4D porosity evolution during pressure-solution of NaCl in the presence of phyllosilicates

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Abstract: Pressure-solution creep is one of the most common crustal deformation mechanisms, inducing changes in the porosity and permeability of rocks. For a variety of rock types undergoing pressure solution, it has been shown that the presence of phyllosilicates may significantly enhance the rate of the pressure-solution process. In this experimental investigation, we present 4-dimensional (three dimensions + time) X-ray microtomographic data that contrast deformation by pressure-solution of a pure NaCl aggregate with that of a mixture of NaCl and biotite. The results show that for mixed samples (NaCl+biotite), phyllosilicates induce a marked reduction in porosity and pore connectivity and contribute to an increase in the local strain rates by an order of magnitude over pure NaCl samples. At the same time, phyllosilicates do not induce strain localization in the sample. We discuss various possible explanations for these observations including a possible positive feedback between the porosity distribution and pressure solution. Our study yields novel insights into the local effects of phyllosilicates during pressure-solution creep and provides full 4-dimensional imaging and characterization of the coupled evolution of porosity and pore connectivity over previously unprecedented experimental time scales.

Rebuttal/list of changes

The discussion can be stronger in determining the potential reasons for the decreased porosity in the biotite zone, and what the relative importance is of these different reasons. The dismissal and acceptance of hypotheses can be better correlated to the images obtained. I do not see how the force chains come in, or where they are shown on the image. Nor am I certain I can see the locations of increased dissolution and preferred location precipitation (last hypothesis), though I do think this hypothesis sounds the most plausible. Perhaps also it can be a combination of the different hypothesis mentioned, where some play a role during fast early compaction, and others during the slower aftermath. Additionally, the comparison with previous literature could also be stronger, i.e. Renard et al indicate a mechanical role of clay particles, which is not mentioned by the authors here. Spiers et al compacted similar size NaCl aggregates and obtained different strain rates at lower stresses. What are the differences between their and these results? In the line-by-line comments on the discussion there are some more details on which lines can use some clarification.

We thank the reviewer for these additional comments and her opinion on our manuscript. In the line-by-line comments, we respond to the reviewer's comments. Combined, they form our response to the points raised above.

Line-by-line comments

1. Highlight 1 & 2 are very similar

We removed the second highlight to improve clarity.

2. Highlight 1: isn't it local porosity, not bulk? Porosity outside biotite zones is less affected. The term bulk porosity implies the complete sample to me.

We agree with the reviewer's comment and changed the text accordingly.

3. Line 20: To avoid confusion (since you didn't test all phyllosilicates) I would suggest rephrasing to 'the results show that for mixed samples (NaCl+biotite), the phyllosilicates induce'

We revised the text according to the reviewer' suggestion.

4. Line 44: somewhat arbitrary selection of references; already from your own reference list you could create a more diverse list of geological materials (from your list you could include: Bos and Spiers - phyllosilicates, Pluymakers and Spiers - anhydrite). Moreover, since there definitely exist other geomaterials than listed here, I'd suggest preceding these references with e.g.

The reference list was modified to include the suggestion of the reviewer.

5. Line 46: change to 'these alterations, may, in turn' - a change in transport properties is not a given - that would depend on the amount of alteration and potentially also direction of transport

We agree and we change the text accordingly.

6. Line 79-80: why not both? i.e. leave the NaCl as geomaterials, since they are geomaterials in salt deposits, and if you want you can state here that they are often used as analogues for

other rock combinations that deform slower, with the appropriate references? This would make more sense than denying NaCl is a geomaterial

We have been careful with our choice of words. We agree that NaCl is a geomaterial in its own right, however, we do note that natural evaporites undergoing deformation do not occur in the form used in these experiments, i.e. highly porous granular NaCl aggregates. Nevertheless, in that form NaCl does provide a useful experimental analogue for other granular geomaterials.

7. Line 102: 'dimensions of the initial salt stack' is this from the reference scan, i.e. after pre-compaction? Unclear

Yes, the dimension of the initial salt scan refers to the reference scan. We modified the text to improve clarity.

8. Figure S2 caption: is this the NaCl+bio sample presented in the main paper or the one presented in the supplementary material?

The caption refers to the NaCl-biotite sample presented in the main paper. The second NaCl-biotite sample presented in the supplementary material was added later, after a different reviewer requested an additional experiment.

9. Line 113: which load? Similar or higher than experimental load? If similar or lower, than there is a possibility that in the beginning of the experiment there will be significant grain rearrangement, leading to fast initial compaction (c.f. Pluymakers et al., 2014). You should be able to determine if this was significant by comparing the scan made at the reference point versus the one made at t=I. Did you determine the porosity in the reference scan? If so, nice to mention here.

The load to pre-compact the samples was similar to the experimental load. We followed the procedure described in Spiers et al. (1990). The pure NaCl sample exhibited an initial porosity of ~36%, while the sample with biotite, due to the different grain packing and an initial different height, exhibited a porosity of ~33%. We interpret grain rearrangements and fast dissolution rates at small contact areas to be the cause of the initial fast compaction (lines 371-372). We have amended the text accordingly (lines 100-101).

10. Line 120: 3 significant numbers, how precise is this load controlled? Really 6.64±0.01 MPa? Was the sample unloaded to make the scans?

Please see our reply in the previous rebuttal The load was kept constant between 6.4 and 6.6 MPa for the duration of the experiment. The samples needed to be unloaded to allow for scan acquisition.

11. Line 127/Fig 2: Fig 2 indicates 11 datasets

That is true, but the first reference scan (0) is not numbered in the figure, thus the experiment led to 12 datasets. We have clarified this in the figure caption.

12. Line 139: Include here if the initial height of the sample is taken from the reference scan, after pre-compaction. (same as line 102)

The text has been revised to include the reviewer' suggestions.

13. Line 146: figures 1-2-3 are not labelled in order, or did I miss figure 3?

Figure numbering now follows the order of their appearance in the manuscript text.

14. Fig 4/line 102-103: in fig 4 initial porosities are 0.36 and 0.33, and in line 102-103 this is 0.39 and 0.33. Is this a difference between for and after pre-compaction?

The reviewer identified a mistake in our reporting of the values in the text. Line 102 has now been modified to include the correct value.

15. Line 130: you need a list somewhere indicating at which time the scans were taken, or you need to indicate in all your figures at which scan-point they are taken. Now it is pretty difficult to relate Figure 4 with Figure 2 - when is 22.5 hours? 208 (V I assume?) and 2455 (XI I assume?) are a bit easier to guess. This also goes for following figures.

A list has been added to the Supplementary Info to indicate at which time the scans were taken, and to which amount of vertical shortening they correspond (Table S2). This list is being referred to in the main text.

16. Line 182: so strain rate was [d<epsilon>(t)- d<epsilon>(t0)]/t(t)? Or [d<epsilon>(t)-d<epsilon>(t-1)]/[t(t)-t(t-1)] If the latter, shouldn't the strain rate for NaCl between VIII and IX be negative?

The values of strain/pixel (derived from the DVC analyses) were divided by the time (in seconds) between each pair of correlated images using the mathematical operation functions available in ImageJ. This operation yields the integrated strain rates (s^{-1}) for each point in the sample over the given time interval. This analysis returns the local strain rates, and not the bulk strain rates (as reported in Fig. 2b, for which negative values have indeed been calculated between VIII and IX compaction step).

17. Line 211: 'microfracturing' This is scale dependent, any microfractures would need an aperture of at least 2x voxel size in order to be visible, which means apertures of 12-13 micrometer at least. That is quite a bit…

We agree with the reviewer's comment, and we modified the text accordingly. We wanted to highlight that no microfracture indicative of brittle deformation could be resolved.

18. Fig. 5: the figure might be easier to grasp if you would plot strain rate instead of strain in this plot, but up to authors. The text clarifies sufficiently.

As the reviewer is happy with the text clarification, we decided to keep the figure as it is.

19. Line 250-266: 15 lines on a figure in the supplement, whereas Fig. 1 is mentioned once. This seems out of balance, consider swapping Figures, or shorten the description of this figure (i.e. only line 250-253, + line 265 for example?)

We revised the text to improve clarity and balance in the description.

20. Fig 7: why didn't you circle all areas of local compaction? What makes the one in the white circle special? Good improvement of Figs 7 and 8 by the way!

