



**HAL**  
open science

# Assessment Of The Hygrothermal, Microstructural And Chemical Evolution Of A Hemp-Based Cementitious Mortar Under Etics Total Weathering Aging Protocol

D Kosiachevskiy, Kamilia Abahri, Anne Daubresse, Evelyne Prat, Mohend Chaouche

► **To cite this version:**

D Kosiachevskiy, Kamilia Abahri, Anne Daubresse, Evelyne Prat, Mohend Chaouche. Assessment Of The Hygrothermal, Microstructural And Chemical Evolution Of A Hemp-Based Cementitious Mortar Under Etics Total Weathering Aging Protocol. Construction and Building Materials, 2022, 10.1016/j.conbuildmat.2021.125471 . hal-03874140

**HAL Id: hal-03874140**

**<https://hal.science/hal-03874140>**

Submitted on 27 Nov 2022

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

# ASSESSMENT OF THE HYGROTHERMAL, MICROSTRUCTURAL AND CHEMICAL EVOLUTION OF A HEMP-BASED CEMENTITIOUS MORTAR UNDER ETICS TOTAL WEATHERING AGING PROTOCOL

D. Kosiachevskiy<sup>1,2,\*</sup>, K. Abahri<sup>1</sup>, A. Daubresse<sup>2</sup>, E. Prat<sup>3</sup>, M. Chaouche<sup>1</sup>

<sup>1</sup> Université Paris-Saclay, ENS Paris-Saclay, CNRS, LMT - Laboratoire de Mécanique et Technologie, 91190, Gif-sur-Yvette, France

<sup>2</sup> Centre d'Innovation Parexgroup, 38070 St Quentin Fallavier, France

<sup>3</sup> Sika Technology AG, Tüffenwies 16, 8048 Zürich, Switzerland

## Abstract

Biobased composites are widely used for building insulation because of their hygrothermal performance, and their economic and environmental benefits. Among the different types of organic fibers for these materials, hemp shives are interesting because of their high availability in France and their low price. Nevertheless, the wide application of the hemp-based insulation mortars is hampered by the lack of data on its durability and behavior over the long term. The objective of this study, in a first part, consists in providing an original accelerated aging protocol inspired by the standardized one used for External Thermal Insulation Composite Systems (ETICS). In a second part, the selected aging weathering cycles were applied to investigate the hygrothermal stresses on the morphology, chemical composition and properties of hemp mortar samples. Experimentally, microstructural characteristics (as total porosity, SEM observation) and hygrothermal properties (as thermal conductivity, Moisture Buffer Value (MBV) and water vapor permeability) were measured and evaluated for both the reference state (after 28 days of conditioning) and after aging (after 28 days of conditioning and the aging cycles).

Furthermore, X-ray diffraction and PH analysis were performed to investigate the chemical composition, especially the effect of carbonation. The obtained results reveal the presence of two processes in the aged hemp mortars: the carbonatation of the mineral matrix and the cracking of the interface between the hemp shives and the cement matrix. This later was quantified too. Despite the microstructural degradation, small deteriorations were recognized concerning the hygrothermal properties and behavior of hemp mortar, that is essential for an insulation material. These results provide data for better forecast on the degradation of the hemp mortars.

**Keywords:** Hemp mortar, ETICS, rain cycles, heat-cold cycles, microstructure, thermal conductivity, moisture buffer value, vapor permeability

---

\* Corresponding author; e-mail: [kosiachevskiy.dima@gmail.com](mailto:kosiachevskiy.dima@gmail.com) (D. Kosiachevskiy)

## 1. Introduction

The building industry is a very energy consuming sector and is responsible for a large share of the world's CO<sub>2</sub> emission. Besides large-scale activity, like transportation, it is due to the use of the energy intensive materials like cement, bricks, steel and others. Moreover, the construction of the buildings represents a small part of the energy consumption in their total life cycle. As for the use of residential and tertiary buildings, it consumed 45% of total final energy and emitted 22% of CO<sub>2</sub> in France in 2016 [1]. The only viable solution for developing the sustainable building production system is to minimize the net CO<sub>2</sub> emission and use alternative low-energy building materials and more energy efficient technologies. That is why the use of bio-based materials based on different types of agricultural products gained a particular interest in France and around the world in order to solve the problem of CO<sub>2</sub> emission. In addition to their low carbon footprint, the use of these materials is promoted due to their hygrothermal properties and the availability of the raw materials [2]. Moreover, the necessity of more thermal insulation to get better energy performance of buildings leads to the problem of air quality, and hemp mortar is the material that can regulate indoor conditions by reducing daily humidity variations. Different natural fibers in building insulation materials are well studied, especially in regard to mechanical, thermal and hygroscopic properties. There are a lot of works on the properties of wood [3,4], straw [5], cork [6–8], jute [9], and hemp [2]. However, the use of bio-based building materials is hampered by the lack of a database relating to its durability.

In this paper, the investigated material is hemp mortar which is used for its thermal, acoustic and hydric performances [10]. Nevertheless, hemp mortar is a material of complex, heterogeneous and anisotropic composition. Like all composites based on natural fibers, hemp mortar must be investigated taking in account its multiscale nature related to its strong heterogeneity: from the macro- and microscale where hemp mortar is represented as a constitutive part of the envelope or as a multiphase material with interphases between hemp particles and a cementitious matrix to the nanoscale of organic cell walls tissues or cement hydrates. Another feature that improves the performances (for example thermal properties) of the insulating hemp-based mortar is that it has a double porosity: intra- (hemp particles porosity) and inter-particular (hemp/matrix interface). Consequently, such compositional and structural complexity complicates the investigation of hemp mortar when talking about the durability.

Another important point that must be taken into account is the hygroscopicity of hemp mortar and its sensitivity to thermal and hydric solicitations and to the environmental conditions of its life cycle. In fact, directly after the hemp mortar cast at early-age, cracks appear at the interfaces between the hemp particles and the cementitious matrix. Later, these cracks increase due to dimensional variations of hemp particles caused by the relative humidity solicitations. This modifies the hemp mortar characteristics and leads to the degradation of the material [11,12]. The evolution of the

microstructural properties must be investigated in order to understand the behavior and durability of the bio-based composite as regards to the hygrothermal stresses.

When talking about the durability of hemp mortars, few researchers studied the hygrothermal stresses impact on the hemp mortar properties. For example, Sentenac *et al.* [13] studied the performance and durability of treated hemp mortar. To estimate the evolution of the different properties and physical characteristics, the full immersion test was used. The conditioning represents 12 to 15 cycles of water immersion at 20 °C during 48 hours and drying at 40 °C during 48 °C depending on the samples. This immersion protocol has often been criticized because it is highly aggressive and also not very representative of the real ageing conditions of the materials.

Other researchers used accelerated wetting and drying cycles to investigate the aging of hemp mortars. For example, Marceau *et al.* [14] adopted the wet/dry cycles protocol at constant temperature of 30°C (each cycle lasts 8 hours). Each conditioning consists of 8 cycles: 4 cycles of 40% / 90% RH and 4 cycles of 40% / 98% RH. They studied the changes in the microstructure (density evolution, hemp particles / cementitious matrix interface) and physical properties (thermal and acoustical performance). Delannoy *et al.* [15] proposed another accelerated aging protocol consisting of 5 days at 98% RH and 2 days of 40% RH constantly at 30 °C. Their study examines the evolution of the different properties of hemp mortar and its chemical composition.

In all of the proposed literature works, non-standardized accelerated aging protocols were adopted that are not representative of the real-life cycle conditions of hemp mortar insulation. This is explained by the total water immersion in the case of the protocol of Sentenac *et al.* [13] or high relative humidity in the works of Marceau *et al.* [14] and Delannoy *et al.* [15], that do not appear under standard conditions of application and use recommended by Piot *et al.* [16]. That is why the main objectives of this paper are to adapt and propose the accelerated aging protocol basing on the standardized one used for External Thermal Insulation Composite Systems (ETICS) with rendering [17] and to investigate the microstructure and properties evolution of hemp mortar due to proposed aging cycles. The main advantages of the proposed protocol are the use of heat-rain and heat-cold cycles and protection against the direct water access that allow to represent real conditions of use of biobased insulation materials with rendering. Experimentally, the evaluation of the hemp mortar characteristics was conducted before and after weathering cycles.

## **2. Experimental procedure**

In this section, the studied material, the accelerated weathering aging cycles conditions and the experimental characterization methods are presented. The studied hemp mortar can be used for both external and internal insulation. Characterization of hemp mortar was conducted on two benchmarks: before and after the exposure to aging protocol. This allows us to compare the results and study the

influence of the conditioning on hemp mortar microstructure, chemical composition and hygrothermal properties.

