

UNRAVELING COUPLED SALT TRANSPORT AND DAMAGE MECHANISMS IN POROUS LIMESTONE USING 4D X-RAY TOMOGRAPHY AND ADVANCED IMAGE ANALYSIS

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How to Cite:

Ben Elhadj Hamida, S., Boone, M. A., Cnudde, V., Moonen, P., & Derluyn, H. (2026). Unraveling Coupled Salt Transport and Damage Mechanisms in Porous Limestone from Core to Pore Scale using 4D X-ray Tomography and Advanced Image Analysis. *InterPore Journal*, 3(3), IPJ020926-4. <https://doi.org/10.69631/2337kj19>

RECEIVED: 30 Nov. 2025

ACCEPTED: 31 Jul. 2026

PUBLISHED ONLINE: 02
Sept. 2026

VERSION OF RECORD: 02
Sept. 2026

ABSTRACT

In this study, time-resolved X-ray microtomography is employed to improve the understanding of coupled hydro-chemo-mechanical processes involved in saline weathering of natural stone. A Savonnières limestone sample, hydrophobically treated at its upper surface, was saturated with a NaCl solution and subjected to a controlled drying and deliquescence cycle while being imaged at a resolution of 9.4 $\mu\text{m}/\text{voxel}$. The volumetric deformations, induced during the cycles, were extracted from the time-resolved 3D images using Digital Volume Correlation (DVC). In combination with deep-learning-based segmentation of distinct pore families, including those below the image resolution, the associated transport and crystallization mechanisms are elucidated at the pore scale. The results reveal that salt crystallization in the hydrophobized region of the sample generates internal stresses leading to fracture formation, while deliquescence promotes partial crack closure due to matrix swelling and salt dissolution. In the untreated part of the sample, shrinkage and swelling was observed during drying and deliquescence, respectively. A chronology of the kinetics of these transport processes was further identified across the different pore families. Overall, this work demonstrates that the combination of X-ray microtomography with advanced image-processing techniques enables in-depth analysis of complex, intertwined phenomena in porous media, even when they occur partly in pores smaller than the image resolution.

KEYWORDS

Salt transport & in-pore salt crystallization; Porous limestone; X-ray microtomography; Digital volume correlation; Deep-learning segmentation

WHY THIS PAPER MATTERS

Salt silently cracks stone buildings and monuments from the inside out, threatening cultural heritage worldwide. Using X-ray imaging without a contrast agent, we developed a novel analysis workflow that reveals how salt is transported and creates cracks at a microscopic scale. This workflow opens new ways for scientists to study similar complex processes in porous materials. The insights inform conservation experts and engineers as they develop strategies to protect materials, buildings, and structures from salt weathering.



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1. INTRODUCTION

The crystallization of salts causes significant weathering of various porous media, particularly of rocks and building stones. More precisely, saline solutions, naturally present in underground reservoirs or infiltrating building structures by capillarity, can become more concentrated due to changes in environmental conditions, leading to supersaturation and salt crystallization. This phenomenon generates a crystallization pressure (37) sufficient to damage rocks or construction materials, hereby creating new pathways for fluid transfer and sustaining the degradation mechanism. The damaging effects of salts on building stones have been widely investigated in the field of construction engineering and conservation science, with sodium sulfate (Na_2SO_4) and sodium chloride (NaCl) being the most studied salts (7, 8, 12, 13, 21, 24, 27, 28, 29, 33, 35). In the field of CO_2 storage, few studies demonstrate precipitation-induced rock damage under representative reservoir conditions. A notable exception is the study by Nooraiepour et al (25), who found that CO_2 -induced salt precipitation leads to a marked impact on rock stiffness and deformation behavior. Furthermore, a review by Hajiabadi et al (16), mentioned that the forces arising from salt crystallization during CO_2 injection are strong enough to reach the tensile strength of porous rocks and cause the formation of microcracks and secondary fracture networks. The observations are hence similar for both application domains, regardless of the significant differences in pressure and temperature. Laboratory experimental studies exploring the damaging effects of salt or mineral crystals under ambient conditions therefore appear to be also relevant for CO_2 injection into saline aquifers.

Experimental studies conducted in the laboratory typically focus on the macroscopic behavior of specimens the size of small building blocks or rock cores. Fewer studies explore the complex interaction between transport phenomena, salt precipitation, crack initiation and propagation at the pore-scale. To study these interconnected phenomena in porous media, X-ray microtomography (micro-CT) has proven to be a very powerful technique (6), allowing the non-invasive morphological characterization of a three-dimensional (3D) volume of the scanned sample. However, discriminating phases featuring poor attenuation contrast, such as calcite and sodium chloride, or quartz and sodium sulphate, remains challenging. To overcome this, a commonly used method involves the addition of contrast agents. These agents, which contain high X-ray attenuation elements, such as iodine-based contrast agents (19), improve differentiation of various phases in samples (23). However, it is important to note that these contrast agents may potentially influence the physical processes of interest. Furthermore, as is the case for all image-based techniques, the smallest observable detail is at most about three to four orders of magnitude smaller than the field of view. The pore sizes within natural stones such as limestone cover an even wider range, implying that a single acquisition cannot cover the full pore size range. Acquisitions of representative sample volumes therefore contain unresolved porosity.

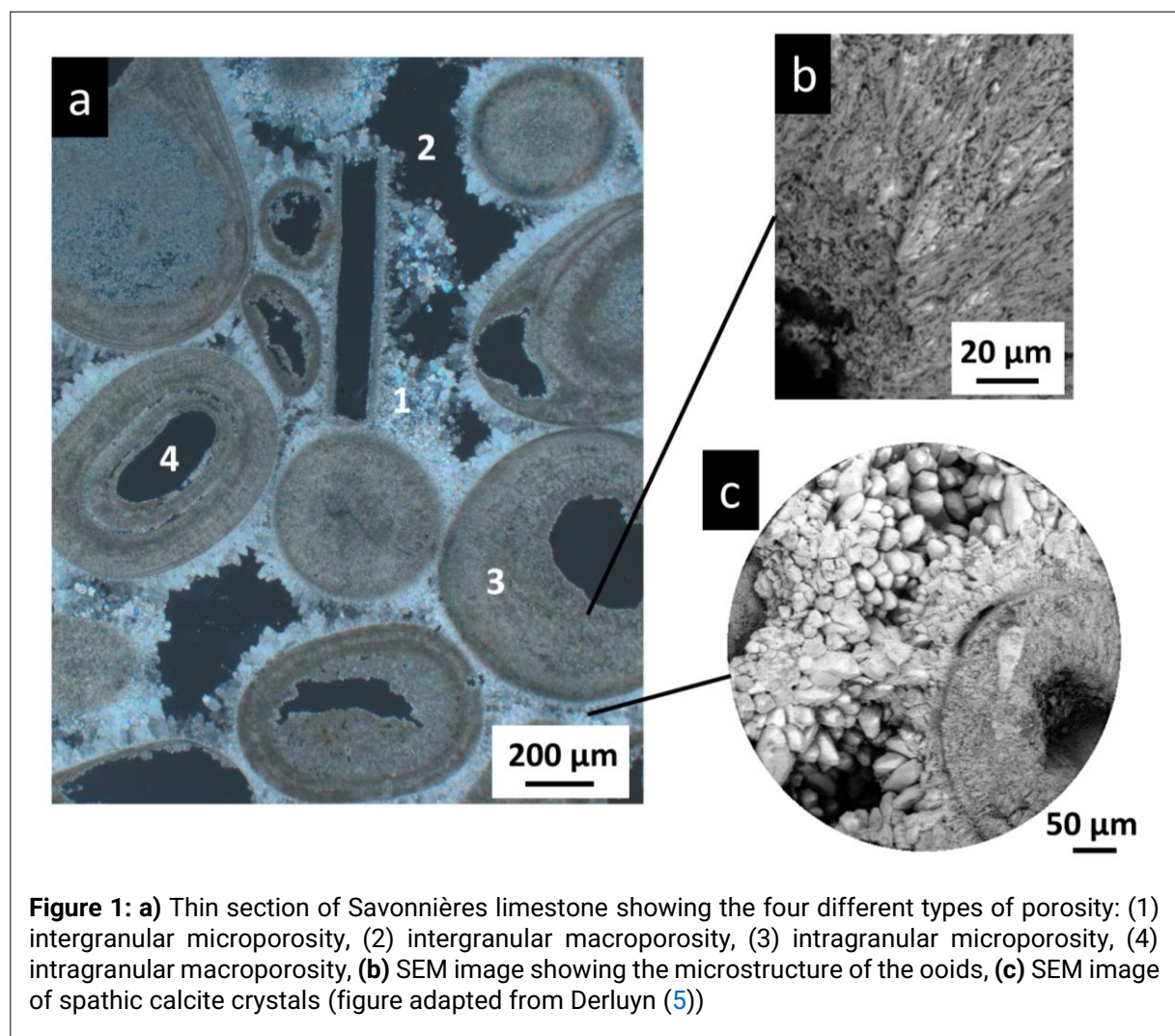
This study serves a double aim. First and foremost, we strive to deepen our understanding of transport, crystallization, and fracturing processes in porous media in the presence of sodium chloride, the most commonly encountered salt in the civil engineering as well as geoenvironmental fields (17, 18, 38). Secondly, we show how advanced image analysis techniques can overcome the challenge of characterizing phenomena that occur partly in regions with pores smaller than the image resolution.