The circles are examples of areas showing highest compaction/strain localisation. Highlighting all areas of compaction with circles would have rendered the figure too clattered and unclear.

21. Line 285-295: any opinion on the local dilation in volumetric strain? Real or artefacts? If real, any idea what they mean?

We speculate that the local dilation areas in the volumetric strain represent local redistribution of grains and changing pore geometries, while the compaction values represent the real compaction and local porosity reduction. We believe that the dilating volumes are indeed real and not artefacts.

22. Line 301: why give first order of magnitude, then 3 x 10¬-8? Looks odd

The text has been improved for clarity.

23. Caption to Fig 9: include what the holes mean.

We modified the caption accordingly.

24. Line 325: the shapes of the curves together with your microstructures forms indeed convincing evidence of PS. Why not include the microstructures in the sentences here?

We agree that the combination of evidence from our compaction curves and from microstructures is convincing evidence of PS. However, in this case, we consider that the existing structure maintains clarity and more effectively guides the reader through our observations and interpretation.

25. Line 331: what was similar in the behaviour, the shape of strain vs time? The absolute rates? With the coarse grains here one would expect interface-controlled PS. In the studies used for comparison here (Renard and Spiers) grains are smaller and stresses were different. Are they nevertheless also expected/proven to be interface-control?

We compared the time-evolution of strain in their data with the one from our data. In these previous studies, the authors observed a very steep initial gradient, as in our data, and similar vertical shortening amounts. In NaCl, kinetics is known to be transport/(diffusion)-controlled (Spiers and Brzesowsky, 1993), while in fine-grained water-saturated quartz compounds the kinetics are known to be interface-controlled (Lehner and Bataille, 1984). We therefore do not feel that the kinetics in the studies presented by Renard or Spiers should be interface-controlled.

26. Line 330-344: a bit unsure what you are trying to say in this paragraph. The first part of the paragraph is on the effects of clay on compaction behaviour, without a conclusion (line 330-340). In Renard et al the increased compaction was explained with the inactive clay propping open the fluid pathways. In your sample this can't be the reason. In the second part (line 340-340) compares to Spiers et al, 1990, which is pure NaCl. Looking at their paper, Fig 5, they show for a grain size comparable to yours (275 ± 25 mu) volumetric strain rates of 8*10-4 to 10-7. Your vertical strain rates for the pure NaCl sample are 10-6 to 10-8 (Fig 2); and the volumetric strain rates from DIC are 10-6 to 10-7 (Fig 9). Spiers et al have a lower effective stress, so theoretically their strain rates should be lower than yours. Can you explain this discrepancy?

Our study explores the time evolution beyond the initial steep compaction rate shown in Renard et al. (2004) and Spiers et al. (1990) and provide a visual assessment of the spatial distribution of local strain rates. However, after ~7 days of deformation, Spiers et al. (1990) reached 20 % vertical shortening (Fig. 3): over that time interval, the integrated bulk strain rates are $\sim 10^{-7}$ s^{-1}, for comparable grain sizes (Fig. 5). In the same time interval, our data show comparable integrated bulk strain rates (Fig. 2b). However, in longer interval times (later in the experiment), our data progressively show lower integrated bulk strain rates. The volumetric strain rates reflect the local distribution of strains within the sample for each time interval between correlated images. At the very beginning, our pure NaCl data shows similar values to Spiers et al., (1990). It is only later on, with more deformation, that lower strain values are obtained. Initial packing and porosity content are likely to affect the overall gradual vertical shortening and have an effect on the bulk strain rates.

The text has been revised to improve clarity.

27. Line 348: after 400 hours you only have 3 datapoints, which seem too have an error of about half the symbol size (comparing VII and VII for example). For me, that would make it difficult to attach strong conclusions to estimate if the trend is linear or constantly decelerating (the latter is what is predicted by PS theory).

We agree with the reviewer, and we modified the text line accordingly.

28. Line 349: indeed a plausible explanation
29. Line 369-382: I am still not convinced about these force chains. In which image would they be visible? Can you indicate them somewhere? I do not see them in Figs 7 or 9. The only clear 'chains' I see are in the NaCl/bio sample in Fig 9, and are formed and explained by the presence of the biotite flakes, which speed up the compaction rates.

We envisage the force chains to be dynamic and to change relatively quickly over time. A careful visual assessment of the maxima of the DVC results in the pure NaCl sample showed how these maxima are transient and how they change positions between pairs of correlated images. To help the reviewer visualising them though, we created a new figure 3, in which an overlay of what we interpret to be *force chains* has been added to the µCT images. This figure has now been included in the Supplementary Material (Fig. S8). However, we do not believe these “outlines” to illustrate definite force chains, as force chains are 3D features. The dynamic force chain model is, at this stage, a speculative one which plausibly explains some of our findings. It remains for further (ongoing) studies to test it.

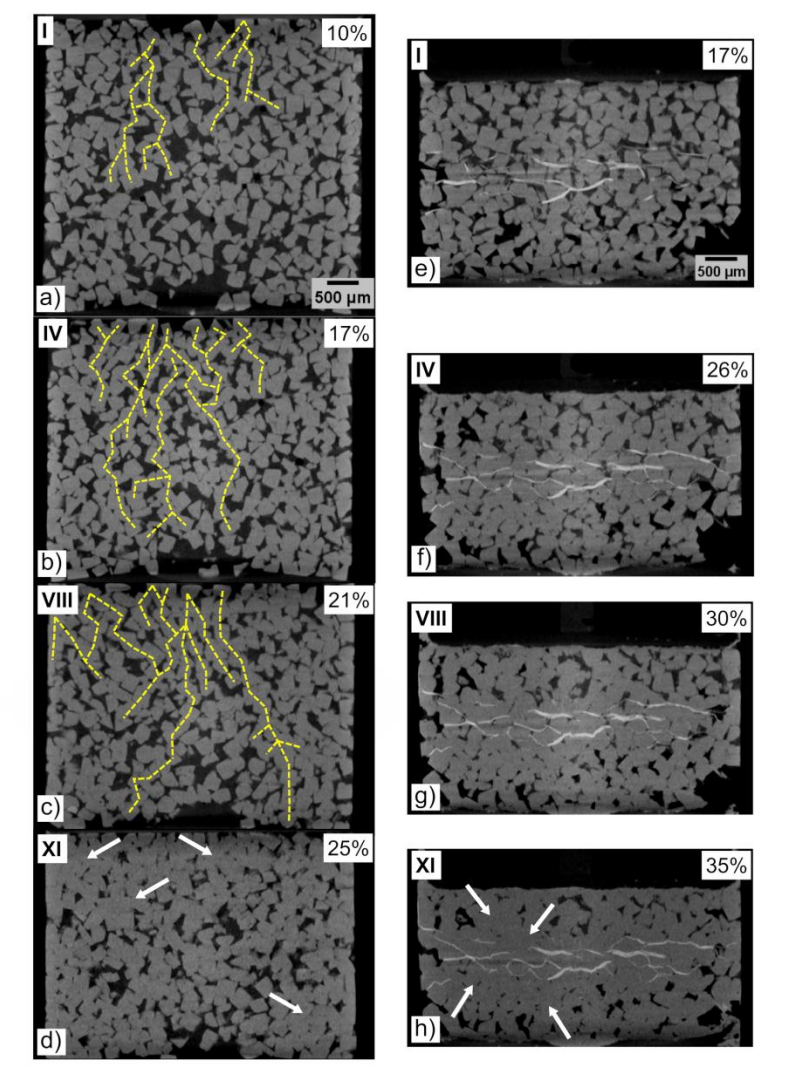

Figure S8 The yellow traced lines estimate the location of *dynamic force chains*, on top of Fig. 3.

30. Line 383-392: is the higher chemical potential a different or a similar explanation from the faster ion exchange? Unclear from this text. Wrt ion exchange, I am a bit confused, given that for PS you also need precipitation. If there is no mechanism liberating Cl, where does the Cl then come from to precipitate extra NaCl? Or are you proposing the NaCl completely dissolves?

The ion exchange hypothesis is a different explanation from the chemical gradient between dissimilar phases. This process would be strongly limited by the ion exchange site availability. The *ion activity product*, $\{Na^+\}.\{Cl^-\}$, must equal the *solubility product constant* Ksp for NaCl, but Na^+ needs not to be equal to Cl^-. Consequently, any rise in Na^+ through an ion exchange process, maintaining charge balance, would increase the ion activity product (which is assumed initially to be in equilibrium with NaCl) and cause NaCl to form. Conversely, removal of either Na^+ or Cl^- via ion exchange would achieve dissolution. Because the biotite flakes are large, the surface area is reduced and therefore the sites may be limited, contributing relatively small to the process. We therefore modified the text accordingly by discarding this hypothesis.

