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EFFECT OF MOISTURE ON TUFF STONE DEGRADATION

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Abstract
Tuff stone elements with a large length/width ratio often suffer damage in the form of cracks parallel to the surface and spalling of the outer layer. The response of tuff to moisture might be a reason for this behaviour. This research aimed at verifying if differential dilation between parts with different moisture content (as outer and inner part of partially encased mullion) can lead to damage.
The effect of moisture on the degradation of Ettringen and Weibern tuff from the Eifel, Germany, has been investigated. A purpose-made weathering test was carried out to simulate the wetting-drying process. Despite no cracks developed during the test, existing cracks widened up and the flexural tensile strength of both materials decreased. The moisture transport properties their porosity and pore size of the stones were determined. Ettringen tuff has a considerable amount of very fine porosity, resulting in slow moisture transport and significant hygroscopic adsorption. Both tuff stones have an extreme hydric dilation. Environmental X-ray diffraction analyses showed that Ettringen tuff undergoes (reversible) mineralogic changes when subjected to RH cycles, whereas this does not occur for Weibern. The results support the hypothesis that moisture gradients in tuff elements may enhance decay in this stone.

1. Introduction

Volcanic tuff stone from the Eifel region (Germany), including Römer (Roman), Weibern and Ettringen tuff stone, is one of the most important stone types used in Dutch architecture since the Roman times [1-5]. Tuff stone is often regarded, but not correctly, as a stone with a limited durability; this may also derive from the fact that the different damage mechanisms affecting tuff stone have not been fully elucidated yet.
In tuff stone elements with a relatively high length to width ratio, as e.g. mullions and window sills, damage often occurs in the protruding, exposed part of the element, in the form of longitudinal cracks leading sometimes to spalling of the outer layer (figure 1). As Ettringen is mostly used in these building elements, damage is mainly observed in this type of tuff stone. However, it cannot be excluded that Weibern tuff could develop a similar decay pattern as well.
The effect of frost and salt decay on tuff stone has been widely researched in the past

[1, 6-10], but the cause of the above described decay patterns is still unclear. One of the hypotheses is that cracks are due to the hygric-mechanic behaviour of the tuff stone. Tuff stone has a high porosity and a bimodal pore size distribution, with both very coarse and very fine pores; these properties lead to a high and fast water absorption and to a slow drying. When the protruding part gets wet due to rain and dry afterwards, differences in moisture content and consequent hygric dilation may develop between the exposed and the encased part of the stone, possibly leading to stresses at the interface. These stresses may cause damage in the form of longitudinal cracks at the interface and spalling of the outer layer of the stone. This hypothesis has been investigated in this research on both Ettringen and Weibern tuff stone. A purpose-made weathering test was carried out to simulate the wetting-drying process due to rain an sun. Additionally, stone properties relevant to this damage mechanism have been investigated as well.

Figure 1. Ettringen tuff mullions showing damage in the form of longitudinal cracks (tower of Eusebius church, Arnhem, the Netherlands)

2. Materials and methods

Ettringen and Weibern tuff stones used in this research originate from the Riedener caldera in the Eifel, Germany [8, 11-13].

The test plan consisted of a weathering test, aiming at simulating in laboratory the wet-dry cycles occurring in the field, and a series of characterization tests, some of them carried out before and after the weathering test. Specimens of different sizes were used for the tests: cubes, prisms, and small scale mullions and window sills (figure 2). The characterization tests, including specimen size and number, are summarized in table 1.

The mineralogical and petrographical properties of the tuff stones were investigated by Polarized and Fluorescent microscopy (PFM). Specimens were prepared by impregnating the stone under vacuum with a UV-fluorescent resin and then cutting and polishing the samples to obtain thin sections of 25-30 μm thickness. PFM observations were also carried out, together with so-called fluorescence macroscopic observations (FMA), to assess the appearance of damage (cracks, mineralogical changes, etc.) after the weathering test.