The paper starts in Section 2 with a description of the sample, the experiment, and the components of the image analysis workflow. Section 3 focuses on the obtained results, demonstrating that a very complete picture of the intertwined mechanisms can be obtained from a single 4D acquisition sequence, even for phenomena occurring at scales below the image resolution in phases featuring hardly any X-ray contrast. The conclusions (Section 4) summarize our findings and discuss their relevance.

2. MATERIALS AND METHODS

2.1. Savonnières limestone and sodium chloride

The Savonnières rock is an Upper Jurassic limestone found predominantly in the northeastern region of France. Savonnières limestone has been used in the construction of historic monuments in France, including the prefecture of Nancy, the Notre-Dame church in Paris, and the Notre-Dame church in Saint-Dizier (9). Internationally, Savonnières stone has adorned emblematic buildings, including Frankfurt's former opera house in Germany and the town hall of Leuven in Belgium (4).



The rock has also been widely used in research focusing on the properties of various subsurface reservoirs, as well as in studies on CO₂ storage in saline aquifers (20, 26, 34).

Savonnières is composed almost entirely of calcite, with a CaCO₃ concentration reaching 98% (15). It is an oolitic stratified sedimentary rock. The ooids form around a nucleus and exhibit a radial and concentric structure, with nuclei that can be either filled or hollow. The cement, filling the pores between the ooids, consists of spathic calcite crystals arranged radially around the grains. This arrangement leads to the creation of triangular-shaped pores. Overall, this heterogeneous rock presents four types of porosity (14, 31): **1)** intergranular microporosity, **2)** intergranular macroporosity, **3)** intragranular microporosity and **4)** intragranular macroporosity (see Fig. 1).

Roels et al. (31) characterized the pore volume distribution of the rock, revealing that the intragranular microporosity of the ooids (type 3) presents the smallest pore sizes below 1 µm, followed by the intergranular micropores between the spathic calcite crystals (type 1), with pore sizes in the order of 10 µm, and finally the inter- and intra-granular macropores (type 2 and 4), with pore sizes in the order of 100 µm.

Sodium chloride (NaCl) is a salt with a single stable anhydrous solid phase above 0°C, namely halite. This phase begins to precipitate at concentrations around 6 molal, representing the solubility limit of NaCl in water, which is only slightly temperature dependent. Below this concentration, the salt remains fully dissolved. In addition, NaCl begins to deliquesce at around 75% relative humidity (RH) under standard atmospheric pressure. Deliquescence refers to the phase transition that occurs when a deliquescent solid, such as NaCl, absorbs water vapor from the atmosphere and dissolves to form an aqueous solution. This process begins at a specific relative humidity, known as the deliquescence point, above which the salt solution becomes thermodynamically stable (11, 22). At this threshold, solid NaCl can coexist with its saturated solution, whereas above it, only the liquid phase remains, and below it, only the solid phase remains.

2.2. Crystallization-deliqescence experiment

The considered experiment focuses on a cylindrical sample of Savonnières rock with a diameter of 8.4 mm and a height of 9 mm. A hydrophobic treatment for limestone based on silane/siloxane^a was applied to the upper surface of the sample in order to prevent efflorescence (similar to the sample preparation by Derluyn et al in (7)). The treatment penetrated the upper part of the stone over a depth of about 2.5 mm. Such treatments are often used in the conservation of historical monuments to prevent water infiltration. Furthermore, in the context of carbon dioxide (CO₂) injection into saline aquifers, the interaction between CO₂, water, and the rock minerals can also lead to wettability alterations (1, 2). After applying the hydrophobic treatment, aluminum tape was carefully wrapped around the sample's circumference, while leaving the top and bottom faces exposed to the surrounding environment, with the purpose of sealing the sample laterally to prevent drying and salt efflorescence via these surfaces as well as the loss of small rock debris. The sample was subsequently put in contact with a 6 molal NaCl saline solution at its base. Capillary imbibition took place over a period of 1.5 hours under room conditions after which the bottom face was also covered with aluminum tape, so that drying could only occur through the hydrophobically treated top side. The capillary saturated sample was then placed in a cylindrical plastic cell connected to a humidity generator^b and equipped with an RH-T sensor^c for independent climate monitoring.

^a SILRES® BS280 from Wacker; <https://www.wacker.com/h/en-us/silanes-siloxanes-silicates/silane-siloxane-mixtures/silres-bs-280/p/000010604>

^b GenRH from Surface Measurement Systems: not in production anymore

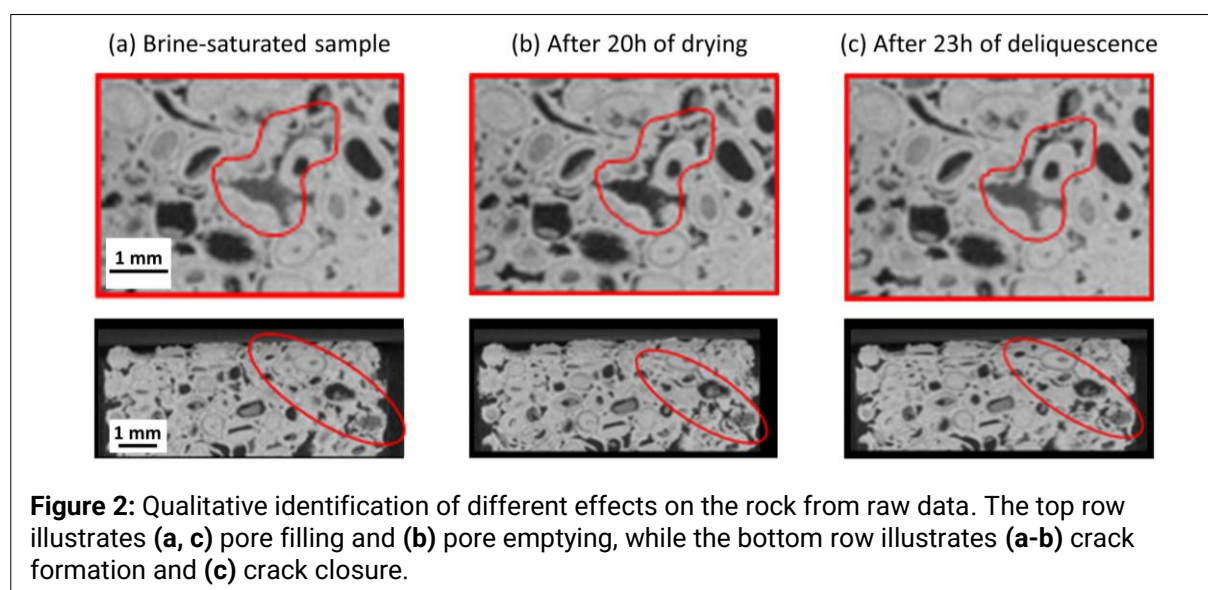
^c SHT75: <https://sensirion.com/products/catalog/SHT75>

The cell was positioned inside the Environmental Micro-CT (EMCT) X-ray scanner^d (10) and time-lapse scanning was performed during drying-induced crystallization and deliquescence. Drying was induced at 5% RH and 26°C with scans taken every 30 minutes over a period of 20 hours. During the next 24 hours, the sample was left to dry in the cell under the same conditions. Scans taken during this period, as well as the advanced image analysis presented in this paper, revealed no further changes in the sample, indicating that it reached a near-dry-state after the first 20 hours. Subsequently, deliquescence was triggered by increasing RH to 93%, with scans acquired every hour for 23 hours.