### 2.1. Materials

The hemp shives of France origin were used for manufacturing the hemp mortar samples. Their visual aspect and granulometric curve are presented in Figures 1 and 2 respectively. The binder represents a commercial mix designed by ParexGroup SA. It is composed of hydrated lime, Portland cement and various additions to achieve the rheology suitable for spray application. Chemical composition of mineral binder is presented in Table 1. The hemp mortar proportion ratios are represented in the Table 2. This mix is suitable for material application by spraying. This requires special attention to the rheology that allows the industrial application of insulating hemp mortar. The studied samples were manufactured in laboratory using the laboratory mixer. The mixing was done in two stages: firstly, hemp shives were mixed with water during 1 minute at 140 rpm, secondly, the binder was added and it was mixed during 5 minutes at 140 rpm. After their laboratory manufacture, the specimens were dried for 28 days at laboratory conditions ( $23 \pm 2$  °C,  $50 \pm 5\%$  RH) as shown in Figure 3. The adopted sample sizes depend on experimental procedure and are presented for every type of test in the paragraph 2.3.



Figure 1. Hemp shives

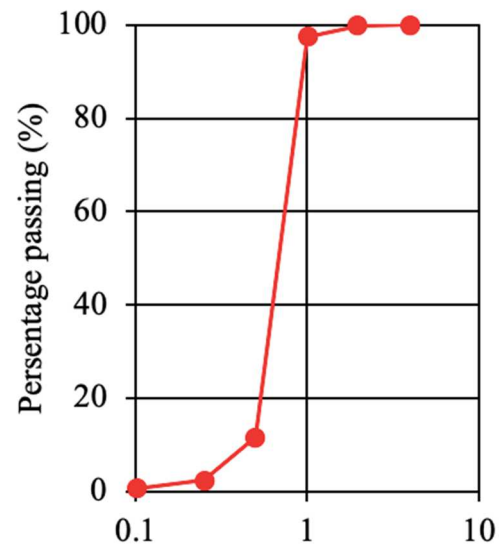


Figure 2. Granulometric curve of hemp mortar by image analysis

Table 1. Oxide analysis of used lime-based binder with hydraulic additions

Oxide	CaO	Al <sub>2</sub> O <sub>3</sub>	Na <sub>2</sub> O	K <sub>2</sub> O	Fe <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	MgO	SO <sub>3</sub>
Percent by mass (wt%)	57.40	2.20	0.15	0.27	0.92	25.40	0.50	3.09

Table 2. Mass ratios of hemp mortar composition

Component	Hemp shives	Mineral binder	Water
Percent by mass (wt%)	12.5 %	25 %	62.5 %

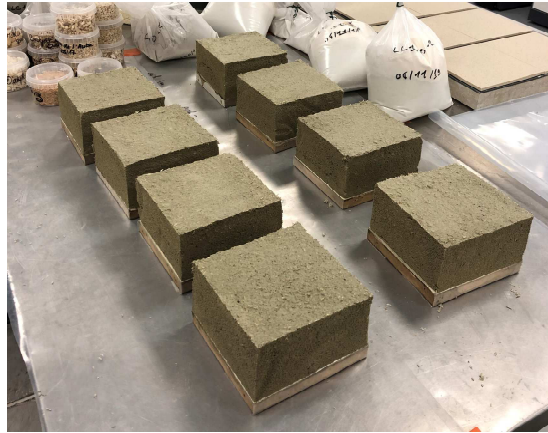


Figure 3. Hemp mortar samples laboratory drying

## 2.2. Accelerated weathering aging cycles

After the 28 days of drying, the hemp mortar samples were conditioned in two different ambiances. Total weathering protocol is represented in Figure 4-A as is explained in the following:

- The first batch of specimens was subjected to the laboratory conditions ( $23 \pm 2$  °C,  $50 \pm 5\%$  RH) for the whole period of the aging cycles. These samples were used later as a reference.
- The second part of the specimens was exposed to the accelerated aging cycles that consist of two parts: heat – rain and heat – cold cycles that are described below:
  - ✓ Heat - rain cycles: Samples are subjected to a series of 80 cycles, comprising the following phases: 1- heating to 70°C (rise for 1 hour) and maintaining at  $(70 \pm 5)$ °C and 10 to 30% RH for 2 hours, 2- spraying for 1 hour (water temperature at  $+ 15 \pm 5$  °C, and amount of water 1 l/m<sup>2</sup> min), 3 – leave for 2 hours (drainage). These cycles are represented in Figure 4-B.
  - ✓ Heat-cold cycles: After a subsequent conditioning of at least 48 hours at temperatures between 10 and 25 °C and a minimum relative humidity of 50 %, the same test rig is exposed to 5 heat/cold cycles of 24 hours comprising the following phases: 1- exposure to  $(50 \pm 5)$ °C (rise for 1 hour) and maximum 30% RH for 7 hours (total of 8 hours), 2- exposure to  $(- 20 \pm 5)$ °C (fall for 2 hours) for 14 hours (total of 16 hours) (Figure 4-C).

Samples of two batches (those used as reference and those subjected to the accelerated aging cycles) were characterized and compared later to assess the aging effect. As hemp mortar is used as insulation material with rendering, it is protected against direct access of water during the accelerated



aging cycles. That is why in this study, we used a roofing to avoid direct spraying of water on the samples, as shown in Figure 5.

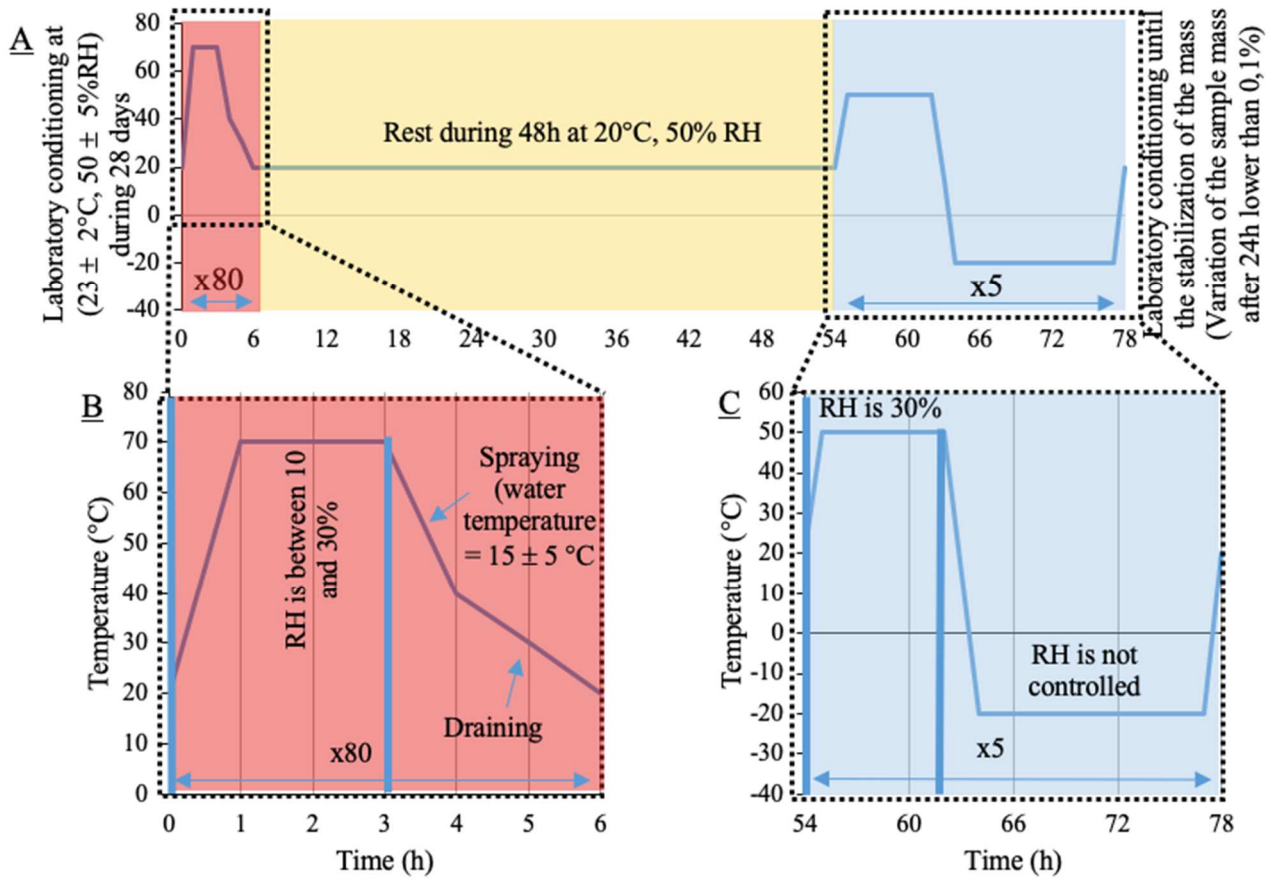


Figure 4. Diagram of the total accelerated aging cycles (A) with detailed heat-rain (B) and heat-cold (C) cycles



Figure 5. Disposition of the hemp mortar specimens in the climatic room before accelerated aging cycles

### 2.3. Experimental procedure

This paper investigates the evolution of the hygrothermal properties, the microstructural and the chemical composition of the hemp mortar before and after aging cycles. Thermal conductivity, water vapor permeability and moisture buffer value (MBV) tests were conducted to analyze the evolution of the properties of the material. Identification of total porosity, pH level values, scanning electron microscope SEM and X-ray crystallography analysis provided more information about microstructural evolution. Experimental protocols for each test are proposed further. For each type of characterization test, three specimens were analyzed before and after the weathering aging cycles. The overall protocol schema is presented in Figure 6.