31. Line 396-403: the grain shape is actually odd: from a shape perspective, the extreme reduction in porosity in the biotite layer is actually odd: there is practically insoluble biotite forming a large planar surface, and then the NaCl cubes make initially a sharp contact. However, already in the first step the contact areas of NaCl against biotite are huge (Figure 8a) compared to the NaCl-NaCl contacts. So whatever makes it speed up, is relatively speaking a stronger driving force than the contact stress, since these must be low in this zone. Or are you proposing that the biotite flakes have an irregular surface (i.e. by the reference to Pluymakers and Spiers)?

We are not proposing that the biotite flakes have irregular surfaces. Rather we propose that initially the geometric arrangement of the grain contacts between a cube and a plate-like mineral may create 'asperities', with lower contact areas, which could have assisted the driving force of the mechanism. With increasing deformation these contacts became progressively smoother and possibly contribute to slow down in the compaction from 400 hours onwards. However, we believe that the chemical potential gradient between dissimilar phases is the main mechanism for the initial fast compaction rates in this sample and it explains the initial DVC local strain rates. The text has been modified to improve clarity (lines 381-385).

32. Line 422: which additional salt? The one from the fast ion exchange?

Our analyses – compaction curves vs porosity evolution – evidence that compaction correlates to porosity reduction, which means that some mechanism other than pore collapse must be responsible for the porosity reduction. In the absence of compaction, this can only be the filling of pores by precipitated salt. We are implying that NaCl from the marginal pure NaCl layers is dissolved, transported and reprecipitated in the central biotite-bearing layer, following Merino's textural model.

33. Line 429: again, where? Can you indicate a specific location?
34. Line 431: Where? Can you indicate a specific location?

The areas where salt has been reprecipitated into the biotite-layer are the areas with reduced porosity and higher grain impingements, essentially the same areas circled in Fig. 3.

35. Line 421-431: as the comment above: I like this theory, but it can be integrated a bit better with the preceding hypotheses. What is the role of each possibility, and in which part of the experiment do they play a role?

The time intervals at which each hypothesis plays a role in the experiment is outlined in the text (lines 383-386, 408 and added line 425). Along these lines, references to the figures have been included to help the readers following the discussion, and we therefore feel that there is enough cross-referencing within the text. The hypothesis outlined in lines 421-431 are only a summary of what is discussed beforehand. We provide a brief summary below:

- 0-150 hours: enhanced dissolution by chemical potential driven by different phases and initial compaction of biotite layer; this caused the biotite-bearing layer to compact, and porosity to reduce in this layer. This also caused the initial steep vertical shortening of the sample;
- 150 - 400 hours: the compaction of the biotite layer induced an increase of the contact areas, lowering the stresses at local points. It also increased dissolution in smaller contact,

higher stress areas located at the marginal salt layers. However, with increasing shortening, the contact areas in the NaCl layer increased, which lowered the stresses at contact points and, as a consequence reduced the driving force for PSC (line 408).

- 400 hours onwards: Porosity reduction in the biotite layer is accommodated by enhanced salt deposition, which lowers the driving force for PSC and slows down the process.

36. Section 4.2: given what you now have observed on the effect of the biotite flakes, do you then think stylolites can only form in rocks with a similar irregularity?

Our results show that biotite flakes contribute to reduce porosity and pore connectivity, and to increase the local strain rates as compared to neighbouring pure salt layers. It is evident from our results how biotite flakes contribute to reduce porosity and pore connectivity, and increase the local strain rates. However, at the same time, they do not seem to facilitate strain localisation (i.e. bulk compaction rates are virtually unchanged). We believe that enhanced pressure-solution is locally caused by a chemical potential driven by dissimilar phases. The effects of particle shapes, size as well as phyllosilicate types remain unclear. These are effects which we would like to explore in future studies, to integrate the current knowledge and provide a better understanding of the pressure-solution process and stylolite nucleation/growth.

List of changes

Line 20: "….The results show that for mixed samples (NaCl+biotite), ~~in NaCl,~~ phyllosilicates induce a marked..".

Line 44: "..(e.g. Aharonov and Katsman, 2009; Angevine and Turcotte, 1983; Bos et al., 2000; Croizé et al.,..)".

Line 46: " These alterations, may, in turn,..".

Line 75-80: "We use NaCl aggregates as analogues for geomaterials and sedimentary rocks undergoing PSC. Previous compaction experiments have demonstrated that NaCl samples deform sufficiently quickly to enable the study of PSC in a laboratory environment at room temperature (Gratier, 1993; Renard et al., 2004, 2001; Schutjens and Spiers, 1999; Spiers et al., 1990). ~~Despite being geomaterials in their own right, in this study, we use NaCl aggregates as analogues for sedimentary rocks undergoing PSC.~~"

Line 99-103: "grains clogging the fluid connections and both samples were saturated with NaCl solution. Initial porosity of the two sample compositions has been calculated from mass of the salt quantitatively added and the dimensions of the initial salt reference stack. The pure NaCl sample exhibits an initial porosity of ~36%, while the sample with biotite, due to the different grain packing and an initial different height (5 mm in NaCl vs 3.7 mm in NaCl-biotite), exhibits a porosity of ~33%."

Line 120: "..uniaxial stress of 6.4 to 6.6 MPa MPa."

Line 140: "…where x_0 represents the initial height of the sample (measured from the reference scans), and.."

Line 212: "..action of cataclasis, nor indented grains to have formed in the first 100 hours."

Line 254-257: "contributed relatively small amounts to the total pore volume (Supplementary Fig. S5c). Similar observations were made in the NaCl-biotite sample. ~~In the subvolume, the number of small pores (volume < 2.6 x 105 µm3) increased from 477 to 664 with increasing deformation (Supplementary Fig. S5b), while the largest pores (volumes > 2.6 x 105 µm3) showed more than a twofold decrease~~."

Line 264-265: "large pores (Supplementary Fig. S6b). ~~Overall, the number of pores decreased from 1144 to 562 over the duration of the experiment.~~"

Line 300: "$10^{-6} - 10^{-7}$ to ~~3 x~~ $10^{-8} - 10^{-9}$ s^{-1}, whereas volumetric strain rates decreased from -10^{-6} to -10^{-8} s^{-1} in…".

Line 329: "Earlier, shorter studies by Renard et al. (~~2001~~, 2004) and Spiers et al. (1990) showed a very similar…".

Line 334-342: "They concluded that the presence of clays favours grain contact healing, reducing porosity and favouring pressure-solution creep. ~~In Renard et al. (2001), mixtures of salt and clays were loaded for 15 days under different loads (0.1, 0.5 and 4 MPa). Smaller grain sizes were used: NaCl was sieved between 106 and 250 µm, while a pottery bentonite was used and sieved between one and five µm. Their results showed how the presence of small clay particles significantly enhanced the compaction rate over time.~~ Spiers et al. (1990) performed tests on brine-saturated NaCl powder (grain size 100-275 µm) at temperatures of 20-90°C and applied stresses of 0.5-2.2 MPa, testing theoretically-derived constitutive equations. For similar grain sizes and time interval, their results showed similar orders of bulk strain rates. "

Line 351: "The ~~non-linear~~ decrease in the…".

Line 382: "chains in the pure NaCl layers (Supplementary Fig. S8)."

Line 386-394: "~~Fast exchange of K+ and Na+ has been observed between biotite grains and NaCl-saturated solutions (Reed and Scott, 1962; Scott and Smith, 2013) and might have been relevant in our experiments (Bray et al., 2014; Hu et al., 2013). It is possible that in our experiments a faster ion exchange occurred between K+ in the biotite and Na+ in the NaCl-solution, making the NaCl-solution undersaturated in Na+ and enriched in K+ over time. This would cause more dissolution of Na+ from NaCl grains and transport of ions in solution, and material to precipitate in nearby pores. This mechanism would explain the reduced porosity within the NaCl biotite layer. While this mechanism probably does not account solely for the localization of PSC around biotite grains, it could have contributed~~."