Each scan lasted 10 minutes, with 1501 projections acquired at 80 kV tube voltage. Reconstructions were performed with the Aquila reconstruction software^e, yielding datasets of 1200 slices each, with a voxel size of 9.4 μm . A total of 62 data series are reported in this manuscript, covering the first 20 hours of drying reaching the near-dry state, and subsequently the 23 hours of deliquescence. To minimize cone-beam artifacts, the sample was positioned so that 80% of its height was covered in the field of view (that is, the upper 7 mm of the sample), while the upper third of the detector viewed the air.

2.3. Image analysis

The image analysis workflow was defined after careful inspection of the raw data (i.e. the data obtained after reconstruction) but prior to further data processing. We first considered the vertical mid-plane of the rock sample and its evolution through time. We specifically focused on the events taking place within the different porosity classes (illustrated in Fig. 2, top row). Visual examination of reconstructed images allowed for the identification of certain macropores, mainly intergranular, filled with saline solution in the initial saturated state of the sample (Fig. 2a, top row) that emptied during the drying process (Fig. 2b, top row). At the end of the deliquescence, these pores refilled, either completely or partially (Fig. 2c, top row). Visual observations of the upper part of the sample (Fig. 2, bottom row) also revealed that cracks appeared in the hydrophobized upper part of the sample that thus suffered damage during the drying phase (compare Fig. 2a and b in the bottom row). Examination of other images revealed that these cracks were small in size and had a non-uniform distribution. During deliquescence, some of these cracks tended to close, either partially or completely (Fig. 2c, bottom row).



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^e Aquila reconstruction software, Tescan XRE. The software is supplied as part of Tescan XRE X-ray CT systems and is not available as a standalone product. The prototype system used in this study was a predecessor of the commercial DynaTOM scanner: <https://tescan.com/product-portfolio/micro-ct/dynatom>

No apparent evidence of calcite dissolution or precipitation was detected over the course of the experiment. This was corroborated through simulations using the PHREEQC geochemical simulation software^f (see Supplementary Material, [available online](#)) confirming that there was no calcite dissolution or precipitation under the applied experimental conditions.

The gray level distribution (not shown) of the capillary-saturated rock sample revealed two dominant intensity ranges: one corresponding to air (darker gray levels) and another to the rock matrix (brighter gray levels). Between these two, an overlapping zone was observed, which could include either the saline solution or represent portions of the matrix with a different amount of (unresolved) porosity, or both. This ambiguity results from partial volume effects, where multiple phases coexist within a single voxel, making it difficult to accurately discriminate between individual phases based solely on gray level intensity. To characterize the transport, crystallization, and deformation processes down to the pore scale, an advanced image analysis workflow is thus required, combining classical segmentation techniques with digital volume correlation and deep learning segmentation, as detailed below.

2.3.1. Classical segmentation

A Python-based image processing workflow was developed using OpenCV^g, Matplotlib^h, and TiffFileⁱ to handle large tomography datasets efficiently and consistently. The process began with median filtering of each transverse slice to improve the signal-to-noise ratio, followed by manual thresholding to separate resolved pores from the other phases (i.e. matrix including unresolved porosity and aluminum). External contours of the sample were then detected using built-in OpenCV functions (`cv2.findContours` and `cv2.drawContours`), allowing isolation of the rock region from the surrounding air and aluminum film. For each slice and time step, the number of pixels belonging to the air-phase (that is, the resolved pores) was computed as well as the total number of pixels within the rock contour. The latter was proportional to the cross-sectional area of the rock sample for a given slice, the proportionality factor being the square of the voxel size (9.4 μm). The evolution of the cross-sectional area through time provided insight into the global sample deformation. For each time step, the total number of pixels belonging to the air-phase, summed over all slices, gave the corresponding (resolved) air volume, and the total number of pixels within all contours gave the rock's volume. The ratio between the two allowed the evolution of the volumetric air content through time to be traced.

2.3.2. Digital Volume Correlation

To account for the internal deformation of the sample, we applied digital volume correlation (DVC) using the SPAM toolkit (36). Digital volume correlation identifies the transformation between two 3D images of the same sample: a reference image and a later image showing deformation. This technique was applied to characterize the deformation during both drying and deliquescence processes. For drying, the first image (sample saturated with saline solution) served as the reference, and subsequent scans acquired every 30 minutes were compared. For deliquescence, the reference was the first scan after 1 hour at high humidity, with comparisons made every hour.

The DVC procedure follows several key steps:

1. **Multiscale registration (spam-reg):** A non-rigid registration aligns the two volumes and provides a global estimate of displacements, including translation, rotation, stretch, and shear.
2. **Local Digital Image Correlation (LDIC) (spam-LDIC):** This step refines the deformation field by performing local, non-rigid correlations on a structured grid of sub-volumes. The

^f <https://www.usgs.gov/software/phreeqc-version-3>

^g <https://opencv.org/>

^h <https://matplotlib.org/>

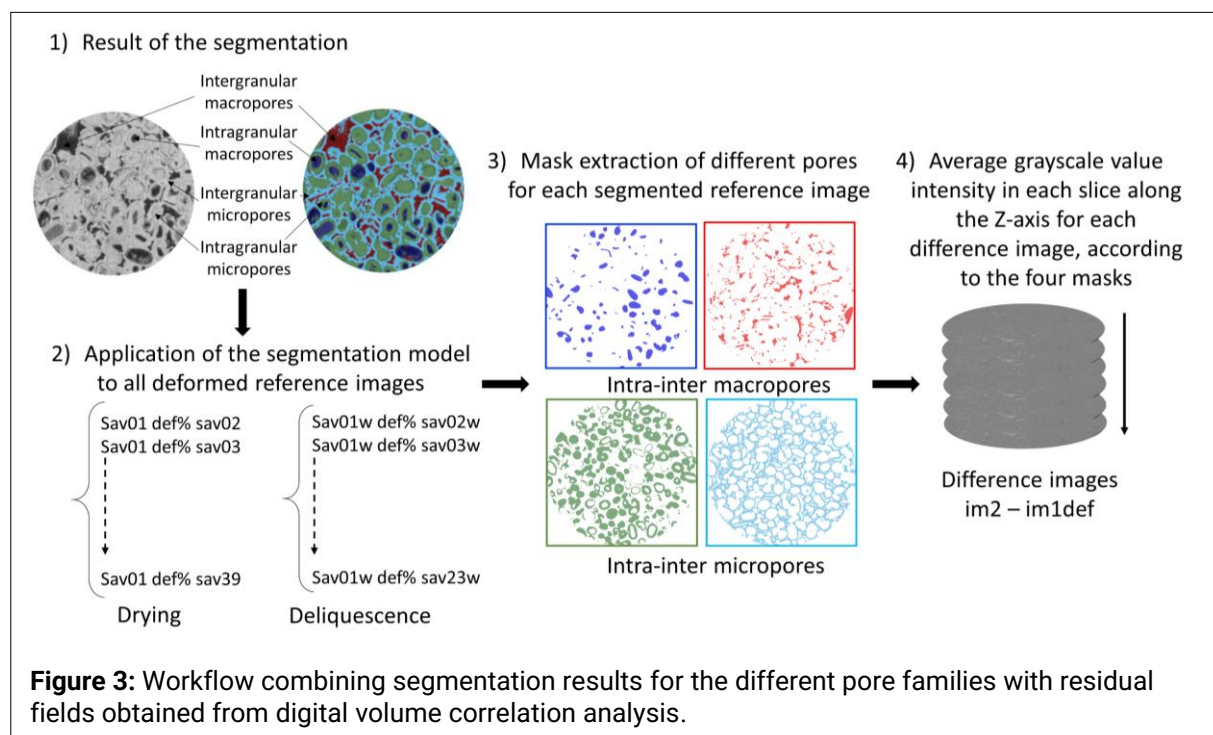
ⁱ <https://pypi.org/project/tiffFile/>

parameters node spacing ($ns = 35$), half-window size ($hws = 15$), and gray level threshold ($glt = 12000$), were found by trial and error and represent a compromise between the level of detail of the deformation field and the convergence behavior of the algorithm. Additional parameters such as the number of iterations and the delta phi norm (convergence threshold) were chosen to optimize accuracy. All the values mentioned were kept constant across all time steps to ensure comparability.