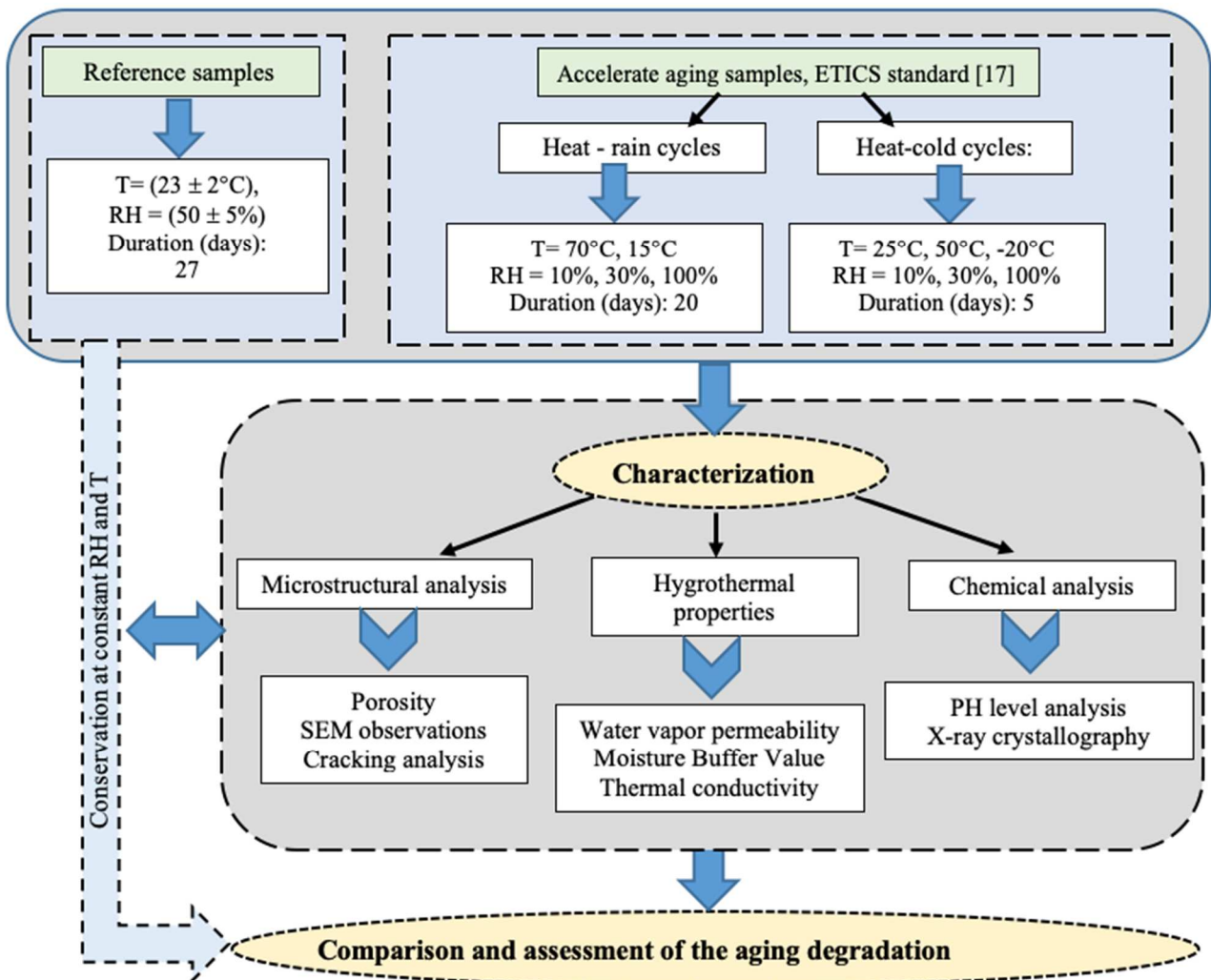


Figure 6. Experimental protocol schema

#### 2.3.1. Microstructural characteristics and chemical analysis



### ***The total porosity tests***

In order to measure the total porosity, the water porosity test was used accordingly the NFP 18-459 standard [18]. It is based on the hemp mortar saturation in water under vacuum. Then different weightings were done to calculate the total porosity of the samples. The scales sensitivity was 0.01 g. One of the weightings was the weighting of wet sample. As hemp mortar becomes fragile after wetting, we used a cotton tissue in order to protect the wet sample from damage (Figure 7). Three specimens (15x15x7 cm<sup>3</sup>) were tested for each time of conditioning to validate the repeatability of the test.

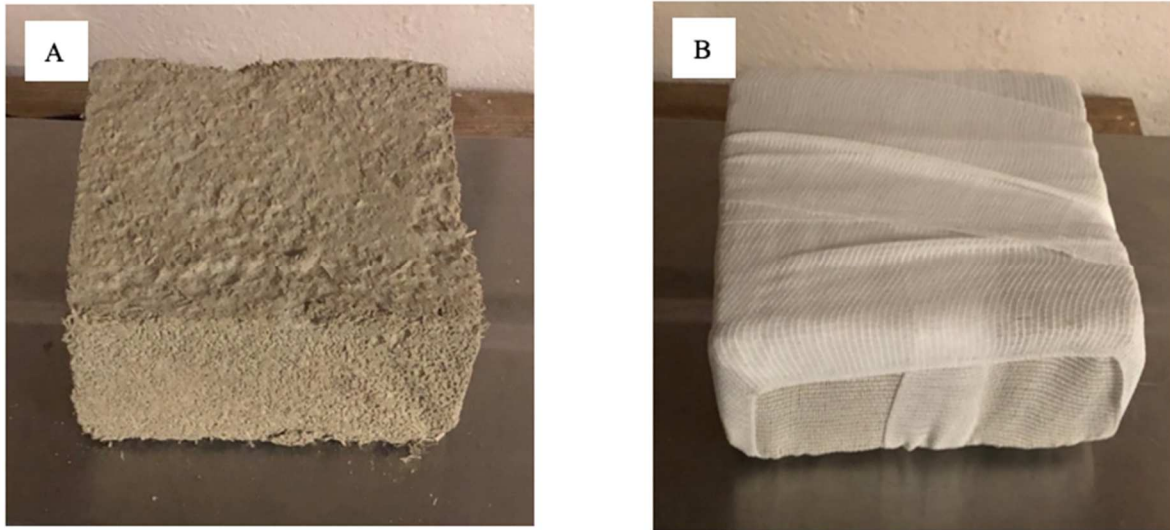


Figure 7. Hemp mortar samples without (A) and with (B) protective cotton tissue

### ***Scanning Electron Microscopy (W-SEM) microstructural analysis***

Scanning electron microscope (W-SEM) analyses were performed by using a Hitachi S-3400N device. The surface of the hemp mortar and the interface zone between cementitious matrix and hemp particles were observed and analyzed before and after aging. Particular attention has been devoted to the identification of the same observation area for these two states.

Micrographs were recorded by using secondary electrons detectors at an acceleration voltage of 15 kV, in high vacuum mode (pressure <1Pa). Previously to observations, the specimens were covered by carbon in order to improve the image quality.

### ***PH level evolution***

There are a lot of different methods of the identification of a pH level. Some authors identify a pH level of interstitial solution by compression that allow to get the most reliable results as we don't modify the pH level adding water [19]. Nevertheless, this method is a heavy procedure that requires special equipment and a large quantity of cement. Others propose another method – of cementitious suspensions [20]. It consists of suspending of some part of the mortar in water and measuring a pH level of obtained suspension. At the same time, the use of suspensions implies problems of dissolution

of the solid phases but especially of dilution, moreover if talking about hygroscopic composite material. That is why in this work a surface electrode WTW Sentix Sur was used [21]. It allowed us to get a reliable result and especially on different parts of samples to evaluate a chemical degradation of the mineral matrix.

In order to better understand the degradation of the samples, we have investigated two different areas for the measurement of pH: the surface and the middle of the sample. The idea is to understand whether or not the degradation caused by hygrothermal stresses has actually propagated inside the sample or not. In fact, most of the works in the literature studying hemp mortars do not pay attention to the position of pH measurement in the sample. Nevertheless, it is important as the chemical degradation starts from the surface and is a long process.

For this test the specimens of the same composition, made with the same raw materials, of the size  $16 \times 4 \times 4 \text{ cm}^3$  were used. For each value three measures were performed at different positions on the specimen. As the pH level measurements are possible just in solution, the constant volume of demineralized water (0.2 ml) was deposited on the studied surfaces. Then, the electrode was kept on the sample surface, so that the electrode's membrane remains in contact with the hemp mortar (Figure 8). At equilibrium, the WTW Sentix Sur displays a constant value of PH which will be subsequently recorded. For the inner pH level measures the protocol is the same, but the measurements were conducted in the middle of the specimen.