Line 395-398: "The initial geometric arrangement of the grain contacts between a cube and a plate-like mineral (i.e. salt vs biotite) may create 'asperities', with lower contact areas, which could assist the driving force of the mechanism. From 400 hours onward, the process might have slowed down because reduced porosity was met by reduced asperities/irregularities in the grains, which became progressively smoother as compaction progressed (lower contact stresses). We envisage that these mechanisms contributed to pressure solution."

Line 424-425: “From 400 hours onwards, the removal of dissolved material leaves less material to carry the load…”.

Overall, the text has been revised to improve clarity.

Highlights:

- Local porosity around biotite grains is reduced by ~100 % during pressure-solution;
- Biotite increases the local strain rates by an order of magnitude;
- Pressure-solution is enhanced due to geochemical self-organization.

4D porosity evolution during pressure-solution of NaCl in the presence of phyllosilicates

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Abstract

Pressure-solution creep is one of the most common crustal deformation mechanisms, inducing changes in the porosity and permeability of rocks. For a variety of rock types undergoing pressure solution, it has been shown that the presence of phyllosilicates may significantly enhance the rate of the pressure-solution process. In this experimental investigation, we present 4-dimensional (three dimensions + time) X-ray microtomographic data that contrast deformation by pressure-solution of a pure NaCl aggregate with that of a mixture of NaCl and biotite. The results show that for mixed samples (NaCl+biotite), phyllosilicates induce a marked reduction in porosity and pore connectivity and contribute to an increase in the local strain rates by an order of magnitude over pure NaCl samples. At the same time, phyllosilicates do not induce strain localization in the sample. We discuss various possible explanations for these observations including a possible positive feedback between the porosity distribution and pressure solution. Our study yields novel insights into the local effects of phyllosilicates during pressure-solution creep and provides full 4-dimensional imaging and characterization of the coupled evolution of porosity and pore connectivity over previously unprecedented experimental time scales.

Keywords (6): pressure-solution creep; phyllosilicates; X-ray microtomography; Digital Volume Correlation; geochemical self-organization;

1. INTRODUCTION

Pressure-solution creep (PSC) is one of the key mechanisms of fluid-rock interaction and can induce significant changes in the transport properties and the composition of rocks. A wide range of geoscience applications, including hydrocarbon extraction, CO_2 storage and geothermal energy harvesting from hot sedimentary aquifers, rely on an accurate knowledge of the porosity and permeability of reservoir rocks. Understanding how these properties are affected by PSC is therefore of crucial importance.

PSC is a solution mass transfer process in response to the application of non-hydrostatic stresses that involves (i) dissolution at stressed contact points, (ii) mass transport through a fluid phase along grain boundaries, and (iii) re-precipitation of the dissolved material in the nearby low-stress areas (pores, veins) (Gratier et al., 2013; Rutter, 1983). PSC induces changes in the microstructure, pore structure, composition and strength of a wide range of geological materials (e.g. Aharonov and Katsman, 2009; Angevine and Turcotte, 1983; Bos et al., 2000; Croizé et al., 2010; Gratier, 1993; Gratier et al., 2005; Hickman and Evans, 1995, 1991; Lehner and Leroy, 2004; Pluymakers and Spiers, 2015). These alterations may, in turn, affect the transport properties of rocks (Sprunt and Nur, 1977, 1976). PSC has also been observed to trigger strain localization leading to the formation of stylolites (Gratier, 2003; Gratier et al., 2013;). Strain localisation during PSC has been recognized as involving feedbacks between solution/precipitation reactions and an evolving pore structure and thus a coupling between chemical, mechanical and hydraulic processes (Merino et al., 1983; Ortoleva et al., 1987).

While the effects of PSC on transport properties have been investigated extensively in a wide range of rocks (see Gratier et al. (2013), and references therein), the dynamic evolution of

these coupled processes at the grain scale remains poorly understood. This is primarily caused by the previously limited possibilities to image PSC in three dimensions over time. Renard et al. (2004) were the first to use 4-dimensional (4D, three spatial dimensions and time) synchrotron-based X-ray microtomography (μCT) to study the effects of PSC during compaction of pure NaCl aggregates. They imaged the reduced pore space and the impingement of grain contacts and inferred that the mean permeability in their samples decreased dramatically during compaction. Despite its pioneering character, this study did not explore PSC in the presence of phyllosilicates, which have been shown to hold the potential to significantly affect the rate of PSC (Aharonov and Katsman, 2009; Bos and Spiers, 2002; Hickman and Evans, 1995; Renard et al., 1997). Here we present 4D X-ray microtomographic data that contrast the deformation of a pure NaCl aggregate by PSC with that of a mixture of NaCl and biotite, in order to investigate the effects of phyllosilicates on the progressive shortening and the concomitant evolution of porosity and pore connectivity of the samples. The combination of μCT with *Digital Volume Correlation* (DVC, e.g. Andò et al., 2012; Tudisco et al., 2017) enables quantification of strain and strain rates at the grain scale and following of the evolution of these parameters during sample compaction. Our findings suggest that in NaCl aggregates: i) bulk porosity and pore connectivity are critically reduced in the presence of phyllosilicates; ii) phyllosilicates contribute to increase the strain rates achieved by PSC by an order of magnitude with respect to the monomineralic NaCl domains.

2. MATERIAL AND METHODS

2.1 Introduction

77 We use NaCl aggregates as analogues for geomaterials and sedimentary rocks undergoing
78 PSC. Previous compaction experiments have demonstrated that NaCl samples deform
79 sufficiently quickly to enable the study of PSC in a laboratory environment at room temperature
80 (Gratier, 1993; Renard et al., 2004, 2001; Schutjens and Spiers, 1999; Spiers et al., 1990).

81 **2.2 Materials**

82 **2.2.1 Sample Preparation**

83 Two different samples were prepared in x-ray transparent uniaxial presses (see section
84 2.2.2) to test the effect of different sample compositions on PSC. The total sample mass was
85 chosen to produce cylindrical samples with diameters of 5 mm and an initial height of ~5 mm.
86 For the first sample, granular NaCl (analytical reagent, Fisher Scientific) was sieved to a 250-300
87 µm grain size. This sample acted as our monomineralic reference. The second sample consisted
88 of a mixture of NaCl and biotite grains at a ratio, by mass, of 4:1. The NaCl was sieved to give
89 the same grain size as the first sample. Biotite flakes with grain sizes between 212 and 750 µm
90 (longest dimension) were obtained by cutting mica sheets from a biotite single crystal using a
91 scalpel. Biotite was chosen because of the contrast in X-ray absorption with respect to NaCl; in
92 µCT data biotite flakes appears as bright grains, NaCl appears as light grey grains, while (brine-
93 filled) pores are dark grey (Fig. 1). In the press, the NaCl/biotite mixture (1.4 mm thick) was
94 confined by two pure NaCl layers (0.95 mm top layer, 1.27 mm bottom layer). In this way, the
95 two pure NaCl layers constitute a 'reference' material within the sample to compare the degree
96 of compaction under the same experimental conditions. The materials were introduced into the
97 presses sequentially as slurries. Biotite within the central layer showed no preferential
98 alignment prior to compaction. In each of the two samples, two discs of filter paper were

placed above and below of the samples, to avoid grains clogging the fluid connections. Both samples were saturated with NaCl solution. The initial porosity of the two sample compositions has been calculated from the mass of the salt quantitatively added and the dimensions of the initial salt reference stack. The pure NaCl sample exhibits an initial porosity of ~36%, while the sample with biotite, due to the different grain packing and an initial different height (5 mm in NaCl vs 3.7 mm in NaCl-biotite), exhibits a porosity of ~33%.

2.2.2. Experimental Setup

The samples were compacted in X-ray transparent presses. These experimental presses were built in-house and comprise a 5 mm diameter cylindrical piston that was fashioned to be a close running-fit to the lower cylindrical sample chamber (Supplementary Fig. S3). The cells were manufactured from PEEK (Polyether-ether-ketone). Loading frames, composed of two 20 mm thick Perspex plates joined by three threaded rods, allowed a pneumatic actuator to apply a constant uniaxial load on the samples. The close running-fit of the piston enabled a narrow annulus of high-vacuum silicone grease to be used to provide a fluid seal between the piston and sample chamber.