3. **Residual field computation:** The calculated deformation is applied to the reference image (yielding $im1def$), and its subtraction from the target image ($im2$) yields a residual field ($im2 - im1def$). This highlights localized changes (e.g., salt formation or dissolution) not visible in raw images or simple subtractions. It enables quantification of salt, monitoring of drying/deliquescence kinetics, and visualization of transport in unresolved pores.
4. **Strain field computation:** Finally, the regular-strain script calculates the strain field from the displacement field obtained by LDIC (in step 2). Among the available methods, the Q8 finite element method provided optimal results for our dataset.

2.3.3. Pore scale analysis with deep learning

As described in Section 1, Savonnières limestone contains four types of porosity: two macropore types (resolved in the scans) and two micropore types (unresolved). Intergranular macropores are located between calcite cements, while intragranular macropores are found inside partially or fully hollow ooids. The macropores have characteristic sizes on the order of $100\ \mu\text{m}$ and are therefore well resolved in the tomography scans acquired with a voxel size of $9.4\ \mu\text{m}$. The micropores are either intragranular (within ooid cores), having a size below $1\ \mu\text{m}$, or intergranular (between cement crystals), having a size on the order of $10\ \mu\text{m}$. They appear as part of the rock's matrix due to their sub-resolution size. Traditional segmentation methods based on gray level contrast were ineffective to segment and distinguish the pores and the regions with sub-resolution porosity belonging to the four different pore families. Therefore, we trained a deep learning-based segmentation model (U-net) (32) to classify them. This model is available on the Dragonfly software platform (3). From a 3D image of the Savonnières limestone (1076 slices), 10 slices were selected and manually labeled to identify the four pore families: intragranular macropores (dark blue), intergranular macropores (red), regions with intragranular micropores (green), and regions with intergranular micropores (sky blue). Data augmentation techniques



(flipping, scaling, distortion, rotation) were then applied, generating 1000 additional annotated slices, resulting in a training dataset of 1010 images. The U-Net model was trained on 80% of this dataset using the *AdaDelta* optimization algorithm (39) and Dice Loss function, across 100 epochs. Model performance was evaluated on the remaining 20% using the Dice coefficient and loss values. Validation showed a stable loss near zero and a Dice coefficient approaching 1, confirming high segmentation accuracy.

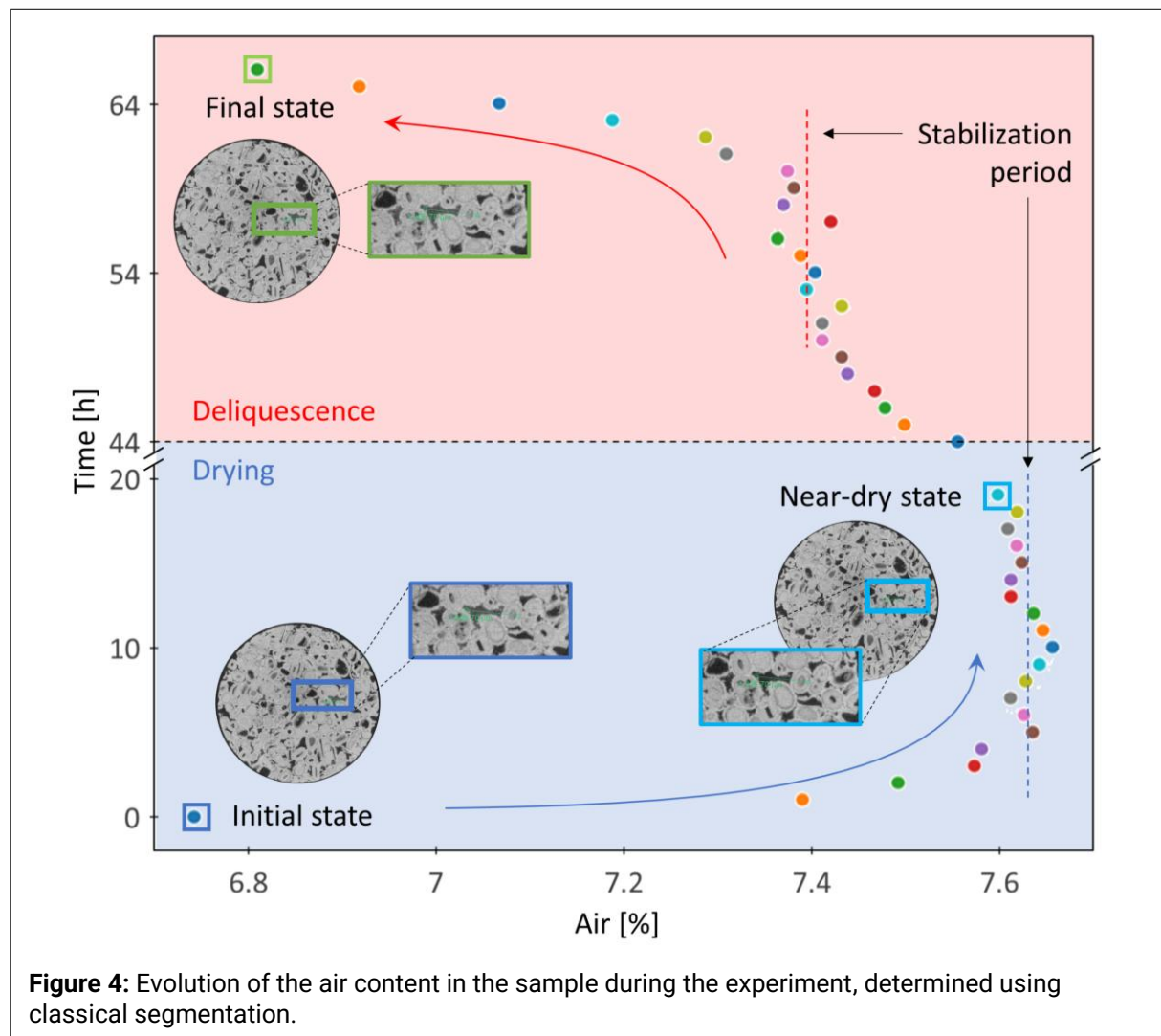
Once trained, the model was applied to track the evolution of each pore family during drying and deliquescence following the workflow depicted in [Figure 3](#). To do so, deformation fields were computed using the DVC workflow between each scan and a reference image (the initial scan of the saturated sample for drying, or the first scan after 1 hour at high humidity for deliquescence). The reference image was deformed accordingly and segmented with the trained U-Net. For example, the reference scan 'sav01' was deformed relative to scan 'sav02' (after 30 minutes of drying) to produce 'sav01deformed%sav02', which was then segmented. This process was repeated across all time steps for both drying and deliquescence. From each segmented, deformed reference image, masks corresponding to each pore family were extracted and applied to the respective difference images (residual field). For instance, the masks from 'sav01deformed%sav02' were applied to 'sav02 - sav01deformed%sav02'. Finally, the average gray level for each pore family was calculated along each slice over the full height of the sample in 3D. This approach ensures accurate alignment between pore family masks and difference images across all time steps, enabling a consistent temporal analysis of salt dynamics in each pore family.

3. RESULTS AND DISCUSSION

Both the classical segmentation and the deep-learning-based segmentation were applied to the same tomographic dataset, as described in [Section 2.2](#), but with complementary objectives. Classical segmentation provides a macroscopic description by separating resolved macroporosity from the rock matrix, allowing identification of global trends during drying and deliquescence. Advanced image analysis combining DVC and deep-learning-based segmentation was subsequently applied to the same images to access finer-scale information, including the identification of the regions with sub-resolution pore families and the transport mechanisms occurring therein.

3.1. Variation of air content

[Figure 4](#) shows the evolution of the air content in the sample during drying and deliquescence obtained by classical segmentation. Initially, the air content corresponds to capillary-trapped air (~6.8%). The air content gradually increases over the first six hours of drying, then stabilizes at approximately 7.6%, forming a plateau (highlighted by the blue dashed line in [Fig. 4](#)). This trend reflects the evaporation of saline solution. During the first three hours of deliquescence, a slight decrease in air content occurs, likely due to moisture entering small fissures at the sample's top. This is followed by a stable period (~9 hours, red dashed line in [Fig. 4](#)), and then a rapid decrease in air content as macropores refill with brine, reaching a value close to the initial state by the end of the experiment. The stability observed at the near-dry state and after 3 hours of deliquescence does not necessarily mean that transport ceased but might indicate that unresolved micropores are slowly filling or emptying, a hypothesis further investigated in [Section 3.5](#).