Figure 8. Measurement of the surface pH level using a WTW Sentix Sur electrode

#### ***X-ray diffraction (XRD) crystallography analysis***

Prior to XRD crystallography analysis, hemp mortar samples of the size  $16 \times 4 \times 4 \text{ cm}^3$  were grinded and sieved to remove organic fibers. This step allowed us to decrease the influence of the dilution on the final result as in this case we analyze only mineral matrix. To link the results of chemical composition to the pH level, the crystallography at the surface and in the middle was analyzed.

Free water was removed using the acetone. Then, the specimens were dried at 50 °C in order not to modify the composition of the sample. XRD crystallography patterns were obtained using Cu K $\alpha$  anode tube ( $\lambda= 1.54182 \text{ \AA}$ ) radiation at 40 kV and 20 mA with a Bruker D8 diffractometer (Bruker, Karlsruhe, Germany). The diffractometer was scanned from 5° to 60° ( $2\theta^\circ$ ) in step size of 0.015° and the counting time per step was 1.2 s.

### 2.3.2. Hygrothermal properties evolution

#### ***The water vapor permeability (WVP) (wet cup method)***

The water vapor permeability is the ability of a material to transfer or retain water vapor due to a difference in the partial pressure of water vapor at the same atmospheric pressure on both sides of the material. It was identified according to the NF EN ISO-12572 standard using the wet cup method [22]. Firstly, specimens of diameter 11 cm and thickness of 2.4 cm (3 samples for each test) were dried at 23 °C, 50% RH during 28 days. Then, they were sealed on cups containing a saturated salt solution that ensures constant relative humidity in the cup (Figure 9). In the case of wet cup, ammonium dihydrogen phosphate was used that maintains the relative humidity at  $93\pm 1\%$ . The cups are conditioned in a constant ambience of 23 °C and 50% RH.

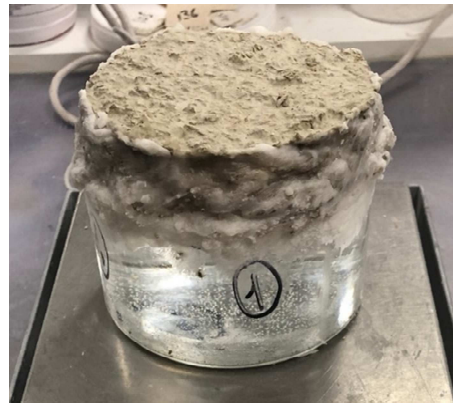


Figure 9. Hemp mortar sample sealed on the experimental cup

The water vapor permeability value was determined in function of the vapor transmission through the specimen. To do so, the mass monitoring was conducted every day during 18 days for each cup to calculate the mass variation velocity:

$$\Delta\dot{m}_{12} = \frac{m_1 \cdot m_2}{t_1 \cdot t_2} \quad (1)$$

where,  $\Delta\dot{m}_{12}$  is a mass variation velocity (kg/s);  $m_1$  and  $m_2$  are masses of the cups with hemp mortar samples at the time  $t_1$  and  $t_2$  respectively (kg);  $t_1$  and  $t_2$  are the successive times of weighting (s).

Then, the water vapor flux density was calculated:

$$g = \frac{G}{A} \quad (2)$$

where, G is an average value of five successive measurements of  $\Delta\dot{m}_{12}$  and A is an exposed area (m<sup>2</sup>).

The water vapor permeance was calculated as

$$W = \frac{G}{A \cdot \Delta p} \quad (3)$$

where,  $\Delta p$  is difference of water vapor during the test.

The water vapor resistance is an inverse value to permeance:

$$Z = \frac{1}{W} \quad (4)$$

In fact, there are several resistances in the experimental montage:

$$Z_m = Z_a + Z_s + Z_1 + Z_2 \quad (5)$$

where,  $Z_m$  is an experimental calculated value of resistance,  $Z_a$  is a water vapor resistance of air,  $Z_1$  and  $Z_2$  are the water vapor resistances that take into account the interface diffusion at the sample/air and air/saline solution interfaces respectively.

Finally, the water vapor permeability value was calculated:

$$\delta = \frac{d}{Z_s} \quad (6)$$

where,  $d$  is a thickness of our sample (mm).

### ***The thermal conductivity tests***

For measuring thermal conductivity, there are two types of experimental methods: steady-state and non-steady-state ones. When talking about measurement setups, we can distinguish four main types: the guarded hot plate (GHP), the heat-flow meter (HFM), the hot wire, and laser flash diffusivity [23]. In this work we used the method of hot wire as it is well adapted for bio-based materials. Thermal conductivity measurements were taken on three different pairs of the 15x15x7 cm<sup>3</sup> samples under transitory conditions using the device CT-Metre (Figure 10). Before measurements, these samples were conditioned at 23 °C and 50 % RH until a constant mass, according to [24].

Measurements were made according to the device precautions on the measurements. The used protocol consists of placing a probe between two samples keeping the maximal parallelism of the surfaces to avoid the distortion of results. The thermal conductivity values were calculated by the CT-Metre automatically, coefficients of correlation of presented results were 0.999.



Figure 10. CT-Metre measurement device

### ***Moisture buffer value (MBV) tests***

The moisture buffer value represents the ability of the material to mitigate the relative humidity changes in ambient air. It refers to the amount of water that could be absorbed/released when exposed to relative humidity change. In this paper, the Nordtest protocol was used to investigate the MBV of the studied hemp mortar. During this test the samples of hemp shives placed in the aluminum recipient of 10x10x3 cm<sup>3</sup> size and hemp mortar samples of 15x15x7 cm<sup>3</sup> size were used. To do this, variable cycles of relative humidity of 75% (for 8 hours) and 33% (for 16 hours) were applied. Samples were weathered at laboratory conditions (23 ± 2 °C, 50 ± 5% RH) beforehand. The MBV value was calculated:

$$MBV = \frac{\Delta m}{S \cdot (HR_{max} - HR_{min})} \quad (7)$$

where  $\Delta m$ : sample mass variation [kg],

S: exposed area of sample [m<sup>2</sup>],

$HR_{max}$ : maximal relative humidity [%],

$HR_{min}$ : minimal relative humidity [%].

According to [25], building materials are classified using the MBV range represented in Figure 11.

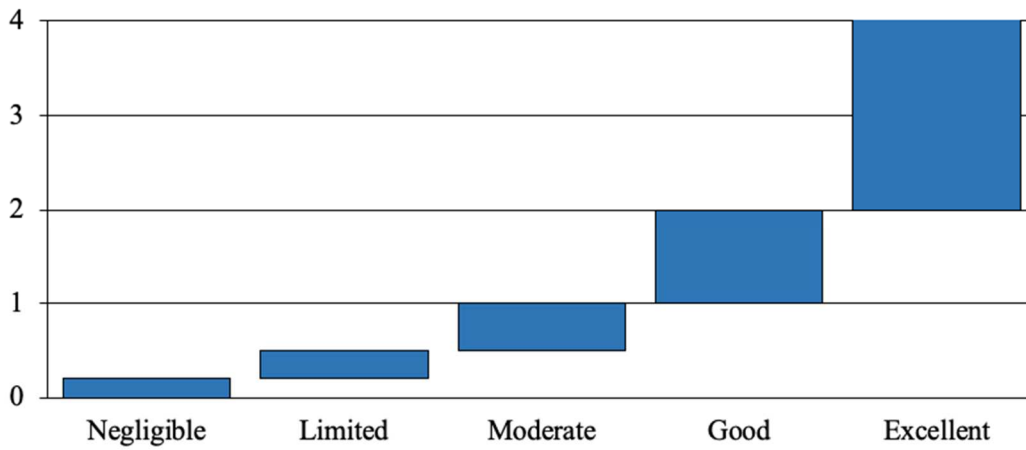


Figure 11. Moisture buffer value [g/(%RH.m<sup>2</sup>)] standardization according to Nordtest protocol [25]

### 3. Results and discussions

In this section, the assessment of the microstructure, the chemical composition and its hygrothermal performance evolution under the accelerated aging cycles are presented.

#### 3.1. Microstructural characteristics and chemical analysis

In order to study the microstructure evolution, firstly the total porosity was analyzed before and after accelerated aging cycles. Then, the interface between cement matrix and hemp particles was observed and the evolution of the chemical composition was studied. The analysis of these results was complemented by SEM.

We note that the studied hemp mortar has a porosity of 85.2% which is more important than the other mortars, presented in literature [10]. This is explained by the organic fibers porosity and the properties of the cementitious matrix that ensure good insulation properties.

According to the results given in Table 3, weathering aging cycles affect very slightly the total porosity, increasing the value from 85.2% to 85.8%. This result remains in the range of a standard deviation. Such slight evolution of porosity could be explained by the dimensional variation of the hemp mortar due to hygrothermal solicitations. In fact, hygrothermal stresses cause swelling or shrinkage of the organic fiber causing cracking of the interface and, consequently, an increase in porosity. At the same time there is another effect – a carbonation of mineral matrix and such porosity change could be explained by competition between these two phenomena. Both effects are investigated in detail.