Initial loading of the sample using the pneumatic actuator for 10 minutes was used to produce a pre-compacted starting material. At this stage, a reference μCT scan was acquired (see section 2.2.3). Subsequently, the inlet and outlet tubes to the cell, as well as the sample, were flushed with a pre - saturated NaCl solution to ensure that no air bubbles remained in the system, enabling the experiments to start saturated with brine. To maintain fluid saturation, a pre-saturated NaCl solution filled reservoir attached to the lower fluid inlet of the cell was

mounted ~40 cm above the cells (Supplementary Fig. S3). The sample was then loaded to a constant uniaxial stress which was stable between 6.4 and 6.6 MPa.

2.2.3 Image Acquisition

During each experiment, 3D µCT data were acquired using the custom-built microtomograph in the School of Geosciences, at the University of Edinburgh (see Cnudde and Boone (2013), for the fundamentals of laboratory X-ray µCT and Table S1 for data acquisition parameters). Combined, the data acquired during the experiments formed a 3D time series, i.e. 4D datasets. Compaction was interrupted for scanning of the samples. For each sample, twelve µCT datasets were acquired over 2455 hours of compaction (Fig. 2, 3, Table S2), allowing monitoring of the evolution of porosity and grain morphologies over time. Initially, the samples were scanned in short time intervals (2, 4, 6 hours), to monitor the mechanical behaviour of the samples, and capture relatively rapid initial deformation. Successively, the time interval between each scan was increased to monitor the progressively slower deformation of the sample (Table S2).

2.2.4 Image Processing

The µCT data were reconstructed using Octopus® software (Dierick et al., 2004) yielding a voxel (3-dimensional pixel) volume of 6.5^3 μm^3. The twelve microtomographic volumes acquired for each deformation experiment were analysed individually, to assess sample compaction and to measure the evolution of porosity and pore connectivity. The height of the samples in between the top piston and the bottom of the cell was measured from the vertical slices through each µCT scan, and the relative shortening was calculated through

$$\varepsilon_t = \frac{x_0 - x_t}{x_0}$$

where x_0 represents the initial height of the sample (measured from the reference scans), and x_t represents the height at a given time *t*. The µCT datasets were denoised using a *median filter* with a kernel size 2. Subvolumes of dimensions 400^3 voxels, equivalent to a cube of 17.6 mm^3 (i.e., 2.6 mm on each side), were cropped from the central regions of the reconstructed µCT volumes to conduct porosity analyses and avoid artefacts from the cell boundaries. The workflow, applied in AvizoFire® v. 8 and Avizo® v. 9, is detailed in Supplementary Figure S4. The pore space was segmented by the application of an *interactive threshold* to produce binarised image stacks from which the total porosity was determined using the *volume fraction* operator (Arns et al., 2002) (Fig. 4a). The sensitivity of the segmentation and interconnectivity analysis to changes in the threshold values was established by repeating the procedure on volumes that were eroded and dilated by one voxel (see Fusseis et al. (2012) and Macente et al. (2017) for details of the analysis). This procedure yielded error bars on the porosity measurements. Slice-by-slice (2-dimensional) porosity analyses were also performed to determine the evolution of the porosity along the direction of loading (Fig. 4b, c, d).

Pore space connectivity was characterized in the segmented data using the *label* function and a voxel coordination value of six, where the *labeling* operator identifies neighbouring voxels that share a cube face and assigns them to an individual pore cluster (Fig. 5, 6). From these clusters, individual pores were separated using the *Separate Objects* operator with a coordination number of six, and a marker extent equal to one (least connected objects). Separation points represent pore throats. Subsequently, the individual pores were analysed for their volume in the individual datasets using the *Label analysis* operator. The smallest pores considered in the pore size distribution analyses were 10 voxels in volume, or 2.7 x 10^3 μm^3.

We treated the central mixed layer and the marginal pure NaCl layers in the NaCl-biotite sample as three discrete subdomains in order to assess and compare their relative evolution. From the 400^3 voxels subvolumes, these subdomains were extracted using the *Extract volume* operator in Avizo, by identifying where the biotite flakes first occur and end vertically along the sample. The marginal NaCl layers were also compared with the pure NaCl sample.

2.3 Digital Volume Correlation (DVC)

Digital Volume Correlation (DVC) was used to measure the local distribution of displacements (from which maximum shear strains and volumetric strains are obtained) in the sample between two successive time steps (see Supplementary Material for details on the technique). DVC, which spatially resolves displacements with a sub-voxel accuracy, was performed on consecutive pairs of µCT 3D datasets using the code *TomoWarp2 (Tudisco et al., 2017)* and computed on the Edinburgh Compute and Data Facility's Terracorrelator cluster (Atkinson et al., 2015). Results were visualized in the open-source software Fiji (Schindelin et al., 2012). Misalignments in x and y, and misorientation in excess of 5° between the two 3D datasets can introduce errors in the outputs of the correlation and needed to be corrected. Two small markers at the base of the experimental cells were used together with the central fluid bore in the press to define the position and orientation of the datasets to enable the correction of misalignments and misorientations between two subsequently acquired time steps using the *rotate* and *translate* operators in Fiji. Figures 7 and 8 show the 3-dimensional results of DVC represented by XZ vertical slices through the middle of the analysis volumes for progressively shortened datasets. Absolute strain-rate values were calculated by dividing the DVC strains by the duration of loading (in seconds) between each analysed µCT scan (Fig. 9).

2.4 Scanning electron microscopy

Secondary electron images were acquired after the deformation experiments to study the grain-scale effects of pressure solution in the sample. Before retrieving the samples, the brine was displaced using compressed air. The samples were then retrieved from the uniaxial cells and portions of the samples carefully extracted. This procedure may introduce physical damage to the sample microstructures. For this reason, we only investigated evidence for PSC (i.e. indented grains, suture zones) in areas that were 'undisturbed' and represented consolidated portions of the original samples. Gold coated samples were imaged using a Carl Zeiss SIGMA HD VP Field Emission SEM with an acceleration voltage of 10 kV. Samples were mounted on an aluminium stub using a self-adhesive conducting pad prior to gold coating.

3. RESULTS

3.1 Bulk deformation

The measurements of vertical shortening show that the *pure NaCl* sample shortened a total of 25 % over 2455 hours of compaction. The compaction curve was initially steep, where the sample accommodated 10 % vertical shortening in 22.5 hours. Between 155 and 255 hours, the compaction rate slowed down. Beyond 400 hours of compaction, the gradient of the curve did not change substantially and we consider the sample to deform in steady state there (Fig. 2a). The compaction curve for the *NaCl-biotite* sample displays an identical pattern. However, the sample reached a total of 35 % vertical shortening at the end of the experiment, with very fast shortening of ~17 % in the first 22.5 hours. Compaction decelerated over the same time interval as the pure NaCl sample, again reaching what appears to be a steady state deformation. In both experiments, compaction was still ongoing at the end of the experiment.

These trends are reflected by the evolution of bulk strain rates over time (Fig. 2b). The data showed an immediate decrease in bulk strain rates after initial compaction. Some differences were present between 208 and 376 hours of compaction, when the two samples deformed less over a significant time period, resulting in lower strain rates (Fig. 2b).

Inspection of vertically centred slices through the μCT data revealed some key differences between the two samples (Fig. 3). The NaCl sample showed no microstructures indicative of the action of cataclasis, nor indented grains to have formed in the first 100 hours. However, after some compaction, porosity was reduced and grains started to agglomerate (Fig. 3c, d). In the NaCl-biotite sample, the porosity reduction was localised in the biotite-bearing layer early on (Fig. 3f). In this layer, NaCl grains lost their cubic habit with increasing compaction. Biotite generally showed a progressive alignment in the direction perpendicular to the loading, although some grains were trapped in their orientation and left without room to move (Fig. 3 e-h, centre bottom). In comparison, the marginal NaCl layers developed fewer NaCl grain impingements, and ~10 % of porosity remained after 2455 hours of compaction. These observations mirrored those from the pure NaCl sample.

3.2 Porosity evolution

3.2.1 Bulk Porosity

We analyzed the distribution of porosity change during compaction in the cropped subvolumes to identify any heterogeneities that could be indicative of strain localization in our samples and to evaluate the evolving transport properties (Fig. 4a). In the case of the pure NaCl sample, as compaction proceeded the bulk porosity was reduced from 36 to ~13 % (Fig. 4a).