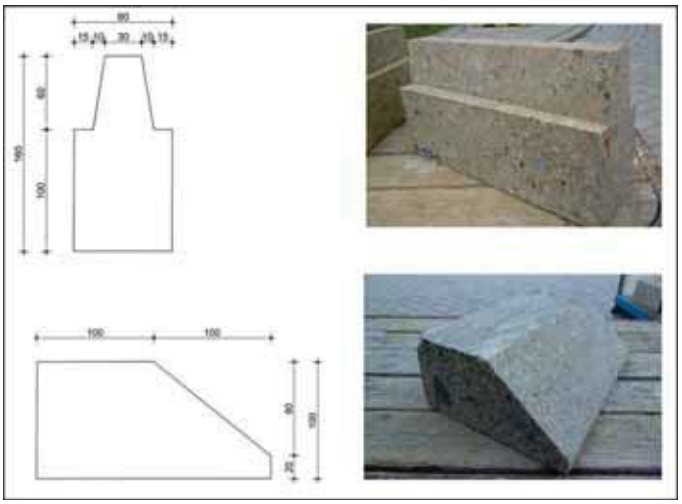

Figure 2. Shape and size (in mm) of small scale mullions and window sills.

Table 1. Characterization tests (B= before weathering test; A= after weathering test).

Test method	**Type of specimen**	**Size**	**B/A**	**Replica's per stone type**
Water absorption	Cubes	10 x 10 x 10 cm^3	B	3
Drying at 20 °C /50% RH	Cubes	10 x 10 x 10 cm^3	B	3
Porosity	Prisms	16 x 4 x 4 cm^3	B	3
Mercury Intrusion Porosimeter	Little pieces	≅ 1 cm^3	B	2
N_2 adsorption	Little pieces	≅ 1 cm^3	B	2
Flexural & compressive strength	Cubes	4 x 4 x 4 cm^3	B, A	5B + 5A
Macroscopic observations on samples impregnated with fluorescent resin	Part of cubes, mullions and window sills		B, A	B:2 cubes + 1 cube with finish A: all
Polarized and Fluorescent microscopy (PFM)	Cubes	10 x 10 x 10 cm^3	B	1
	Mullions (with finish)	See figure 2	B, A	1B, 1A
	Mullions	See figure 2	B, A	1B, 1A
Hygric dilation	Prisms	16 x 4 x 4 cm^3	B	3
Hygric dilation	Prisms	16 x 4 x 4 cm^3	B	1
Thermal dilation (20-40°C)	Prisms	16 x 4 x 4 cm^3	B	3
X-ray diffraction	Powder	n.a.	B	2
X-ray diffraction (RH cycles)	Powder	n.a.	B	1

Phases were identified by Bruker Eva 2.0 software and the crystallographic databases ICDD PDF2 (2011) and ICSD (2011). The XRD analyses for the identification of the mineralogical composition were carried out at 20 °C /40 % RH. Additionally, in order to check whether any chemical transformation occurs due to RH changes, XRD diffraction analyses were carried out at different RH's: at first the RH was increased, with steps of 10% RH, from 40 to 90% and then lowered again, with similar steps, to 10 % RH.

The physical properties of the tuff stones were studied by a combination of methods and techniques. The water absorption of the stone at 20 °C 50% RH was measured, according to EN 13755 [14], on cubes 10 x 10 x 10 cm^3 sealed with epoxy resin on the lateral sides. The wetting front in the stones was photographically monitored. After absorption, the specimens were fully saturated by immersion in water and then dried at 20 °C 50% RH through one surface. Their weight was monitored at regular time intervals during drying.

The porosity of the stones was measured according to the RILEM CPC 11.3 on 4 x 4 x 4 cm^3 cubes [15]. Additionally, porosity and pore size distribution were measured by Mercury Intrusion Porosimeter (MIP) using a Micrometrics Autopore IV9500A. By the use of this instrument pore entrances of diameter size between 0,007 and 366 μm can be measured. Smaller pores were measured by nitrogen adsorption (Micrometrics Tristar 3000 Adsorption Analyzer); adsorption and desorption curves were measured at 77 K (-196 °C).

The thermal and hygric dilation were determined on 4 x 4 x 16 cm^3 specimens. The thermal dilation between 10°C, 20°C, 30°C and 40°C was measured by means of a dilatometer (precision 0,001 mm), after conditioning the specimens at each temperature in a climatic cabinet. Similarly, the hygric dilation was measured after conditioning the specimens at different RH conditions (30%, 50%, 65% and 93%) and in water, at a stable temperature of 20 °C. Additionally, hygric dilation was continuously monitored by means of linear variable differential transformers (LVDT) when cycling the RH between 30% (72 hours) and 93 % RH (24 hours) at a constant temperature of 20 °C during 12 days.

The flexural and compressive strength of the stone was assessed on 4 x 4x 16 cm^3 specimens, according to NEN EN 196-1 [16], before and after the weathering test. The load was applied with a speed of 300 N s^{-1} and a pre-loading 10 N.