Table 3. Total water porosity values (%) evolution of hemp mortar samples before and after the weathering aging cycles

Values	Before the weathering aging cycles	After the weathering aging cycles



Total water porosity value of the sample 1 (%)	83.9	87.7
Total water porosity value of the sample 2 (%)	86.5	84.8
Total water porosity value of the sample 3 (%)	85.2	84.9
Average total water porosity value (%)	85.2	85.8
Standard deviation (%)	$\pm 0.87$	$\pm 1.23$

In order to study the hemp mortar cracking before and after proposed weathering aging cycles, the W-SEM observation of the hemp particles / cementitious matrix interface was conducted. To do this, the surface areas of 2 samples for each type of conditioning were observed. For each sample, 10 interfaces were examined and the crack sizes were measured in 10 spots all along the crack. example of the image before and after aging are present in Figure 12. We notice the cracking of the hemp mortar and mineral matrix interface that show the nature of the hemp mortar damage. That explains the results of the water vapor permeability evolution. Moreover, the mean results of the crack size show the important increase of the crack size from  $5.5 \pm 1.5 \mu\text{m}$  to  $24.8 \pm 3.1 \mu\text{m}$  in the longitudinal direction of fibers. These results emphasize the influence of hygrothermal solicitations on the morphology of the hemp mortar.

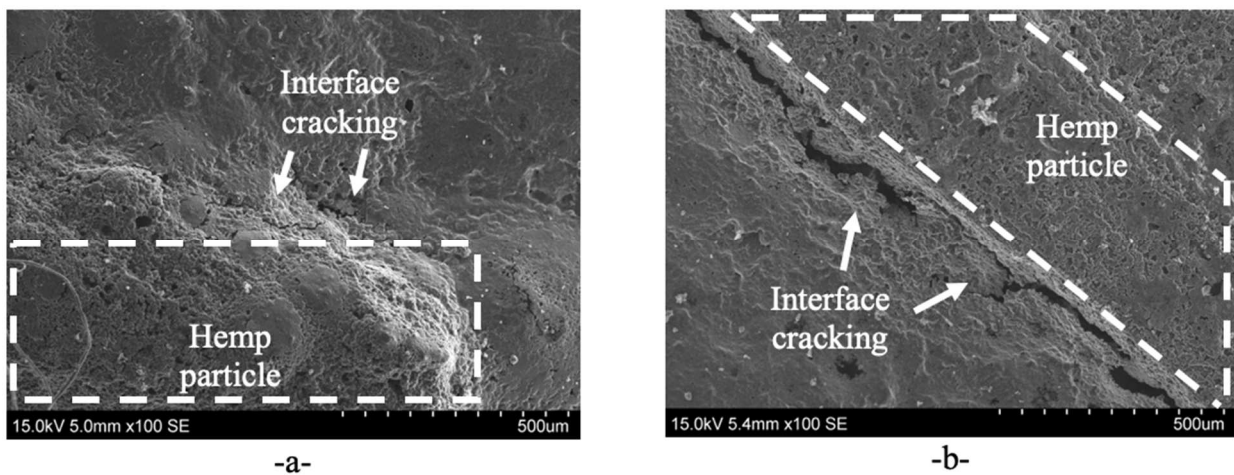


Figure 12. W-SEM observation of the surface of the specimens before (a) and after (b) the weathering aging cycles

The carbonation of the cementitious matrix was investigated by the X-ray diffraction crystallography (XRD). It was conducted for the samples before and after the weathering aging cycles

which allowed estimate the chemical composition evolution. For each specimen the analysis of the composition on the surface and in the middle of the studied samples was performed.

The XRD patterns presented in Figure 13 showed the main mineralogical compounds of the hemp mortar as  $\text{Mg}_4\text{Si}_6\text{O}_{15}(\text{OH})_2 \cdot 6\text{H}_2\text{O}$  (sepiolite),  $\text{Ca}_6\text{Al}_2(\text{SO}_4)_3(\text{OH})_{12} \cdot 26\text{H}_2\text{O}$  (ettringite),  $\text{Ca}(\text{OH})_2$  (portlandite),  $\text{CaCO}_3$  (calcite),  $\text{SiO}_2$  (quartz),  $\text{Ca}_3\text{SiO}_5$  (tricalcium silicate),  $\text{Ca}_2\text{SiO}_4$  (dicalcium silicate) and  $\text{Ca}_3\text{Al}_2\text{O}_6$  (tricalcium aluminate). Figures 13 (a) and 13 (c) present the crystallographic analysis of the hemp mortar specimen before the weathering aging cycles in the middle and on the surface, respectively. We note that after one month of drying at 23 °C and 50% RH there is non-hydrated  $\text{Ca}_3\text{SiO}_5$  (tricalcium silicate),  $\text{Ca}_2\text{SiO}_4$  (dicalcium silicate) and  $\text{Ca}_3\text{Al}_2\text{O}_6$  (tricalcium aluminate), which means that hydration is not completed. As for carbonatation of the cementitious matrix, we note that the quantity of the ettringite and portlandite is lower on the surface, is more visible in Figures 14 (I (a and c)) and 14 (II (a and c)) respectively. On contrary, Figure 14 (III-a and III-c) shows the increase of calcite.

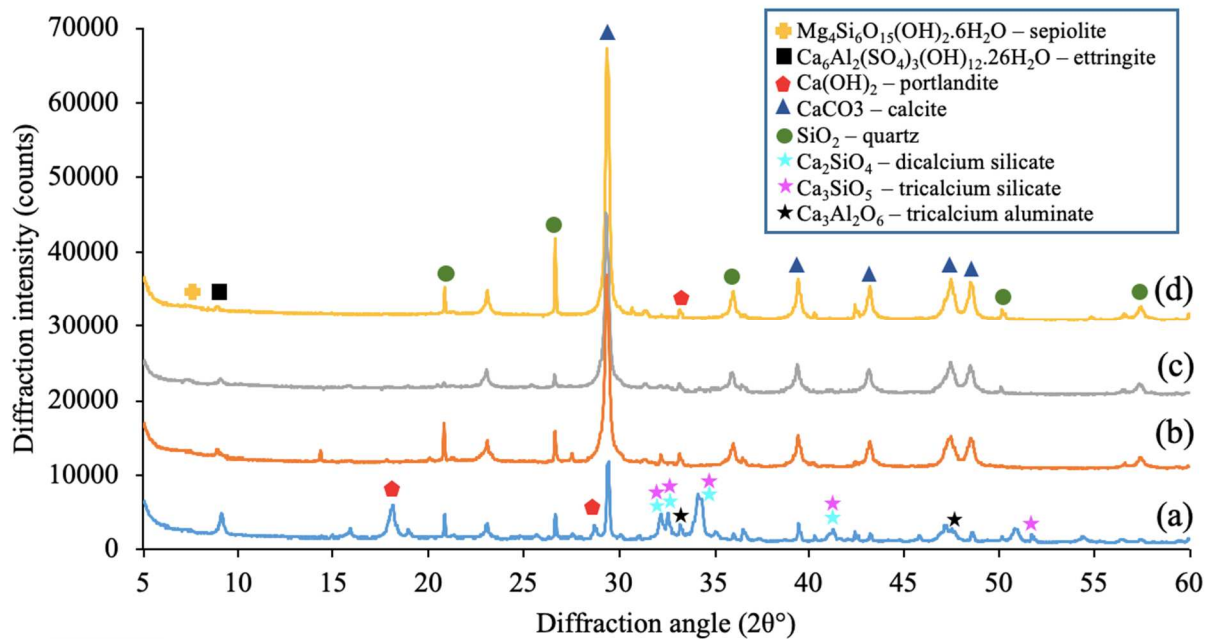


Figure 13. X-ray diffraction patterns of: (a) interior part of the hemp mortar samples before the weathering aging cycles; (b) interior part of the hemp mortar samples after the weathering aging cycles; (c) surface part of the hemp mortar samples before the weathering aging cycles; (d) surface part of the hemp mortar samples after the weathering aging cycles.