Slice-by-slice porosity measurements along the vertical axis of the sample (Z-direction) confirm that this porosity reduction was evenly distributed in the sample and that no strain localization had taken place (Fig. 4b, c, d). The upper portion of the sample showed a slightly larger decrease in porosity as compared to the rest of the sample. In the case of NaCl-biotite sample, bulk porosity decreased by ~25 % over the duration of the experiment (Fig. 4a). The slice-by-slice analysis shows that at some vertical positions porosity decreased to 0 % after 2465 hours of compaction (Fig. 4d).

To better illustrate the effect of sample composition, shortening was evaluated separately for the biotite-bearing layer and the marginal NaCl layers of this sample (Fig. 5). The marginal NaCl-bearing layers were combined in the analysis. Essentially, shortening was accommodated to roughly equal parts by all three layers. The biotite-bearing layer shortened marginally more (36 %), with its thickness decreasing from 1.4 mm to 0.9 mm over 2455 hours (Fig. 5). In the same time interval, the NaCl layers shorten from 2.2 mm to 1.5 mm in total (35 % shortening). This analysis also reveals how shortening was partitioned between the three layers (Fig. 5). Initially, both layers compacted at about the same rate. At 91 hours, the shortening was partitioned into the marginal NaCl layers, a development that continued to 376 hours. At 137.5 hours the pore connectivity in the biotite-bearing layer broke down (see section 3.2.2 below), as shown by the grey shaded area in Fig. 5. This event was followed by accelerated compaction in the biotite-bearing layer and a deceleration of compaction in the NaCl layers. From 376 hours onwards, both layers compacted at about the same rate, until the end of the experiment.

We further analysed the pore size distribution in the two sample compositions (Supplementary Fig. S5) for three compaction steps (beginning, middle and end of the compaction). In both samples, the absolute frequency of larger volume pores decreased and that of smaller pores increased with increasing compaction. In the NaCl sample, small pores (volume < 2.6 x 10^5 μm^3) doubled in frequency with increasing deformation. However, these pores contributed relatively small amounts to the total pore volume (Supplementary Fig. S5c). Similar observations were made in the NaCl-biotite sample. To better understand the effect of the central biotite-bearing layer, we analysed the pore size distributions in this layer and the marginal NaCl layers in five increasingly shortened datasets (Supplementary Fig. S6). The pore size distribution for the marginal NaCl layers showed a general increase in smaller pore sizes (volumes < 2.6 x 10^5 μm^3) and a corresponding decrease in the number of larger pores, which resembled the evolution of the pure NaCl sample (Supplementary Fig. S6a). However, the central biotite-bearing layer showed a much more marked reduction for large pores (Supplementary Fig. S6b).

3.2.2. Pore Connectivity

We further analysed how compaction affected pore connectivity via pore throats (as opposed to grain boundaries, which we could not resolve in our data) in the two samples. The homogeneity of the pure NaCl sample is assumed to be responsible for the preservation of a percolating pore network throughout the sample until the end of the experiment (Fig. 6). The largest connected pore cluster (volume > 2.7 x 10^8 μm^3) initially accounted for all the pore space; however, in later compaction stages ~3 % of the porosity was accommodated by

smaller, isolated pores (Fig. 6 top). In contrast, the NaCl-biotite sample was characterized by a marked difference in the evolution of pore connectivity in the central NaCl-biotite layer and the marginal NaCl layers (Fig. 6 bottom). While the pore space was initially connected throughout the entire sample, with 99.8 % of the segmented porosity being accommodated by a single pore cluster, porosity started to disconnect vertically across the NaCl-biotite layer after 137.5 hours of compaction. Eventually two large pore clusters, disconnected from each other, remained present at the top and bottom of the sample, in total constituting 88 % of the total pore space (Fig. 6). A significant proportion of porosity was accommodated by smaller, isolated pores, mostly located at the interfaces with the marginal NaCl layers (not shown in Fig. 6).

3.3 Local strains and strain rates: results from DVC analysis

DVC was applied to every successive pair of compaction steps, yielding the evolving displacement fields during the entire experiment using eleven DVC datasets per sample. The DVC analysis of the pure NaCl sample showed a relatively homogeneous distribution of shear strains, with local maxima (Fig. 7, Supplementary Gif1). These zones correspond to zones where the volumetric strain is negative, indicating compaction (white circles in Fig. 7).

In comparison, the DVC analysis of the NaCl-biotite sample showed a much more heterogeneous shear strain distribution at the beginning and again at the end of the experiment, with a concentration of shear strains in the NaCl-biotite layer (Fig. 8, Supplementary Gif2). The analyses of volumetric strains in this sample showed a concentration of compaction in the centre of the sample (white circles in Fig. 8), with the highest strains being accumulated in the early and late stages of the experiment.

Calculating the local integrated strain rates from DVC analyses enabled contrasting the bulk compaction rate of each sample with locally resolved strain rates on the grain scale (Fig. 9). While in both samples the bulk compaction rates decreased by two orders of magnitude during the duration of the experiment (10^{-6} to 10^{-8} s^{-1}, Fig.2), locally (i.e. on the grain scale) both samples showed significant deviations in strain rates (Fig. 2). In the pure NaCl sample, the local shear strain rates decreased from 10^{-6} – 10^{-7} to 10^{-8} – 10^{-9} s^{-1}, whereas volumetric strain rates decreased from -10^{-6} to -10^{-8} s^{-1} in the most compacted areas. However, these values were evenly distributed within the sample (Fig. 9). In contrast, the shear strain rates reached maximum values of 10^{-7} s^{-1} in the presence of biotite, while the highest volumetric compaction rates were -6×10^{-8} s^{-1}. The high shear- and lower volumetric strain rates occur in the central NaCl-biotite layers (Fig. 9) and are an order of magnitude higher than in the rest of the sample.

3.4 Microstructural evidence for PSC

SEM images of the recovered samples were acquired in order to verify the microstructural changes and to collect evidence for PSC, if present, on the grain scale (Fig. 10). Indented contacts would provide evidence for PSC consistent with that proposed by Passchier and Trouw (2005) (page 31, box 3.2). In the pure NaCl sample, grains that have lost their original cubic habits are clearly visible (Fig. 10a, b). In some areas, it is possible to recognize triple junctions (Fig. 10a) and suture zones between grain interfaces, where the two grain contacts are highly indented (Fig. 10b). In the presence of phyllosilicates, indentations are obvious and NaCl grains can be seen that have dissolved against (less soluble) biotite grains, and NaCl grains that have lost their cubic habit and have become more rounded are also

315 evident (Fig. 10c, d). In the pure NaCl portions of this sample, indentations between NaCl grains
316 can still be observed, but the grains have retained more of their original habit (Fig. 3).

317 **4. DISCUSSION**

318 **4.1 Bulk and local deformation evolution**

319 Our experiments enable a direct comparison of bulk compaction behaviour with local
320 grain-scale responses and thus allow novel insights into the effects of sample composition on
321 the deformation processes. The bulk compaction curves of the two investigated sample
322 configurations (Fig. 2a) show a strong non-linear decrease in the compaction rate over time,
323 which is in agreement with findings from previous studies and support our conclusion that NaCl
324 deforms by PSC (Hickman and Evans, 1995, 1991; Renard et al., 2004; Schutjens and Spiers,
325 1999; Spiers et al., 1990). However, the duration of our experiments considerably exceeded
326 that of many published studies (Renard et al., 2004; Rutter and Wanten, 2000; Spiers et al.,
327 1990;), which allows a more detailed assessment of the evolution of compaction behaviour of
328 halite samples. Earlier, shorter studies by Renard et al. (2004) and Spiers et al. (1990) showed a
329 very similar compaction behaviour despite slightly different experimental setups. Renard et al.
330 (2004) used a smaller grain size of sieved NaCl (100-150 µm), and smaller loads (0.1 to 0.6 MPa)
331 and conducted their experiment at room temperature for seven days. The compaction rate
332 decreased over time, showing an initial steep gradient, as in our data, and reaching 18 % of
333 vertical shortening after three days. They concluded that the presence of clays supports grain
334 contact healing, reducing porosity and favouring pressure-solution creep. Spiers et al. (1990)
335 performed tests on brine-saturated NaCl powder (grain size 100-275 µm) at temperatures of
336 20-90°C and applied stresses of 0.5-2.2 MPa, testing theoretically-derived constitutive

337 equations. Their grain sizes and time intervals are similar to ours, and their results showed
338 similar orders of bulk strain rates. The data presented here explore the time evolution beyond
339 the initial steep compaction rate shown in these studies and provide a visual assessment of the
340 micro-scale processes, feeding into a better understanding of the role of phyllosilicates.