3.2. Radial deformation

The second result from the classical segmentation concerns the evolution of the cross-sectional area over time along the sample height, as defined in Section 2.3.1, and shown in Figure 5a for three points in time: initial saturated state (blue curve), near-dry state (orange curve), and end of deliquescence (green curve). Only three curves are shown to maintain figure readability. Differences are observed in the top 2-3 mm of the sample, where expansion during drying (compare blue and orange) is linked to crack formation (confirmed by raw images, see Fig. 2), followed by a contraction during deliquescence (compare orange and green), indicating partial crack closure.

To quantify this process for all scans acquired during drying and deliquescence, we first calculated the deformation relative to the initial state according to the following equation:

$$deformation = \frac{S_{current} - S_{initial}}{S_{initial}} \quad (1)$$

where $S_{initial}$ and $S_{current}$ represent the cross-sectional area delimited by the contour at the initial state of the sample and at the “current” time point of interest, respectively. Then, the average deformation over the top 277 slices (2.6 mm, i.e. the hydrophobic part) was computed (Fig. 5b) at each point in time. Each data point therefore represents the mean deformation at each time step during drying and deliquescence, a more detailed discussion is provided in Sections 3.4 and 3.5. Figure 5b shows:

- A rapid increase during drying, reaching 1.4% deformation at the end

- A slower decrease during deliquescence, ending at 0.9% residual deformation

The bottom of the sample remains largely unchanged (not shown), with only minor variations (slight shrinkage during drying, slight expansion during deliquescence), but these are too small for detailed interpretation at this scale. Therefore, a more local analysis is required, as described in the next sections.

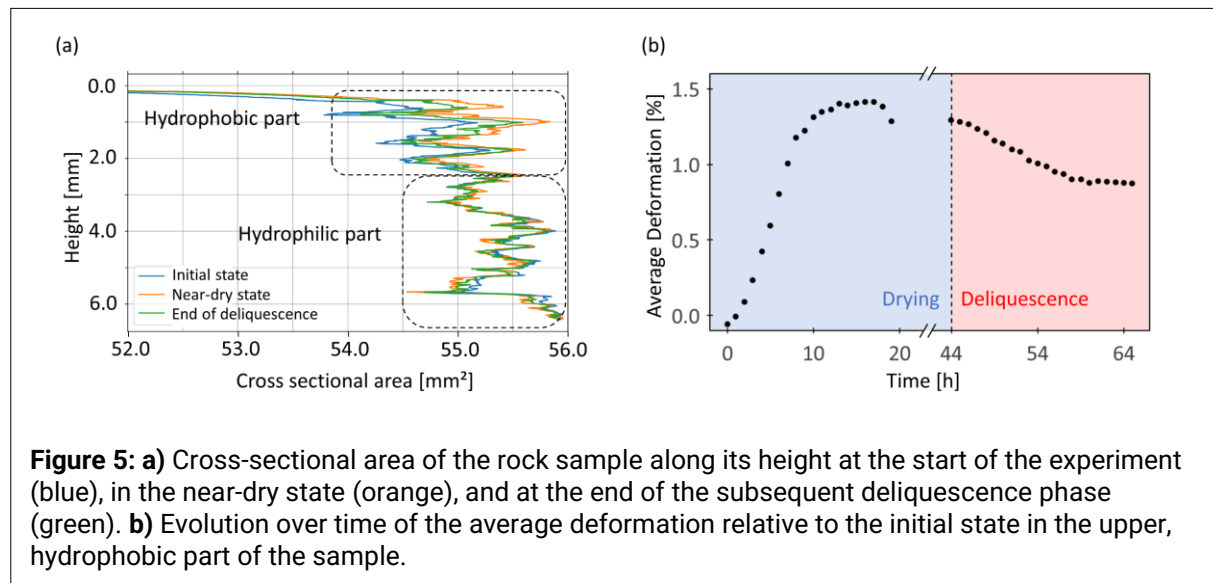


Figure 5: **a)** Cross-sectional area of the rock sample along its height at the start of the experiment (blue), in the near-dry state (orange), and at the end of the subsequent deliquescence phase (green). **b)** Evolution over time of the average deformation relative to the initial state in the upper, hydrophobic part of the sample.

3.3. Salt distribution during drying

Based on the deformation-adjusted difference images (residual field, see Section 2.3.2), it was possible to distinguish and quantify precipitated NaCl salt despite its similar contrast with the rock matrix. The curve representing the salt quantity as a function of the sample's height (Fig. 6a) reveals a clear accumulation of salt at the top of the sample. This region corresponds to the part that received hydrophobic treatment. The salt quantity increases progressively over the drying period, specifically in this hydrophobized region. To monitor the rate of salt formation, the total salt volume throughout the sample was calculated at each time step (Fig. 6b) by integrating the salt distribution profiles as depicted in Figure 6a over the height. At the beginning of the drying process, up to 300 minutes, the salt precipitation rate is high, indicating that crystallization starts rapidly as soon as water begins to evaporate (part 1 of the curve in Fig. 6b). Between 300 and 600 minutes, the slope of the curve decreases, suggesting a decrease in crystallization rate, either due to the reduced availability of saline solution or because the salt concentration falls below the solubility limit of NaCl (part 2 of the curve in Fig. 6b). In the final phase (part 3 of the curve in Fig. 6b), the curve reaches a plateau, indicating that most of the water has evaporated and that the crystallization process is approaching completion. The hydrophobic treatment penetrated the sample in a non-uniform manner, resulting in a gradual decrease in hydrophobicity with depth rather than a sharp hydrophobic-hydrophilic interface (7, 8). As evaporation proceeds, salt is transported toward the drying front, which is initially located within the hydrophobized region. This explains the preferential accumulation of salt crystals in that region (8).

3.4. Mechanical behavior in the hydrophobically treated part of the sample

In the hydrophobized part of the sample, complex physical interactions are observed, including mechanical behavior characterized by crack formation and closure, associated with salt accumulation. These observations are supported by the calculation of the volumetric strain using the DVC workflow (Section 2.3.2). The distribution of volumetric strain across the sample height during drying and deliquescence exhibits distinct behavior during drying and deliquescence, as discussed in the remainder of this section.

