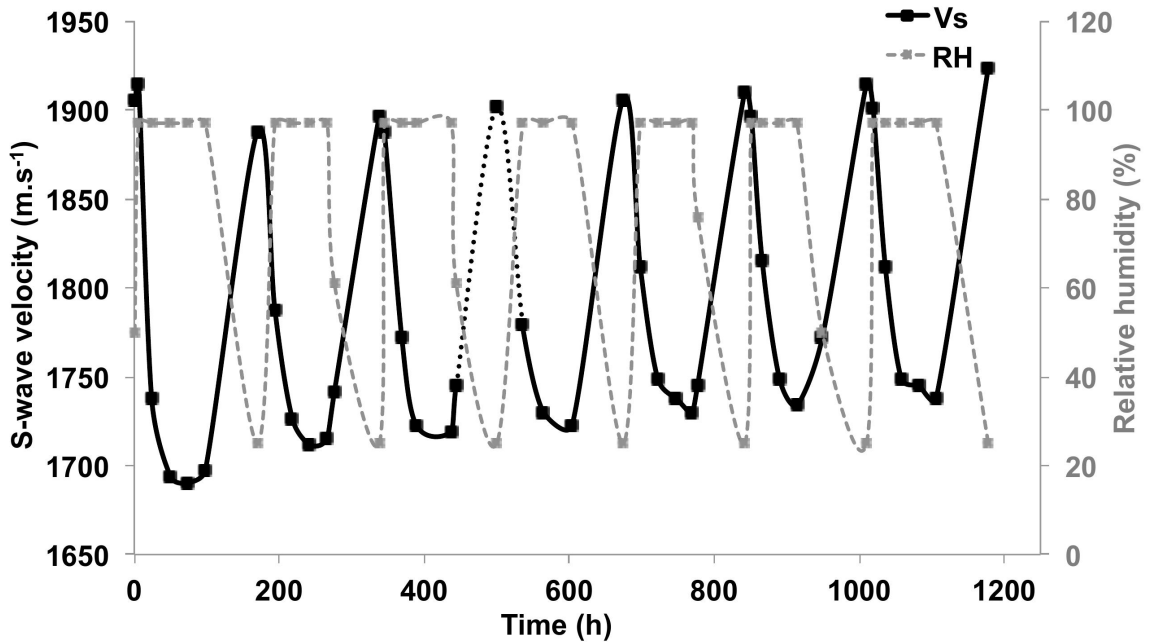


Figure 8: S-wave velocity V_s within Thüringer sandstone during the seven cycles of moisture variations from 25% to 97% of relative humidity (dot section is extrapolated for the fourth cycle) in black line, and RH variations in grey line

identified, is considered. In their paper, [Yurikov et al., 2018] calculate the fluid pressure using the following equation, adapted for our notation:

$$\Delta P_f = M_{pl} \varepsilon_S \quad (1)$$

with M_{pl} the pore load modulus (after [Prass et al., 2009]) and ε_S the free swelling strain.

After [Zimmerman et al., 1986], the M_{pl} modulus can be defined as:

$$M_{pl} = \frac{3}{C_{bp}} \quad (2)$$

with C_{bp} the bulk pore compressibility, related to the drained bulk moduli K and the bulk modulus of the solid K_S , estimated from the Reuss average ([Reuss, 1929, Yin et al., 2019]) as:

$$C_{bp} = \frac{1}{K} - \frac{1}{K_S} \quad (3)$$

Based on the measured values for the Thüringer sandstone, presented in [Yin et al., 2019], $K = 7$ GPa and $K_S = 33,8$ GPa, with respect to the amount of the various phases identified in the stone.

For moisture variations from 25% to 97% of RH, the free swelling strain measured is $\varepsilon = 0.68 \text{ mm.m}^{-1}$. After Equation (1), the fluid pressure induced by adsorption in the Thüringer sandstone is $\Delta P_f = 18$ MPa. Such pressure is assumed to create a substantial evolution of the elastic moduli.

Then, the comparison of the two ultrasonic measurements performed i) during RH increase and adsorption and ii) in a triaxial cell, supports the hypothesis. Indeed, when the confining pressure is increased from 5 to 25 MPa [Yin et al., 2019], V_S increases of 250 m.s^{-1} , a value similar to the one obtain during adsorption: V_S changes of an average 200 m.s^{-1} (Figure 7) for a variation of the solvation pressure estimated at 18 MPa.

5 Discussion

Adsorption and desorption processes and their consequences on a clay-bearing sandstone behaviour are investigated using elastic waves propagation during moisture variations cycles.

Focusing on the adsorption phase, a significant decrease in ultrasonic velocity, correlated with the elastic weakening induced in the stone, is identified. This phenomenon has already been observed and hypotheses on the surface energy evolution, the modification of the grain boundaries [Pimienta et al., 2014, Nakao et al., 2016], and the solvation pressure generated by the adsorption of water [Yurikov et al., 2018], have been discussed. With our carried out protocol, wave velocities measurements underline the water-mineral phases interactions during adsorption and illustrate the sorption kinetic within the studied sandstone. Indeed, the P-wave velocity loss presents a delay during humidification, which can be a consequence of the progressive moisture content equilibrium from the surface to the bulk material. This process is in close correlation with the porous distribution and the mineralogical composition. Moisture-sensitive phases, with respect to their nature and

accessibility, influence the kinetic of the interactions as well as the elastic weakening of the stone.

Furthermore, the suggestion [Yurikov et al., 2018] that the adsorption-induced elastic weakening is the consequence of the variation of fluid pressure in specific nanopores seems confirmed by the experimental protocol on this clay-bearing sandstone. The higher magnitude of solvation pressure observed here is the consequence of i) the stone porosity and the specific bimodal distribution, ii) the nature, amount and localisation of clay phases identified in this sandstone.

The long-term behaviour of the stone, thanks to these repeated solicitations, is also studied. Continued elastic wave measurements during adsorption from 25 % and 97% of RH illustrates a 5 m.s^{-1} increase of the S-wave velocity V_S . It means that the natural swelling behaviour of this sandstone induces an irreversible increase of the elastic properties over cycles. SEM observations and fracture toughness measurements, combined with the evolution of the porous distribution after the 7 cycles of hygric solicitations are in correlation with the increase of the elastic properties. The complete set of data and results are described in details as part of another research publication [Tiennot et al., 2017]. A reorganisation upon repeated adsorption and a new preferred orientation of the illitic filament-shaped clays were identified. After 7 cycles, a local redistribution appeared, leading to the filling of some previously accessible spaces. This effect can be correlated to the slight increase of the wave propagation velocity measured.

Thus, the impact of clay minerals on stone elastic weakening during adsorption, as well as on the elastic properties irreversible evolution, has been illustrated over several cycles of moisture variations.

6 Conclusion

The long-term behaviour of a clay-bearing sandstone has been investigated. Identification and consequences of adsorption and desorption over repeated moisture cycles are highlighted using ultrasonic P- and S- waves velocities. The continuous analysis along multiple cycles developed in this research improves the quantification of stone elastic properties' evolution with moisture variations. Thanks to these experimental results, the solvation pressure as well as the related elastic weakening induced by adsorption are also evaluated. Moreover, these measurements illustrate that the elastic weakening is in close correlation with the clay minerals' nature, distribution and moisture sensitivity.

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