341 The striking observation from our own experiments is that both samples follow
342 essentially the same bulk compaction path, with a very rapid initial compaction characterized
343 by variable rates (Fig. 2 a, b), replaced by what appears to be steady state compaction after
344 about 400 hours. We interpret mechanical grain rearrangements and accelerated dissolution
345 rates at initially relatively small and, therefore, highly stressed grain contact areas to be
346 responsible for the high compaction rates during the early stages of the experiments. The
347 decrease in the compaction rate up to about 400 hours might broadly reflect an increase in
348 grain contact area and the associated decrease in local normal stresses as drivers for
349 dissolution.

350 From about 400 hours until the end of the experiments at 2455 hours, both experiments
351 compacted by PSC at virtually the same constant rates (Fig. 2b), which raises questions about
352 the actual effect of biotite upon PSC in the NaCl-biotite sample. In our interpretation, we
353 consider it of importance that, while the local shear strain rates reached up to 3×10^{-8} s^{-1} in the
354 pure NaCl sample, these maxima tended to be transient in terms of their location and moved
355 throughout the sample (cf. Fig. 9a). We interpret this as the capture of dynamic
356 rearrangements of stresses in the compacting sample (see below). The NaCl-biotite sample
357 showed similar magnitudes of local strain rates, but in that case, these were clearly associated
358 with the loci of biotite flakes and remained so over the entire duration of the experiment (Fig.

359 8, 9). We further observe that the presence of biotite was clearly related to the reduction of
360 porosity (Fig. 4 b-d), which, in the absence of strain localization in the central NaCl-biotite layer
361 (Fig. 5), cannot be ascribed to pore collapse there and rather points to pores having been filled.
362
363 On the basis of these observations, we interpret PSC in the NaCl and NaCl-biotite layers,
364 to have operated under different conditions in the two layers. We envisage the overall load in
365 pure granular NaCl samples, at least initially, to have been distributed over “force chains”. The
366 concept of force chains in compacting granular materials considers the overall load to be
367 distributed heterogeneously among the grains, with a minority of grains that carry above-
368 average loads forming a strong load-bearing framework, while the majority of grains constitute
369 a weak granular aggregate (e.g. Peters et al., 2005). In our conceptual model, a force chain will
370 localize pressure solution in a granular NaCl aggregate along an axis that is broadly aligned with
371 the bulk shortening direction and over an initially small grain contact area. The idea of *dynamic*
372 *force chains* entails that the lifetime of a particular force chain is limited by the dynamic
373 redistribution of loads within the granular aggregate as locally increased normal stresses lead to
374 enhanced PSC and accelerated shortening, and a relative increase of the grain surface area
375 along the force chain. When loads are redistributed, the load-bearing framework changes its
376 geometry. Our DVC analysis indicates that the strain within the pure NaCl sample varies in
377 space and time; we interpret this to indicate a rearrangement of stresses and the emergence of
378 new dynamic force chains in the pure NaCl layers.
379 While, in principal, the conceptual model described above may also apply to the NaCl-
380 biotite layer, our DVC analysis indicates that compaction there was tied to the biotite grains and

381 we interpret PSC to have been accelerated by the higher chemical potential associated with
382 phase boundaries (Aharonov and Katsman, 2009; Hickman and Evans, 1995). Factors such as
383 particle shape, particle size, and contact area may also have played a significant role in
384 controlling the evolution of PSC. Meyer et al. (2006) and Van den Ende et al.(2018)
385 demonstrated how particle shapes and the shape of the contact between the particles
386 influence compaction rates in pressure-solution. Pluymakers and Spiers (2015) emphasized the
387 significance of the nature of the contact surfaces where irregularities lead to faster dissolution.
388 The initial geometric arrangement of the grain contacts between a cube and a plate-like mineral
389 (i.e. salt vs biotite) may create 'asperities', with lower contact areas, which could have assisted
390 the driving force of the mechanism. From 400 hours onward, the process might have slowed
391 down because reduced porosity was met by reduced asperities/irregularities in the grains,
392 which became progressively smoother as compaction progressed (lower contact stresses). We
393 envisage that these mechanisms contributed to pressure solution. Whichever the reason, the
394 local strain rate data from our DVC analysis suggest that this enhanced PSC drove the initial
395 compaction in the NaCl-biotite layer (Fig. 9). However, with increasing shortening, the contact
396 areas in the NaCl layer increased, which lowered the stresses at contact points and, as a
397 consequence reduced the driving force for PSC. DVC reveals that both the local volumetric and
398 the shear strain rates decreased by almost an order of magnitude in the NaCl-biotite layer
399 between 150 and 400 hours of compaction (Fig. 9, central column), which we consider to be
400 supporting evidence for this interpretation.

401 Compaction of the central NaCl-biotite layer was accompanied by a significant reduction
402 of the porosity, to such a degree that pore connectivity broke down across the central layer

403 early in the experiment (Fig. 3, 4 and 6). However, at the same time, this layer did not compact
404 significantly more than the surrounding NaCl layers (Fig. 5). It is generally assumed that the
405 transport distances during pressure solution of NaCl in a stagnant, saturated brine are on the
406 order of the grain size and material is re-precipitated in the nearby pore space (e.g. Gratier et
407 al., 2013; Gundersen et al., 2002; Weyl, 1959). For our experiments this would mean that the
408 porosity in the NaCl and NaCl-biotite layers should have decreased in proportion with the
409 compaction. Given that this was clearly not the case, porosity reduction in the NaCl-biotite layer
410 cannot have been accomplished by pore collapse during compaction. We speculate that the
411 disproportional decrease of porosity in the NaCl-biotite layer was caused by additional salt
412 being deposited there and, invoke Merino et al.'s model for a texture-pressure solution
413 coupling to explain this (Dewers and Ortoleva, 1988; Merino et al., 1983; Ortoleva et al., 1987).
414 Merino suggests that, in a positive feedback loop, dissolved material should diffuse towards
415 areas with a relatively lower porosity and, therefore, larger grain contact area and
416 correspondingly lower stresses. In our experiment, this would mean that material was
417 transported from the pure NaCl layers to the central NaCl-biotite layer. In the former, the
418 removal of dissolved material leaves less material to carry the load, which increases the driving
419 force for PSC. At the same time, loads are distributed over an increasingly larger area at the
420 deposition sites in the NaCl-biotite layer, which should slow down PSC there. We interpret our
421 quantitative observations to provide evidence that dissolved salt has indeed been transferred
422 from the marginal NaCl into the central NaCl-biotite domain. Once connectivity into the NaCl-
423 biotite was lost, the interface between the two domains would have become the locus for
424 enhanced deposition (Fig. 3).

425 **4.2 Implications for fluid flow and permeability in rocks deforming by PSC**

426 Our experiments provide new insights into the effects of PSC on the spatio-temporal
427 evolution of porosity and pore connectivity, which influence transport properties, and
428 consequently fluid flow. These effects have been intensively studied, particularly for the case of
429 stylolites. Stylolites commonly develop in carbonates, silicate rocks, sandstones and mudstones
430 (Benedicto and Schultz, 2010; Gratier et al., 2013; Heap et al., 2014; Park and Schot, 1968).
431 They can induce up to 50 % of dissolution in the host rock with important implication for solute
432 transport and fluid flow (Aharonov and Katsman, 2009). However, it remains unclear whether
433 stylolites act as barriers to fluid flow (Alsharhan and Sadd, 2010) or become fluid conduits
434 (Heap et al., 2014;). Our experiments, which were conducted under the reported, specific
435 boundary conditions, show that the disconnection of the pore space with increasing
436 deformation leads to a very efficient hydraulic compartmentalization of the sample. The
437 porosity evolution in the central layer would suppress any potential fluid flow across the layer
438 and constrain fluid movements to the NaCl layers, at least at the scale and resolution of our
439 data. At the same time, it would allow fluid that is trapped inside the NaCl-biotite layer to
440 equilibrate chemically with its local host, whereas fluid in the percolating NaCl layers would
441 form part of an open chemical system. Our results support the interpretation by Heap et al.
442 (2014) that stylolites may compartmentalize fluid flow, allowing movement in the marginal
443 layers and in the directions perpendicular to the shortening direction.
444
445 Our findings feed into an improved understanding of fluid transport properties in
446 hydrocarbon reservoirs where PSC is active (Ghanbarzadeh et al., 2015; Torabi et al., 2015).