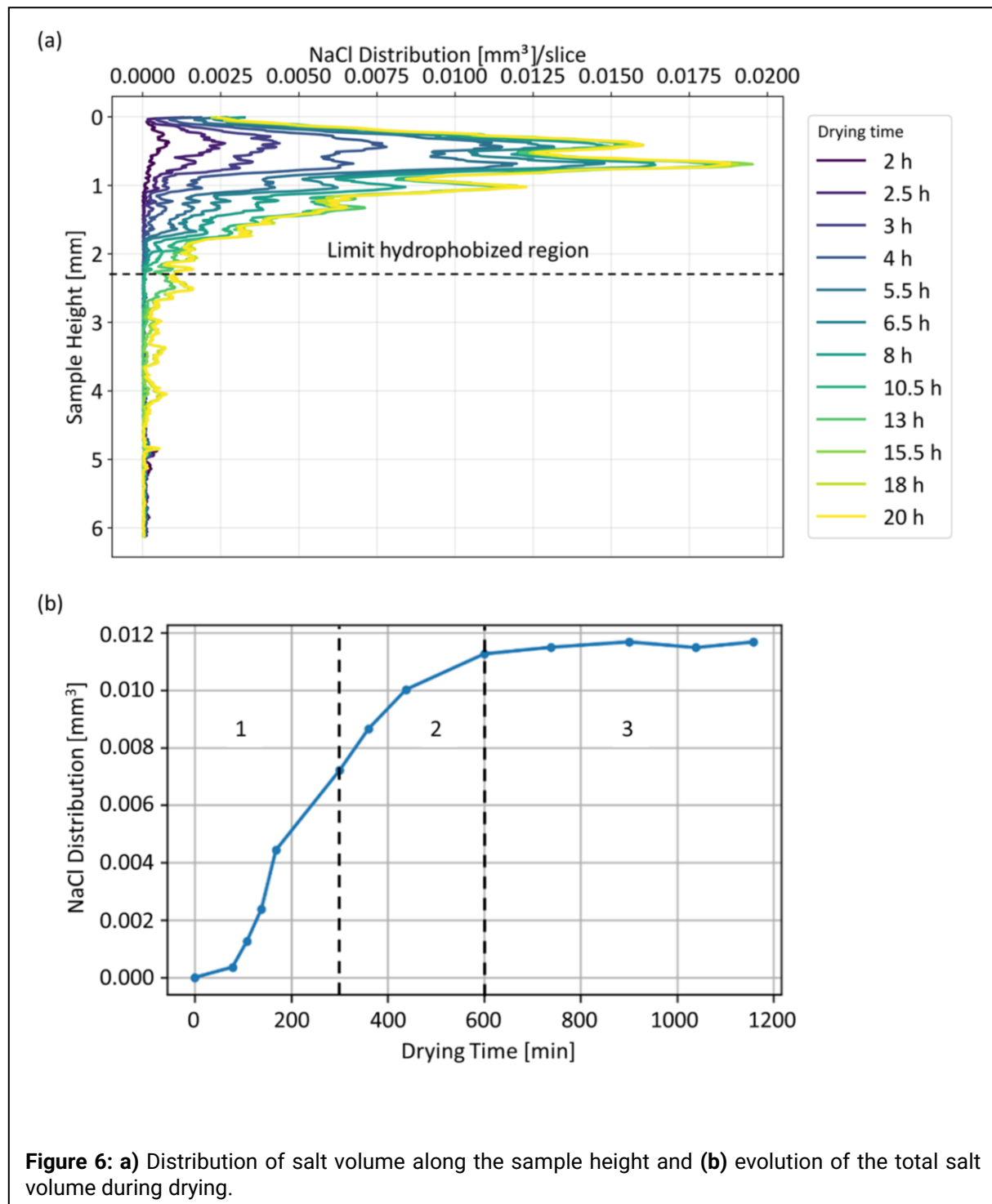
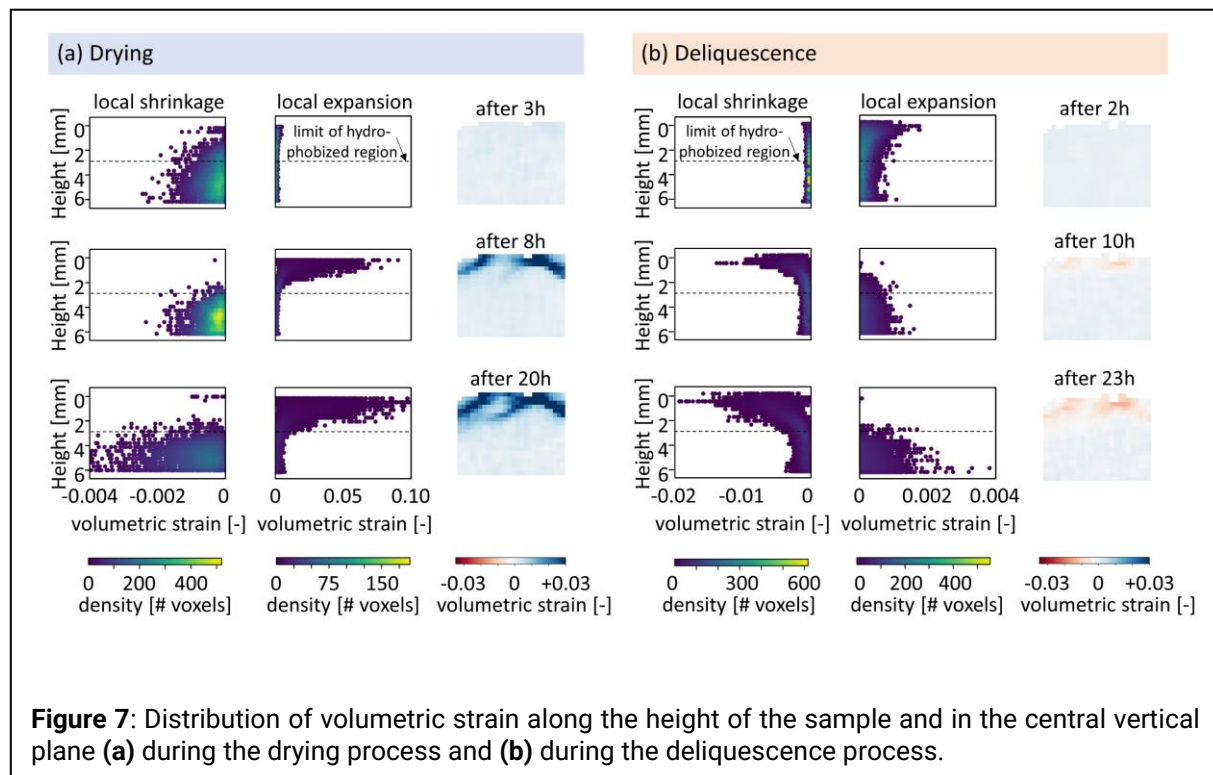


Figure 6: **(a)** Distribution of salt volume along the sample height and **(b)** evolution of the total salt volume during drying.

During drying: At the onset of drying, the distribution of volumetric strain across the sample height shows small negative volumetric strains, caused by a slight shrinkage due to evaporation (Fig. 7a, left side). Over time, volumetric strain becomes positive in the top part of the sample, primarily due to salt crystallization in this region, leading to local material expansion. As the expansion is not homogeneous, this results in heterogeneous internal stresses. When the stress state locally exceeds the rock's mechanical resistance, cracks can form. Digital volume correlation identifies these cracks as regions of high volumetric deformation, as seen in the volumetric strain field representations of the central vertical plane in Figure 7a (right side), and comparison with the raw, unprocessed, X-ray data confirmed this interpretation.

During deliquescence: The first stage of the deliquescence process is characterized by positive volumetric strains (Fig. 7b), left side), indicating swelling with respect to the reference state due



to moisture absorption. This is attributed to the hygroscopic behavior of NaCl, which absorbs moisture from around 75% relative humidity upward. As pores expand, local swelling occurs. Subsequently, negative volumetric strain indicates salt dissolution, reducing the volume of salt crystals and causing shrinkage. Volumetric strain fields of the central vertical plane (Fig. 7b right) show that the cracks which formed during drying start to close as negative volumetric strain increases. This closure may result from several phenomena linked to high humidity: swelling of the matrix leading to crack closure (especially for thin cracks), and dissolution of NaCl, reducing crystal volume that allows to reduce crack openings.

Using pore family segmentation masks on the difference images, further insights are gained. Figure 8a illustrates the evolution of the average gray level intensity for different pore families throughout the drying process. Each curve on this graph represents a pore family: the red curve intergranular macropores, the dark blue curve intragranular macropores, the sky-blue curve intergranular micropores, and the green curve intragranular micropores. The variation in average gray level with time (Fig. 8a) shows that intragranular macropores exhibit the strongest increase in intensity, indicating significant salt accumulation in these pores compared to intergranular macropores. A moderate increase is observed in both intragranular and intergranular micropores, consistent with their relatively smaller size. The greater salt accumulation in intragranular macropores may result from non-uniform fluid transport in the hydrophobized region. During imbibition, the intergranular macropores, being more exposed to the exterior, might have been preferentially affected by the hydrophobic treatment, leaving the intragranular macropores more accessible to the saline solution. Regarding the increase in salt within intragranular micropores, which include the micropores in and around the ooids (Fig. 8b), the difference images reveal salt deposition around the ooids (Fig. 8a, c). A subsequent decrease in gray level intensity in both types of micropores corresponds to crack formation (Fig. 8a, c). More fissures are observed in intergranular micropores than in intragranular ones. This suggests that fractures initiate in areas of significant salt accumulation around ooids, where mechanical fragility is highest. Consequently, ooids are subjected to two sources of salt-induced pressure: one from macropore crystallization