The weathering test aimed at reproducing the wet-dry cycles to which tuff stone mullions and window sills are subjected when positioned in building masonry. A test set-up was developed to this purpose consisting of:

- A frame, on which the specimens were placed, positioned on an angle in order to allow flowing away of the water.
- Two pipes with hoses to sprinkle the specimens with water (reproducing rain)
- Four infrared lamp to lighten and warm up the specimens (reproducing the effect of the sun)
- Thermocouples to measure the surface temperature of the specimens. The thermocouples were connected to a computer, so that the intensity of the infrared lamps could be automatically adjusted to keep the temperature constant at 40 °C during the "sunny" period.

The following wet-dry cycle was used: 8 h rain, 64 h drying, alternating a 4 h period of drying at 40 °C and 4 h drying at room temperature. The length of the cycles was chosen based on the water absorption and drying properties of the tuff stone and with the aim of providing an accelerated, but still realistic, reproduction of the situation in the field.

The specimens (type, number and size specified in table 1and figure 2) were sealed with resin on those sides which are normally encased in masonry.
During and at the end of the weathering test, the appearance of new cracks or the widening of existing cracks was visually and photographically monitored. During the last wet-dry cycle, the moisture distribution in the exposed and encased part of mullions and window sills was assessed, before and after the rainy period, by drilling powder samples at different depths and determining their moisture content gravimetrically.

3. Results

3.1 Characterization tests

3.1.1 PFM

The PFM observations show that Ettringen tuff has more stone fragments (basalt, sandstone and schists) and less pumice than Weibern tuff. The pumice has been zeolitized and contain inclusions of xenocrysts and/or phenocrysts (Ti-augite, leucite, quartz, opaque minerals, phlogopite and sanidine). Some voids are filled with calcite. Weibern tuff has a higher porosity than Ettringen, also because of the presence of a larger amount of pumice. Next to pumice, stone fragments (sandstone, schist and siltstone) are present; these are smaller in size and lower in number than observed in Ettringen tuff stone. Xeno- and/or phenocrysts are in this case constituted by Ti-augite, quartz, biotite / phlogopite and tourmaline.

3.1.2 XRD analyses

The XRD pattern of Ettringen tuff stone shows the presence of quartz, albite, sanidine, leucite,clinopyroxene and muscovite, next to philipsite-Ca as the only zeolite. In the Weibern tuff stone, quartz, sanidine, augite, phlogopite and illite are present, next to analcime as the only zeolite. The zeolite assemblages in the current samples differ from those encountered in the past for both tuff stones (e.g. the lack of chabazite; [1]).
The XRD patterns of Weibern tuff do not change when collected at different RHs. In contrast, the XRD pattern of Ettringen varies with RH, indicating that crystallographical changes due to RH cycles; these differences are reversible and seem to be caused by changes in the crystal structure of philipsite-Ca.

3.1.3 Porosity and pore size distribution

The total porosity measured by saturation under vacuum according to RILEM CPC 11.3 is 35. vol% (standard deviation 0.49) and 43. vol% (standard deviation 0.72) for Ettringen and Weibern, respectively. The porosity and pore size distribution of Ettringen and Weibern tuff stones, as measured by MIP, are reported in figure 3. The graph shows that Weibern has a higher open porosity than Ettringen, but their pore size distribution in the range measured by MIP is similar. The open porosity values measured by MIP are (slightly) lower than those obtained by immersion, fact which might be due to the presence of pores larger than 366 μm (largest size measured by MIP) and/or to the lower representativeness of the small samples used for MIP measurements.Pores smaller than 0,1 μm were measured by N_2 adsorption (figure 4). These results show that Ettringen has a larger amount of very small pores (2-4 nm) than Weibern tuff. This can significantly affect the hygric behaviour of the stone.

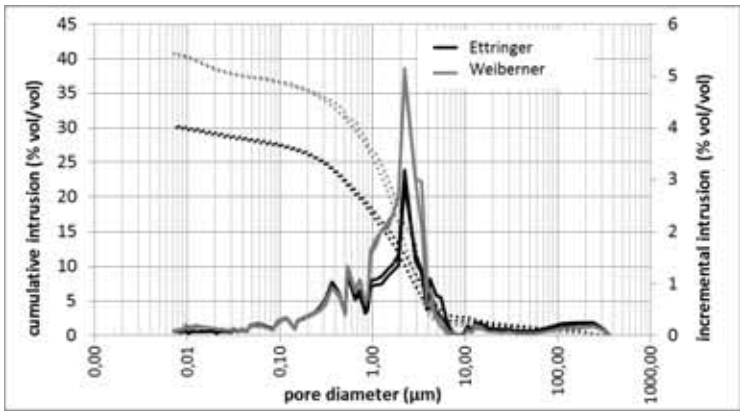

Figure 3. Open porosity and pore size distribution of Ettringen and Weibern as measured (in twofold) by MIP (continuous line: incremental intrusion; dashed line : cumulative intrusion)