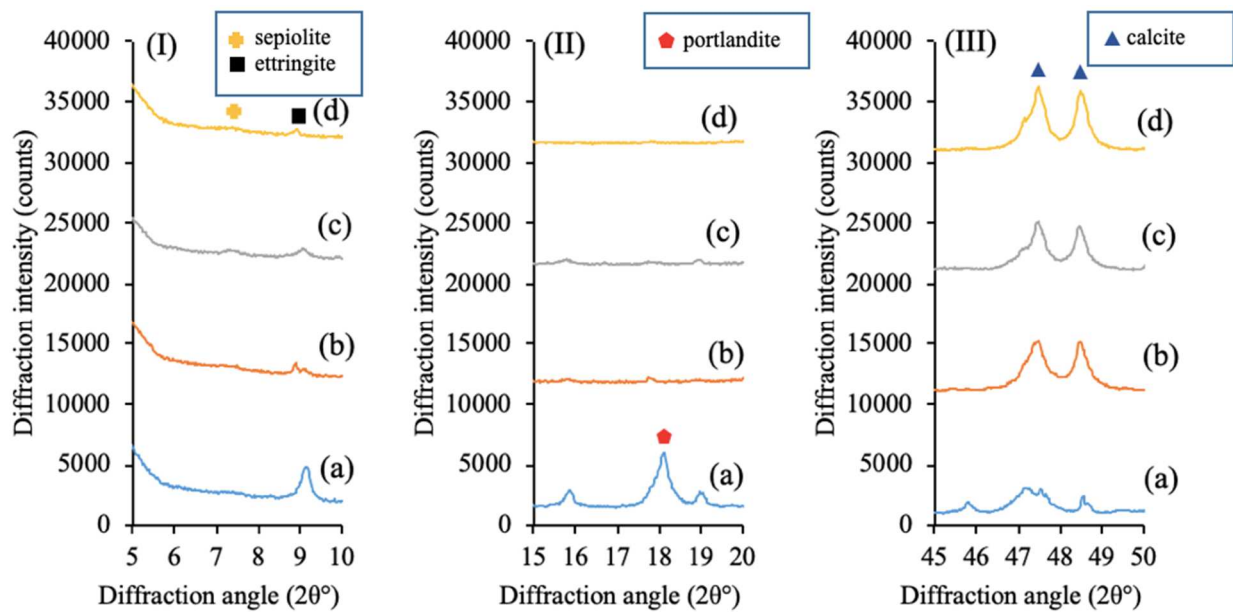


Figure 14. Zoomed X-ray diffraction patterns for ettringite (I), portlandite (II) and calcite (III) evolution: (a) interior part of the hemp mortar samples before the weathering aging cycles; (b) interior part of the hemp mortar samples after weathering aging cycles; (c) surface part of the hemp mortar samples before the weathering aging cycles; (d) surface portion of the hemp mortar samples after the weathering aging cycles.

The same pattern was noted for the sample after the weathering aging cycles. The results show that the ettringite and portlandite almost vanished in the middle and on the surface of the sample (figure 14 (I) and (II)), and calcite quantity increased (Figure 14 (III)). This shows that after the aging protocol the matrix is almost carbonated.

The carbonation of the calcium hydroxide is accompanied by a decrease in pH. That is why the measurements of the pH level were carried out to study the influence of the pH on the crystallographic composition. The results presented in Figure 15 show that the pH levels on the surface and in the middle of the sample were equal to 8.46 and 12.18 respectively before the weathering aging cycles and 8.71 and 8.69 – after. It demonstrates the effect of the carbonation of the cementitious matrix. In fact, the diffusion of the air dioxide carbon in the hemp mortar provokes its reaction with the portlandite and decrease the pH level. As the surface of the sample is exposed to the air, the quantity of portlandite at the surface is lower because of the carbonation. Later, that leads to the degradation of the ettringite, as it is very sensible to the ambient pH level.

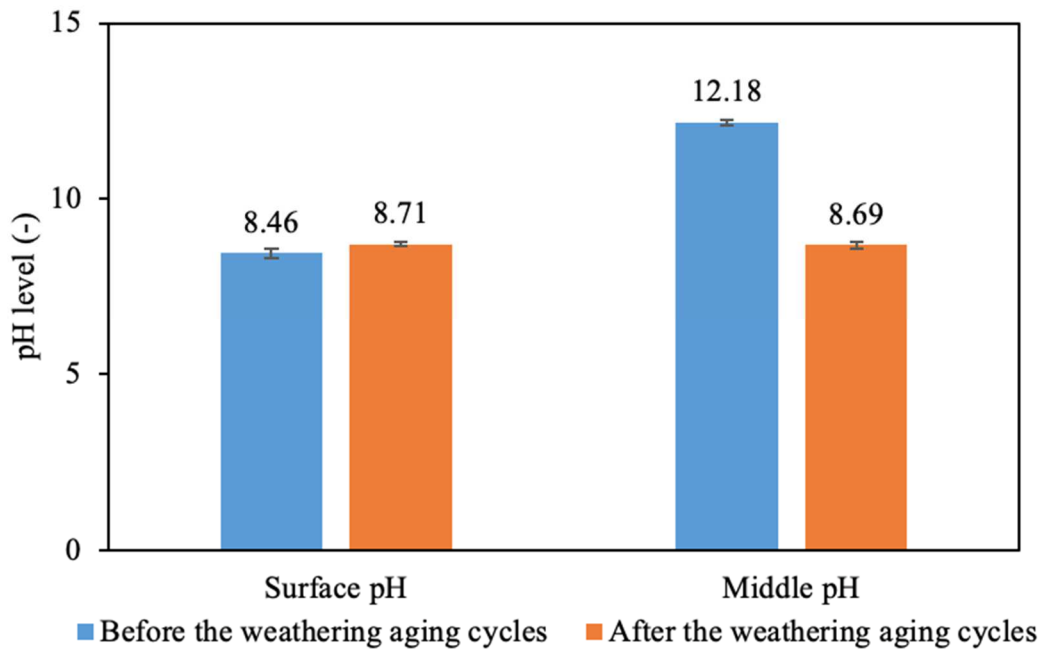


Figure 15. Diagram of pH level evolution on the surface and in the middle of the samples before and after the weathering aging cycles

### 3.2. Hygrothermal properties evolution

Hemp mortar is used for external or internal insulation applications. It is also considered as a hygroscopic material with a good ability to moderate variations in relative humidity of the external environment. The results of their evolution investigation are present below.

First, the water vapor permeability evolution due weathering aging cycles was investigated. This parameter expresses the quantity of water vapor that passes through sample's porosity. Thus, the evolution of this parameter also gives information on the evolution of the porosity and cracking caused by the hygrothermal dimensional variations after aging. The results of water vapor permeability before and after the weathering aging cycles are present in Figure 16. Firstly, one can notice that the results obtained from this study are much higher than the values measured for other formulation of the hemp mortar in the literature [10]. The measured value at the age of 30 days for wet cup method was  $2.02 \text{ E}^{-10} \text{ kg.s}^{-1}.\text{m}^{-1}.\text{Pa}^{-1}$ . That is explained by the composition, the particle size of hemp particles, and different binder types that allow to produce more aerated hemp mortar in case of studied formulation. Also, we notice the increase of the average water vapor permeability after aging from  $2.02\text{E}^{-10}$  to  $3.41\text{E}^{-10} \text{ kg.s}^{-1}.\text{m}^{-1}.\text{Pa}^{-1}$ , that justify the results of the increase of the porosity. This is explained also by the cracking of the hemp mortar, which increases its vapor diffusion capacity and thus its permeability.

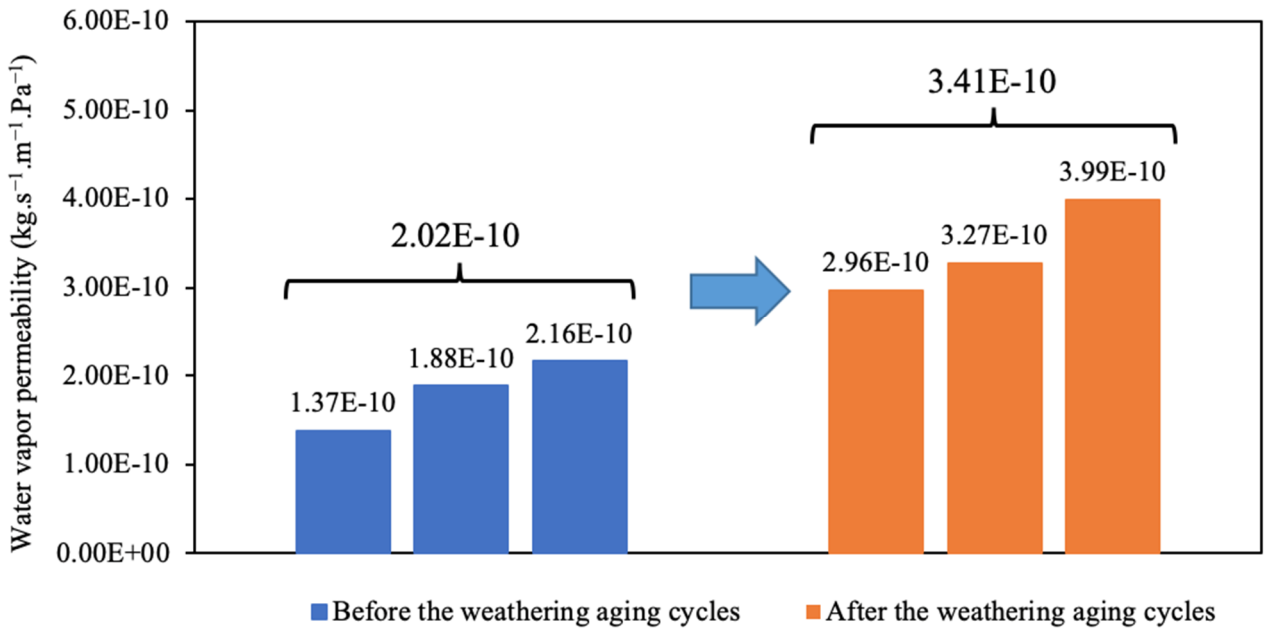


Figure 16. Water vapor permeability ( $\text{kg}\cdot\text{s}^{-1}\cdot\text{m}^{-1}\cdot\text{Pa}^{-1}$ ) evolution of hemp mortar samples before and after the weathering aging cycles

The result of the evolution of the thermal conductivity before and after aging are presented in Figure 17. The thermal conductivity was slightly increased after aging from  $0.087 \text{ W}/(\text{m}\cdot\text{K})$  to  $0.091 \text{ W}/(\text{m}\cdot\text{K})$ . In fact, this parameter is very dependent on the porosity. Thermal conductivity increased after aging because of the degradation of the plant fibers.