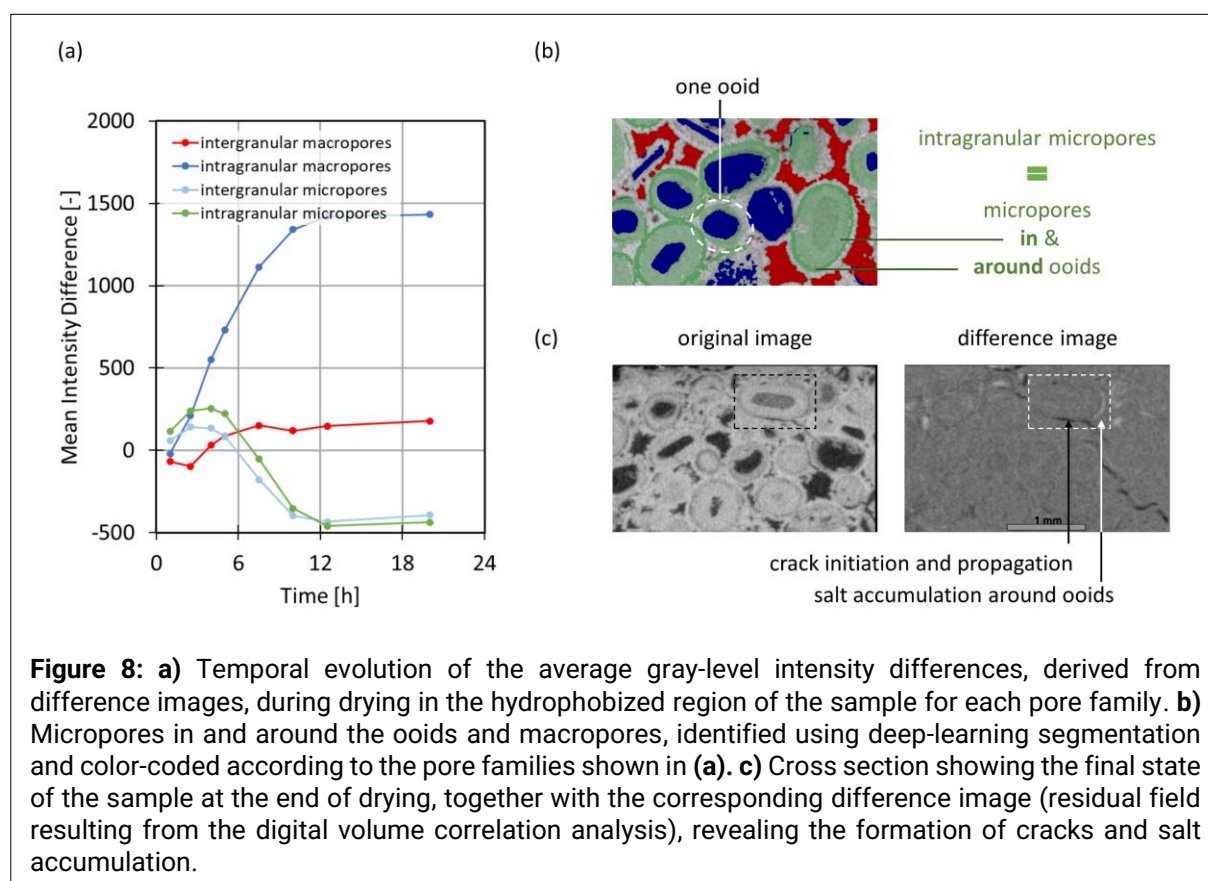


Figure 8: **a)** Temporal evolution of the average gray-level intensity differences, derived from difference images, during drying in the hydrophobized region of the sample for each pore family. **b)** Micropores in and around the ooids and macropores, identified using deep-learning segmentation and color-coded according to the pore families shown in **(a)**. **c)** Cross section showing the final state of the sample at the end of drying, together with the corresponding difference image (residual field resulting from the digital volume correlation analysis), revealing the formation of cracks and salt accumulation.

and another from crystals in the surrounding microporosity. Cracks thus begin in these pores and propagate toward the surrounding calcite (segmented as intergranular micropores).

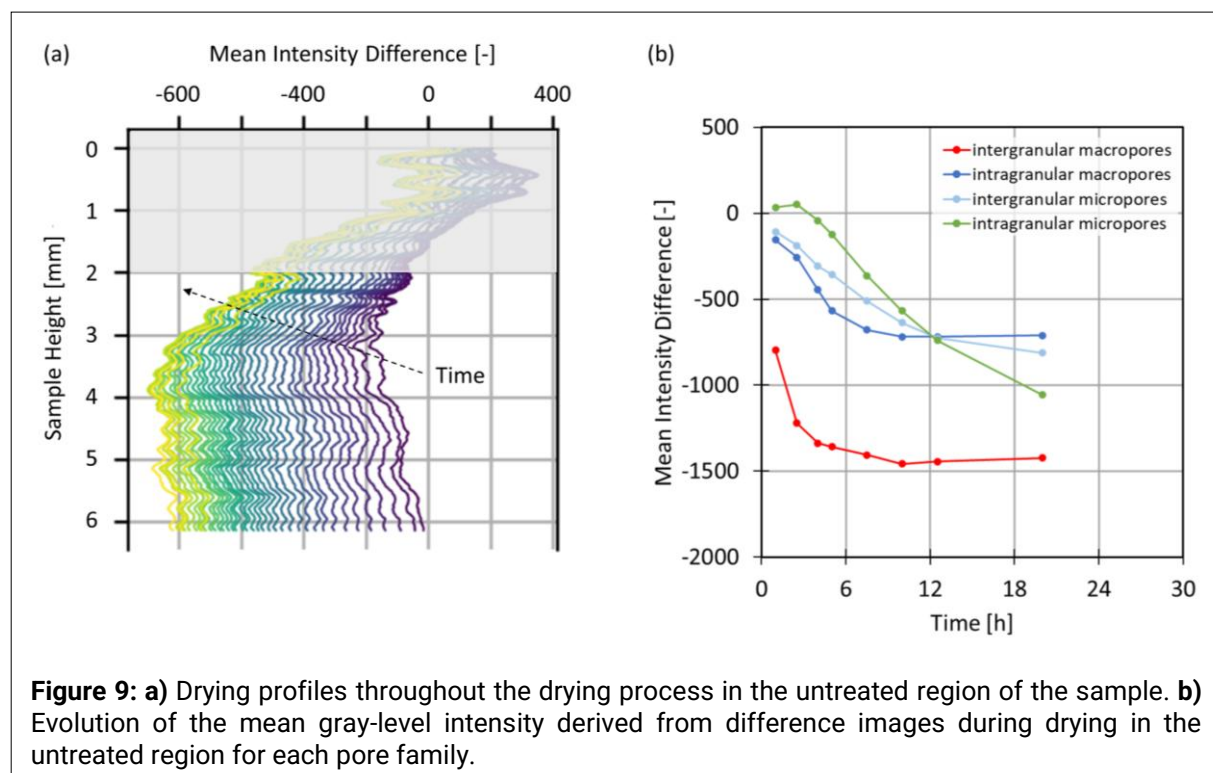
3.5. Chronology of drying and deliquescence kinetics in each pore family in the untreated part of the sample

The untreated part of the sample exhibits no notable damage or significant salt accumulation, which enables a more focused analysis of the local transport kinetics in the Savonnières rock.

During drying: From the 3D difference images obtained via the DVC workflow, the mean gray level intensities were calculated along the sample height (Fig. 9a). During drying, the curves trend uniformly towards negative values, indicating a shift toward darker gray levels. This trend toward darker shades is interpreted as a loss of saline solution and thus as an indicator of the drying process of the sample, showing progressive pore emptying. The drying rate is initially constant but slows down towards the end. Figure 7a shows a combination of small negative and positive values of volumetric strain. Persistent negative values indicate shrinkage due to ongoing drying. Positive deformation values, appearing towards the end, reflect minor salt crystallization in small pores (Fig. 6). These small deformations are therefore caused by the crystallization pressure generated by these salt crystals in the microporosity.

The combination of segmentation using the U-Net model and difference image analysis from the DVC workflow provides a detailed picture of the solution distribution in each pore family. Figure 9b illustrates that the initial value of each curve corresponds to the average gray level intensity observed when the pores are saturated with saline solution. It is important to note that the

intergranular macropores have very different initial intensity values compared to the other pore families. This difference can be attributed to the fact that large intergranular pores begin to dry earlier and faster. The first data point in **Figure 9b** corresponds to the sample after one hour of drying. It is therefore likely that the large intergranular pores had already dried significantly during this first hour, consistent with the lower initial intensity values in the difference images of the subsequent analysis. During drying, a general decrease in average intensity is observed for all pore families. The initial slope of each curve provides information about the drying rate of the pores: a steeper slope for instance indicates faster drying. During the first hours of drying, the intergranular macropores are the first to dry, followed by the intragranular macropores, intergranular micropores, and finally intragranular micropores. This chronology corresponds to the pore size hierarchy, where the intra- and intergranular macropores are followed by intergranular micropores and, finally, the smallest pores, the intragranular micropores (30). Regarding the macropores, a rapid decrease in intensity is observed at the beginning, indicating fast drying, followed by a gradual stabilization of the drying process. For micropores, particularly the intragranular ones, the initial trend shows a slight decrease that becomes more pronounced over time, indicating an acceleration of drying. This acceleration in micropores' drying coincides with the stabilization of drying in macropores, meaning that the macropores are connected to the microporosity. In both raw and difference images, intergranular macropores can often be seen filled with saline solution, whereas intragranular macropores rarely appear in this state (**Fig. 2**). Thus, the difference between the curves for these two types of macroporosity can be attributed to the greater number of intergranular macropores filled with saline solution compared to the intragranular ones.