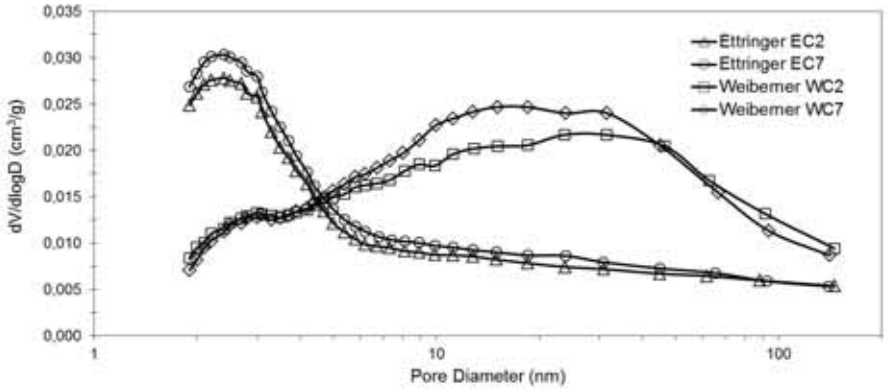

Figure 4. Pore size distribution of Ettringen (E) and Weibern (W) measured (in twofold) by N_2 adsorption

3.1.4 Water absorption and drying

Figure 5 shows the water absorption curves of the tuff stones: Weibern has a higher total absorption than Ettringen, fact which corresponds to its higher porosity. The water absorption coefficient (WAC) of Weibern (0,316 kg m^{-2} $s^{-0.5}$) is higher than that of Ettringen (0,064 kg m^{-2} $s^{-0.5}$), indicating the faster absorption of the first with respect to the second. The measured WAC for Ettringen is slightly higher than values earlier measured in this stone type (0,05 kg m^{-2} $s^{-0.5}$, reported in [6]); the WAC measured for Weibern lies in the range reported in literature (0,24 – 0,38 kg m^{-2} $sec^{-0.5}$, as derived based on [6,17-18]). During absorption it was observed that the wetting front proceeds much faster in Weibern than in Ettringen: this difference is probably explained by the pore structure of Ettringen and/or by the presence of very small pores (2-4 nm) in Ettringen tuff stone, which delay the penetration of the wetting front. The drying of both tuff stone is quite slow: after more than 3 months the specimens are not fully dry yet. Similarly to the absorption, the drying of the Ettringen stone is slower than that of Weibern.

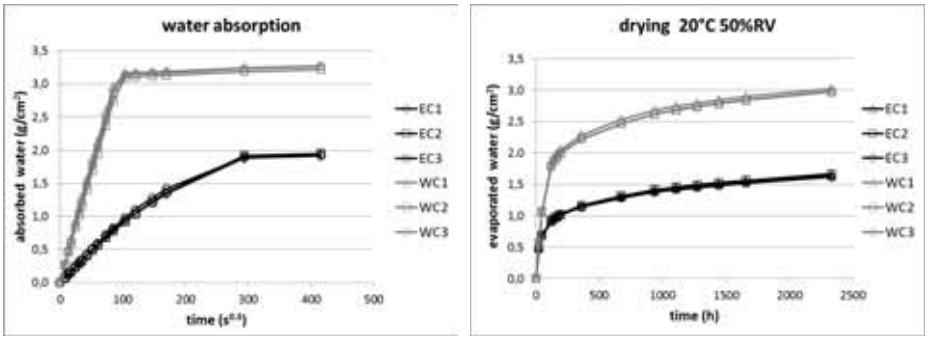

Figure 5. Water absorption (left) and drying (right) of Ettringen (EC) and Weibern (WC) tuff.

3.1.5 Hygric and thermal dilation

The hygric dilation of Ettringen and Weibern, calculated with respect to the specimen size at 20 °C 30% RH, is given in figure 6. The hygric dilation of both stones is high, with a maximum of about 1.2 μm/mm, reached by immersion of the specimens in water. Ettringen tuff stone shows a significant hygric dilation already at low RH. The hygric dilation corresponds to the hygroscopic adsorption of the specimens: Ettringen specimens, due to the presence of very small pores (see 3.2), start to adsorb moisture already at low RH values (figure 6).