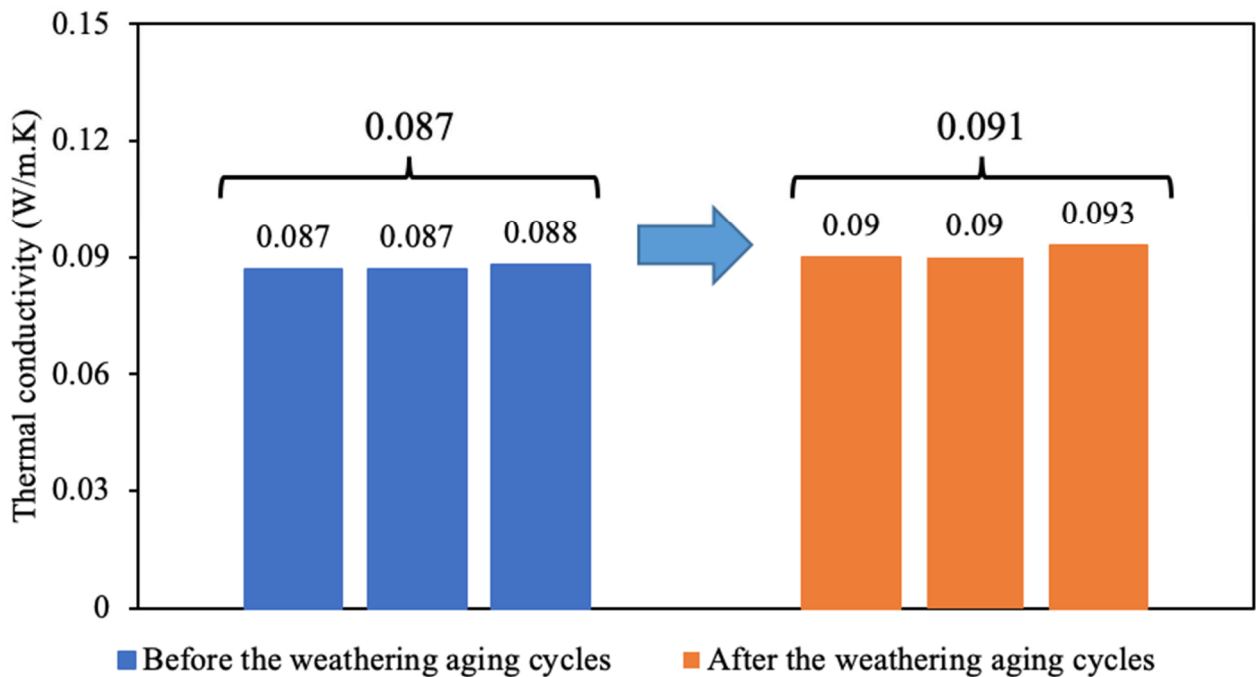


Figure 17. Thermal conductivity [ $\text{W}/(\text{m}\cdot\text{K})$ ] evolution of hemp mortar samples before and after the accelerated aging cycles

Figure 18 shows the mass and relative humidity evolution that occurred during the first 5 MBV experimental cycles for the second sample of hemp mortar before the weathering aging cycles. The temperature was held constant at 23 °C.

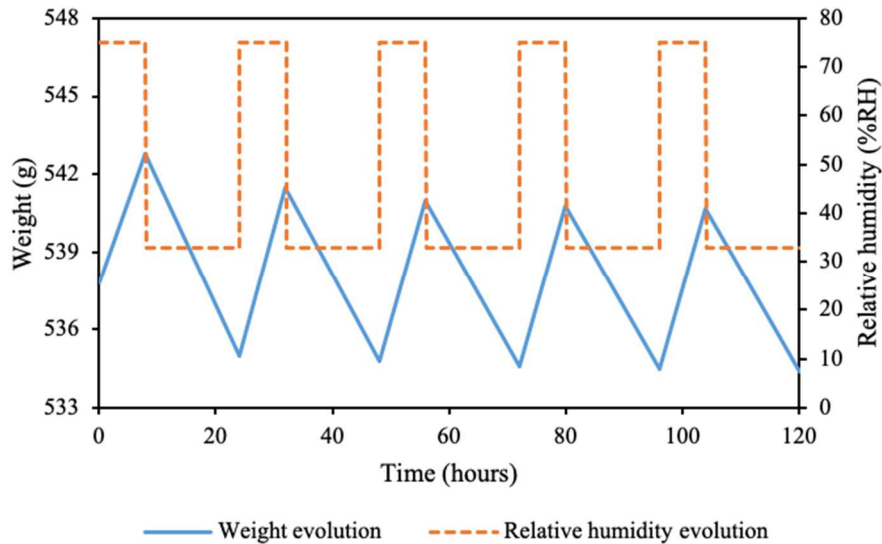


Figure 18. Masse (g) and relative humidity (%RH) evolution in function of the time

Concerning the moisture buffer value, one can note that the MBV of hemp mortar is bigger than the MBV of hemp shives before aging cycles. It is explained by the fact that the studied hemp mortar is much more aerated and lightweight than conventional hemp mortars that allows it to preserve the hygric properties of hemp shives. Nevertheless, we notice that the accelerated aging cycles did not affect the hemp shives moisture buffer value. It stays excellent with 2.25 moisture buffer value according to Nordtest protocol (Figure 11) [25]. Unlike hemp shives, hemp mortar represents the degradation of its MBV value. It decreased from 2.22  $\text{g}/(\%RH.m^2)$  to 2  $\text{g}/(\%RH.m^2)$  (Figure 19). In fact, there is the competition between the interface damage of the hemp mortar samples and the carbonatation of the mineral matrix during the hygrothermal solicitations caused by the accelerated aging cycles. So, the carbonatation of the matrix caused the closing of the microporosity and complicated the air access to the hemp particles that finally led to the moisture buffer value decrease. Despite the decrease of the moisture buffer value, we note that the MBV of the hemp mortar stays excellent.



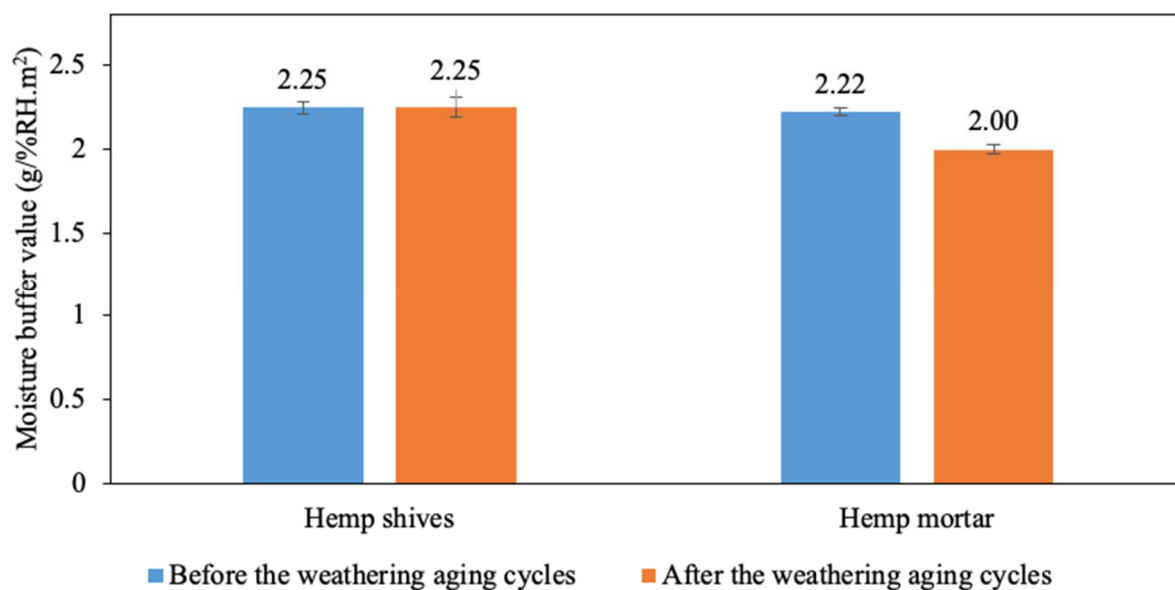


Figure 19. Moisture buffer value  $g/(\%RH.m^2)$  evolution of hemp shives and hemp mortar samples before and after the accelerated aging cycles

#### 4. Conclusion

In this work, an original accelerated weathering aging protocol inspired by the standardized one used for External Thermal Insulation Composite Systems (ETICS) with rendering was proposed. The particularity of this protocol consists in the representativeness of the real cycles of ageing applied to the building walls without damaging the material by total immersion in water. The proposed cycles of relative humidity and temperature have been studied and normalized to represent the climatic conditions of these insulation materials with rendering.