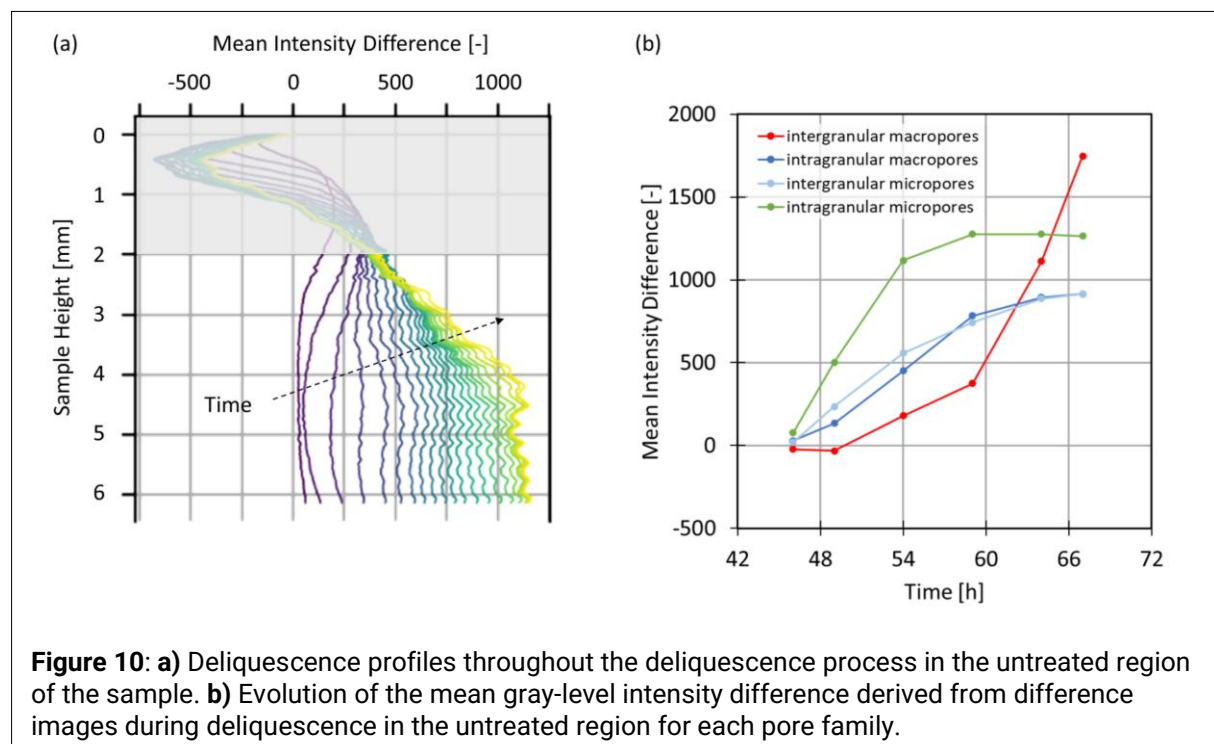
During deliquescence: the deliquescence phase unfolds in three stages characterized by a slow onset, a second acceleration stage, and ultimate slowdown. Initially, precipitated salt deliquesces; then, dry pores facilitate absorption (**Fig. 10a**). As saturation increases, the absorption rate declines.

Figure 7b shows sustained positive values of volumetric strain, confirming continuous swelling due to saline solution absorption. This aligns with the hydrophilic nature of this part of the rock. Simultaneously, small negative volumetric strain values suggest dissolution of residual salt

crystals formed during drying. Their dissolution leads to minor pore shrinkage, a subtle effect that is however consistent with the expected reduction in crystal volume.

The pore-filling sequence during deliquescence (Fig. 10b) is the inverse of the sequence observed during drying: intragranular micropores fill first, followed by intergranular micropores, intragranular macropores, and lastly intergranular macropores. When micropore gray level intensities begin stabilizing, macropores start filling, confirming a link between macro- and micropore behavior. Notably, intergranular macropores appear to absorb more saline solution than other pore families, corroborating their state observed at the beginning of drying.

The observation presented in Section 3 regarding the stabilization of the air content within the sample (Fig. 4) at the end of drying and after 3 hours of deliquescence, can be explained by the drying and refilling activity of the micropores. This phenomenon, which takes place in pores with sizes below the resolution of the X-ray images, could be detected in our work through advanced image-processing techniques while this is beyond the reach of more traditional approaches.



4. CONCLUSION

This study offers new insights into the coupled transport-mechanical behavior of a porous limestone subjected to drying and deliquescence, with a particular focus on salt crystallization and dissolution. By combining X-ray tomography, difference imaging with local deformation correction by DVC, and deep-learning-based pore segmentation, we were able to visualize and quantify, in 3D and with sub-voxel precision, the evolution of saline solution distribution and salt precipitation within different pore families. A key originality of this work lies in the ability to distinguish between gray-level variations caused by deformation and those induced by transport processes, even in damaged areas of the sample. This enabled the precise localization of salt crystallization, particularly in the hydrophobized part of the rock, where traditional segmentation methods would fail due to the similar contrast between the different phases in the sample. Our results show that salt preferentially accumulates in intragranular macropores and micropores, especially around ooids, which represent the initiation sites of cracks that we could follow in real time, from their onset to their propagation through the rock matrix (intergranular microporosity). These results confirm the observations of Fronteau (14), who, based on the study of vacuolarian

oids with microcracks, conceptually reconstructed the evolutionary process of microcracking and identified the rock phases affected by this evolution. Fronteau (14) observed that the cracks cut perpendicularly through the cortex of the ooids and connected the intra- and intergranular macropores and cements, showing that the intergranular micropores were fractured by the microcracks passing through the ooids. In the untreated part of the sample, we identified a clear chronology of drying and deliquescence at the pore scale, revealing an inversion between macropore-to-micropore emptying during drying and micropore-to-macropore filling during deliquescence. This sequence, obtained through image-based analysis, confirms and extends previous experimental findings, providing a refined view of pore-scale fluid dynamics in multimodal porous materials. Finally, the identification of crack formation and partial closure as a response to crystallization and dissolution stresses brings valuable understanding of the mechanical implications of salt weathering. Moreover, by combining different image-processing techniques, it becomes possible to extract pore-scale information from non-destructive imaging alone. This integrative approach provides a powerful framework for future investigations of salt damage in cultural heritage materials, porous building stones, and natural reservoir rocks.

STATEMENTS AND DECLARATIONS

Supplementary Material

The PHREEQC simulation with its input and the main output results are provided in the Supplementary Material that can be accessed [online](#).

Data, Code & Protocol Availability

The datasets generated and analyzed during the current study are available from the corresponding authors upon request.

The DVC analysis uses an open-source software, SPAM, its source code is publicly available on <https://gitlab.com/spam-project/spam>.

All relevant methodological details and protocols are described within the article.

Author Contributions

S. Ben Elhadj Hamida: Conceptualization, Methodology, Software, Visualization, Writing-original draft, Writing-review & editing; **M. A. Boone:** Investigation, Writing-review & editing; **V. Cnudde:** Resources, Writing-review & editing; **P. Moonen:** Conceptualization, Supervision, Writing-review & editing; **H. Derluyn:** Conceptualization, Investigation, Funding acquisition, Project administration, Supervision, Writing-review & editing

Conflicts of Interest

The authors declare no conflicts of interest.

Funding Received

Syrine Ben Elhadj Hamida and Hannelore Derluyn acknowledge support from the European Research Council (ERC) under the European Union's Horizon 2020 research and innovation programme (grant agreement No 850853). Hannelore Derluyn acknowledges furthermore the support from the Research Foundation – Flanders (FWO) for her postdoctoral fellowship 1276814N during which the X-ray micro-CT data were acquired, and thanks Erika Tudisco for exchanges about the digital volume correlation method. Veerle Cnudde acknowledges the financial support of FWO Grant G065224N and BugControl (project number VI.C.202.074) of the NWO Talent programme, financed by the Dutch Research Council (NWO).

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