The reversibility of the dilation was checked by continuously monitoring the dilation during RH cycles. This test shows that the dilation is fully recovered (within the test period) (figure 7).

The thermal dilation between 10 and 40 °C at 65 % RH is similar for both tuff stones and equal to 0.15 μm/mm; based on these results it can be concluded that the thermal dilation is much less relevant for damage development than the hygric dilation.

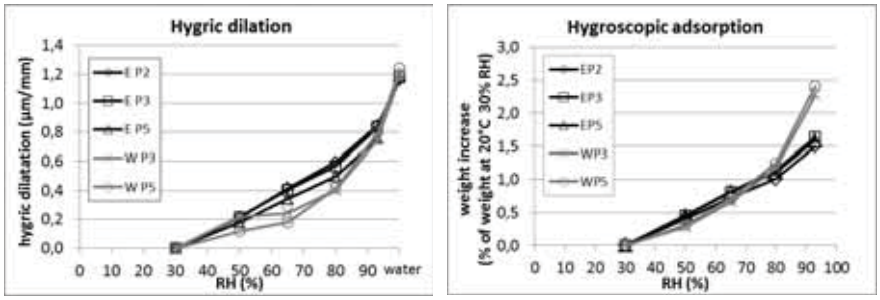

Figure 6. Hygric dilation (left) and hygroscopic moisture adsorption (right) of Ettringen (E) and Weibern (W) tuff stones.

3.1.5 Mechanical strength

The flexural and compressive strength of the stones before the weathering test are given in table 2. The flexural and compressive strength of Ettringen tuff stone is about double than that of Weibern. The strength values measured for Ettringen tuff show a large standard deviation, indicating that the properties of this tuff stone can significantly vary even within blocks from the same quarry (as found in previous studies).

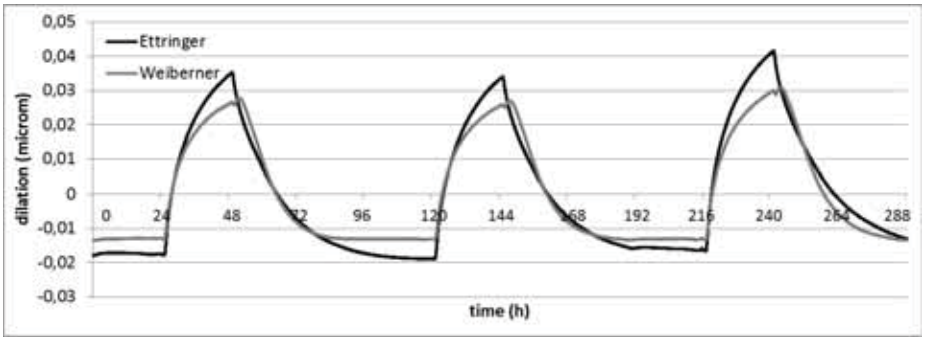

Figure 7. Dilation of Ettringen and Weibern tuff stone during RH cycles between 30 and 93% RH, at the constant temperature of 20 °C

Table 2. Flexural and compressive strength of Ettringen and Weibern tuff measured before the weathering test (average of 5 specimens ± standard deviation)

	Flexural strength (N/mm^2)		Compressive strength (N/mm^2)	
	Ettringen	Weibern	Ettringen	Weibern
Before weathering	8.10 ± 1.80	4.24 ± 0.40	29.41 ± 3.18	13.24 ± 1.44

3.2 Weathering test

The weathering test ran during about 3 months. During and at the end of this period the specimens were visually examined to check the appearance of cracks. According to the supposed damage mechanism, cracks would develop longitudinally, parallel to the exposed surface, in the exposed part of the stone elements. No cracks with these features could be observed with the naked eye. However, the randomly oriented cracks already present before the test seem, based on visual observation, to have widened up. FMA (on all specimens) and PFM (on a selection of 8 samples) observations carried out at the end of the test confirmed the absence of cracks which could be due to the supposed damage mechanism.