Experimentally, the hemp mortar microstructure, its composition and properties evolution were investigated. To do this, all characteristics were studied before and after the hemp mortar exposure to accelerated aging cycles. Firstly, the hemp mortar total water porosity, its water vapor permeability, its mineralogical composition and pH level evolutions were analyzed. Also, the SEM observation of the hemp particles / cementitious matrix interface was realized. Then, the characterization of evolution of the hygrothermal properties as thermal conductivity and moisture buffer value was conducted.

Results of the total porosity evaluation show a slight increase of the porosity but it remains in a standard deviation range. This is explained by a compensation of two processes that occurs and are in contest during the hygrothermal stresses: the cracking of the hemp particles / cementitious matrix interface that increase the hemp mortar porosity and the carbonatation that provoke the filing of the porosity by calcite. To study the cracking effect the evolution of water vapor permeability was analyzed. It increased from  $2.02E^{-10}$  to  $3.41E^{-10} \text{ kg.s}^{-1}.\text{m}^{-1}.\text{Pa}^{-1}$  that shows the increase of the ability to transfer a water vapor. This change was also noticed using a SEM observation because of the

increase of the interface cracks on the surface of the mortar. The XRD analysis demonstrated the carbonation of the mineral matrix – the increase of the quantity of the calcite and decrease of the quantity of the portlandite and ettringite due to the aging cycles. The carbonation was also demonstrated by the study of pH level evolution since the pH level of the samples decreased to 8.69 in the middle and to 8.71 on the surface after the weathering cycles. As for the hygrothermal properties, the value of thermal conductivity dependent on the porosity follows the same as total porosity pattern – it slightly increases at the range of the standard deviation. The moisture buffer value decreases from 2.22 g/(%RH.m<sup>2</sup>) to 2 g/(%RH.m<sup>2</sup>) that is explained by the closing of the hemp mortar microporosity and complication of the air access to the hemp particles. Obviously, both the interface cracking and the carbonation processes present the considerable influence on the durability of the hemp insulating mortar. At the same time, we note that the studied hemp mortar saves their previously declared hygrothermal properties (as the thermal conductivity and the moisture buffer value). It is primordial when talking about the durability of the insulating material. As recommendation for using the proposed hemp mortar, it must be used according to the construction rules. The control of the continuity and the regularity of coating surface is a critical point because the access to water could lead to the molds proliferation.

This study provides literature with a less aggressive and more representative accelerated aging protocol with an improvement in knowledge when talking about the degradation and the hygrothermal behavior of the biosourced materials considered as very heterogeneous. These results represent a database for the durability of hemp mortars. Efforts are still needed to validate the accelerated aging tests with a comparison to an in situ experimental campaign.

## **Acknowledgments**

The authors gratefully acknowledge ParexGroup SA for financing this project.

## **References**

- [1] ADEME. Climat, Air et Energie. Chiffres clés - édition 2018 2018.
- [2] Walker R, Pavia S, Mitchell R. Mechanical properties and durability of hemp-lime concretes. *Construction and Building Materials* 2014;61:340–8. <https://doi.org/10.1016/j.conbuildmat.2014.02.065>.
- [3] El Hachem C, Ye P, Abahri K, Bennacer R. Fiber's hygromorphic effect on thermal conductivity of wooden fibrous insulation characterized by X-ray tomography. *Construction and Building Materials* 2017;150:758–65. <https://doi.org/10.1016/j.conbuildmat.2017.06.013>.
- [4] El Hachem C, Abahri K, Bennacer R. Original experimental and numerical approach for prediction of the microscopic hygro-mechanical behavior of spruce wood. *Construction and Building*

Materials 2019;203:258–66. <https://doi.org/10.1016/j.conbuildmat.2019.01.107>.

[5] Ren J, Zhao Z, Zhang J, Wang J, Guo S, Sun J. Study on the hygrothermal properties of a Chinese solar greenhouse with a straw block north wall. *Energy and Buildings* 2019;193:127–38. <https://doi.org/10.1016/j.enbuild.2019.03.040>.

[6] Panesar DK, Shindman B. The mechanical, transport and thermal properties of mortar and concrete containing waste cork. *Cement and Concrete Composites* 2012;34:982–92. <https://doi.org/10.1016/j.cemconcomp.2012.06.003>.

[7] Pacheco Menor MC, Serna Ros P, Macías García A, Arévalo Caballero MJ. Granulated cork with bark characterised as environment-friendly lightweight aggregate for cement based materials. *Journal of Cleaner Production* 2019;229:358–73. <https://doi.org/10.1016/j.jclepro.2019.04.154>.

[8] Jerónimo A, Soares C, Aguiar B, Lima N. Hydraulic lime mortars incorporating micro cork granules with antifungal properties. *Construction and Building Materials* 2020;255:119368. <https://doi.org/10.1016/j.conbuildmat.2020.119368>.

[9] Garikapati KP, Sadeghian P. Mechanical behavior of flax-lime concrete blocks made of waste flax shives and lime binder reinforced with jute fabric. *Journal of Building Engineering* 2020;29:101187. <https://doi.org/10.1016/j.jobbe.2020.101187>.

[10] Bennai F, Nabil I, Abahri K, Belarbi R, Tahakourt A. Experimental characterization of thermal and hygric properties of hemp concrete with consideration of the material age evolution. *Heat and Mass Transfer* 2017. <https://doi.org/10.1007/s00231-017-2221-2>.

[11] Bennai F, El Hachem C, Abahri K, Belarbi R. Influence of hydic solicitations on the morphological behavior of hemp concrete. *RILEM Technical Letters* 2019;4:16–21. <https://doi.org/10.21809/rilemtechlett.2019.80>.

[12] Bennai F, El Hachem C, Abahri K, Belarbi R. Microscopic hydic characterization of hemp concrete by X-ray microtomography and digital volume correlation. *Construction and Building Materials* 2018;188:983–94. <https://doi.org/10.1016/j.conbuildmat.2018.08.198>.

[13] Sentenac C, Sonebi M, Amziane S, Pascal UB, Pascal I, Clermont-Ferrand P. Investigation on the performance and durability of treated hemp concrete with linseed oil 2017;35:9.

[14] Marceau S, Glé P, Guéguen-Minerbe M, Gourlay E, Moscardelli S, Nour I, et al. Influence of accelerated aging on the properties of hemp concretes. *Construction and Building Materials* 2017;139:524–30. <https://doi.org/10.1016/j.conbuildmat.2016.11.129>.

[15] Delannoy G, Marceau S, Glé P, Gourlay E, Guéguen-Minerbe M, Amziane S, et al. Durability of hemp concretes exposed to accelerated environmental aging. *Construction and Building Materials* 2020;252:119043. <https://doi.org/10.1016/j.conbuildmat.2020.119043>.

[16] Piot A, Béjat T, Jay A, Bessette L, Wurtz E, Barnes-Davin L. Study of a hempcrete wall exposed to outdoor climate: Effects of the coating. *Construction and Building Materials*

2017;139:540–50. <https://doi.org/10.1016/j.conbuildmat.2016.12.143>.

- [17] EOTA. Guideline for European technical approval of external thermal insulation composite systems (ETICS) with rendering 2013.
- [18] AFN. NF P18-459 - Concrete - testing hardened concrete - testing porosity and density 2010.
- [19] Alonso C, Fernandez Luco L, García J, Hidalgo A, Huertas F. Low-pH Cementitious Materials Design and Characterisation 2007.
- [20] Codina M. Les bétons bas pH - Formulation, caractérisation et étude à long terme. phdthesis. INSA de Toulouse, 2007.
- [21] Tran TH, Govin A, Guyonnet R, Grosseau P, Lors C, Damidot D, et al. Influence of the intrinsic characteristics of mortars on their biofouling by pigmented organisms: Comparison between laboratory and field-scale experiments. *International Biodeterioration & Biodegradation* 2014;86:334–42. <https://doi.org/10.1016/j.ibiod.2013.10.005>.
- [22] ISO 12572:2016 Hygrothermal performance of building materials and products — Determination of water vapour transmission properties — Cup method 2016.
- [23] Yüksel N. The Review of Some Commonly Used Methods and Techniques to Measure the Thermal Conductivity of Insulation Materials. *Insulation Materials in Context of Sustainability* 2016. <https://doi.org/10.5772/64157>.
- [24] ISO 12571:2000(E) Hygrothermal performance of building materials and products. Determination of hygroscopic sorption properties: n.d. <https://doi.org/10.3403/30259581>.
- [25] NORDTEST project. BYG DTU R-126 Moisture buffering of building materials 2005.