During the last wet-dry cycle, the moisture content in mullions and window sills before and after the wet period was gravimetrically determined. The results (figure 8) show that the difference in moisture content (MC) between the encased and exposed parts can be high for both mullions and window sills. This implies that the difference in hygric dilation between exposed and encased parts can be relevant (see 3.1.4). The mechanical strength of the specimens subjected to the weathering test was assessed and compared to that measured before the test (table 3). A decrease of the flexural strength is observed for both tuff stone types after the weathering test. Differently, the difference in compressive strength before and after the weathering test lies within the range of the standard deviation and is thus not significant.

Table 3. Flexural and compressive strength of Ettringen and Weibern tuff measured after the weathering test (average of 5 specimens ± standard deviation)

	Flexural strength (N/mm^2)		Compressive strength (N/mm^2)	
	Ettringen	Weibern	Ettringen	Weibern
After weathering	6.11 ± 1.11	3.07 ± 0.56	27.65 ± 1.78	14.38 ± 1.54

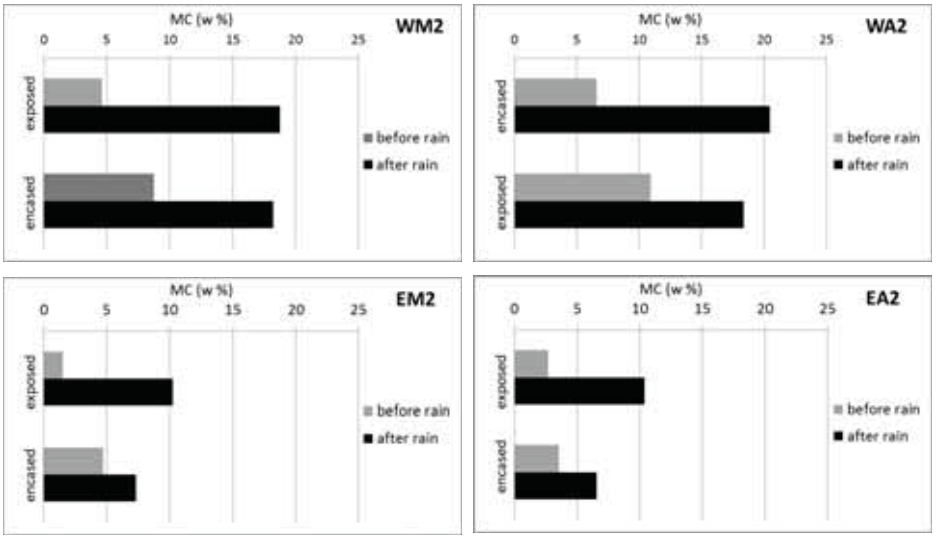

Figure 8. Moisture content (MC) in Ettringen (E) and Weibern (W) mullions (M) and window sills (A)

4. Discussion and conclusions

This research aimed at verifying if a differential dilation between parts of tuff stone elements with different moisture content can lead to damage in the form of longitudinal cracks between the protruding and encased part of tuff stone elements. To this scope, the effect of moisture on the degradation of Ettringen and Weibern tuff has been thoroughly investigated by means of different methods and techniques. Moreover, a purpose-made weathering test has been carried out to simulate the wetting-drying process. Despite no cracks developed during the test, which could definitely confirm the supposed damage mechanism, the results obtained from the characterization tests support the hypothesis that the hygric behaviour of the stone plays an important role in the decay mechanism of Ettringen and Weibern tuff.

First of all, because of the presence of both coarse and very fine pores, both these stones show fast water absorption but very slow drying: this behaviour makes them particularly prone to moisture related damage mechanisms, as biological growth, frost and salt crystallization. Besides, both stones were shown to have a high hygric dilation, which would lead to high stresses at the interface between parts of the stone with different moisture contents, as those which develop during wet-dry cycles. Despite hygric dilation was shown to be reversible (at least in the short term), a decrease in the flexural strength of the stones was measured after the weathering test, suggesting that repeated cycling can lead to weakening of the stone.

Ettringen seems to be more sensitive for damage than Weibern due to the presence of very fine pores, which lead to hygroscopic adsorption and hygric dilation even at low RH. The sensitivity of Ettringen to RH is shown also by the mineralogical changes undergone by the stone (most probably by the philipsite-Ca in de zeolites assemblage) during RH cycles. All factors suggest that Ettringen might be more susceptible to moisture related damage than Weibern. This high susceptibility might be (partially) compensated by its higher mechanical

strength. The behaviour of Ettringen and Weibern tuff with different zeolite assemblages (cf.[1]) and relationship between these assemblages and hygric behaviour and RH sensitivity require further investigations.

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