Alsharhan and Sadd (2010) and Heap et al. (2014) show that stylolites are capable of reducing permeability when the rock is enriched in minerals such as phyllosilicates. The effect of biotite on the permeability of the halite rocks remains unclear and will be addressed using a modelling approach in a follow-up study.

5. CONCLUSIONS

We have analysed the first 4D microtomographic dataset that documents pressure solution creep in NaCl-biotite aggregates over several months. Our results indicate that phyllosilicates contribute to reduce bulk porosity and induce a significant decrease in pore connectivity, causing a hydraulic compartmentalization of the sample. The measured local strain rates indicate that phyllosilicates enhance the pressure-solution process by increasing the local strain rates. These results highlight the key role of phyllosilicates in affecting the rock microstructure under pressure-solution creep and provide new insights into the spatio-temporal evolution of transport properties.

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FIGURE CAPTIONS

Figure 1 a) Horizontal XY µCT slice and b) vertical XZ µCT slice of the NaCl-biotite sample, showing the different attenuation values of the materials: biotite in bright colour, NaCl grains in light grey and (brine-filled) pores in dark grey.

Figure 2 a) Compaction curves for the two samples (top). Roman numbers indicated the number of the 12 µCT dataset acquired: the reference scan is not numbered (see section 3.2). b) Evolution of bulk strain rates with increasing deformation for the two samples, in log-log scale.

Figure 3 Vertical µCT slices of four different shortened datasets, for pure NaCl (left column) and NaCl-biotite samples (right column), respectively. Roman numbering refers to Figure 2a. White arrows indicate areas of reduced porosity, where NaCl grains indent more, losing their cubic habit, and/or precipitation sites.

Figure 4 a) Evolution of the pore volume (in μm^3) in the two sample compositions for increasing compaction time (hours). The percentages inside the bars indicate the porosity, calculated through the *Volume Fraction* operator from the 400^3 voxels subvolumes (see section 2.2.4). Initial porosity fraction (ϕ_0), calculated from the undeformed 400^3 voxels subvolumes, is given in the legend for the two samples. Errors bars obtained through erosion and dilation of original segmented pore space. b, c, d) Temporal evolution (in hours, 'hs') of porosity slice by slice ("Porosity 2D") as a function of the vertical Z axis for the two samples.

Figure 5 Relative shortening of the biotite-bearing and NaCl-bearing layers. The grey shaded area indicates the time interval over which pore connectivity breaks down within the NaCl-biotite layer (see section 3.2.2).

Figure 6 Interconnectivity of the pore space as a function of deformation for the two samples. Different colours indicate disconnected pore clusters.

Figure 7 DVC results for the NaCl sample, for increasing compaction time (indicated in hours, '*hs*') and vertical shortening (in percentage). Images are obtained as vertical slices through the

middle of the sample. For a better comparison, maximum shear strain (*Shear Strain*) and volumetric strain (*Volum. Strain*) results are reported together with the pair of *x-ray μCT* images used for the correlation for each interval time. Black areas identify locations where the correlation failed. Higher values of maximum shear strain are represented in the images by warmer colours, while negative values of volumetric strain (indicating compaction) are represented by blue colours (white circles). Note the relatively homogeneous distribution of shear strains, with local highs.

Figure 8 DVC results for the NaCl-biotite sample, for increasing compaction time (indicated in hours, '*hs*') and vertical shortening (in percentage). Images are obtained as vertical slices through the middle of the sample. For a better comparison, maximum shear strain (*Shear Strain*) and volumetric strain (*Volum. Strain*) results are reported together with the pair of *x-ray μCT* images used for the correlation for each interval time. Black areas identify locations where the correlation failed. Higher values of maximum shear strain are represented in the images by warmer colours, while negative values of volumetric strain (indicating compaction) are represented by blue colours (white circles). Note the extreme intense values in areas where the biotite flakes are located (high shear strains, negative volumetric strains).

Figure 9 The images show the distributions of local strain rates (in s-1), calculated from DVC results, for both samples: a) maximum shear strain rates, b) volumetric strain rates. To obtain the strain rates, the strain values, obtained from the DVC results, are divided by the time interval between each pair of correlations. Black areas identify locations where the correlation failed.

643 **Figure 10** SEM images of recovered samples: a) triple junction (highlighted by white arrows)
644 between grains in the pure NaCl sample; b) close up from the red square in a, note the suture
645 zone (white arrows) between NaCl grains; c, d) NaCl grains indenting biotite flakes (Bt) in the
646 NaCl-biotite sample.

647

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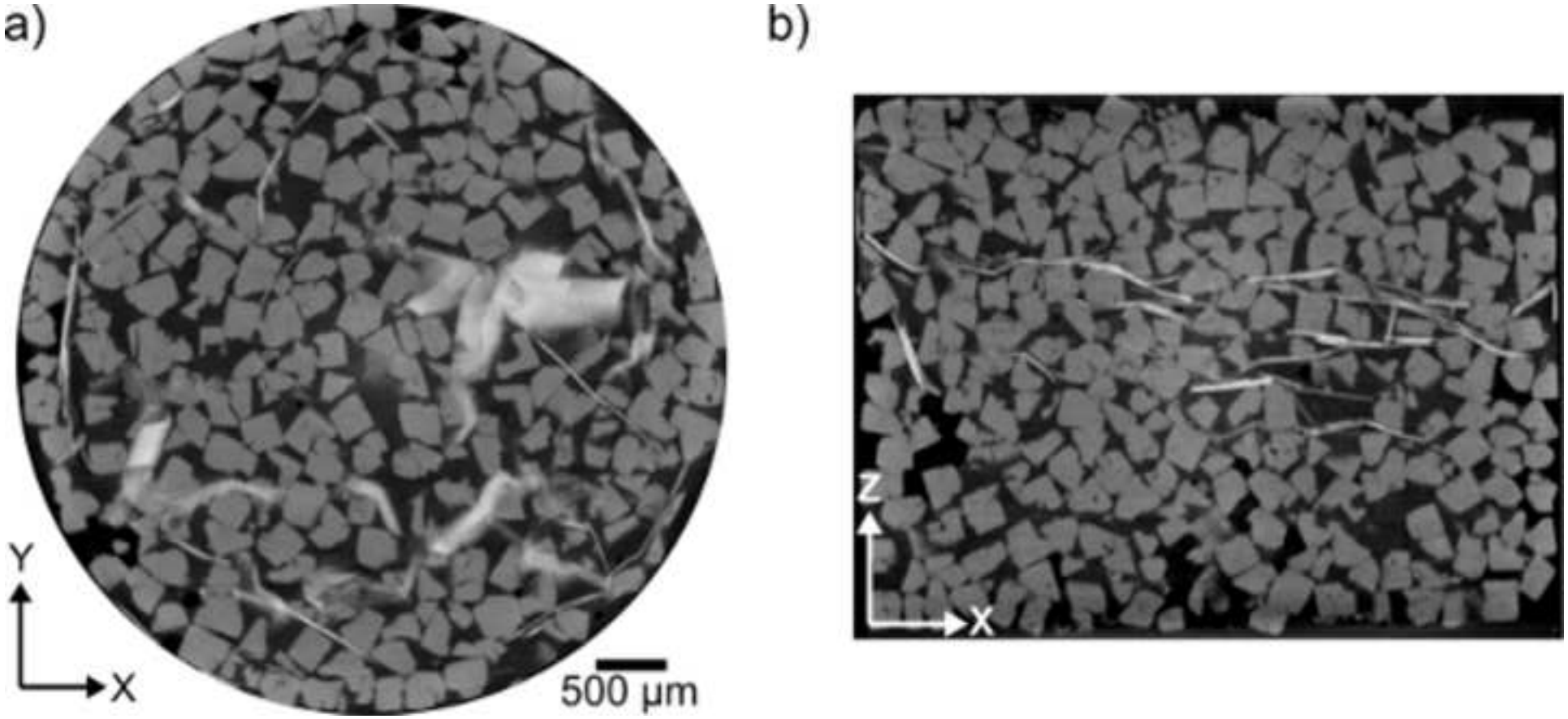

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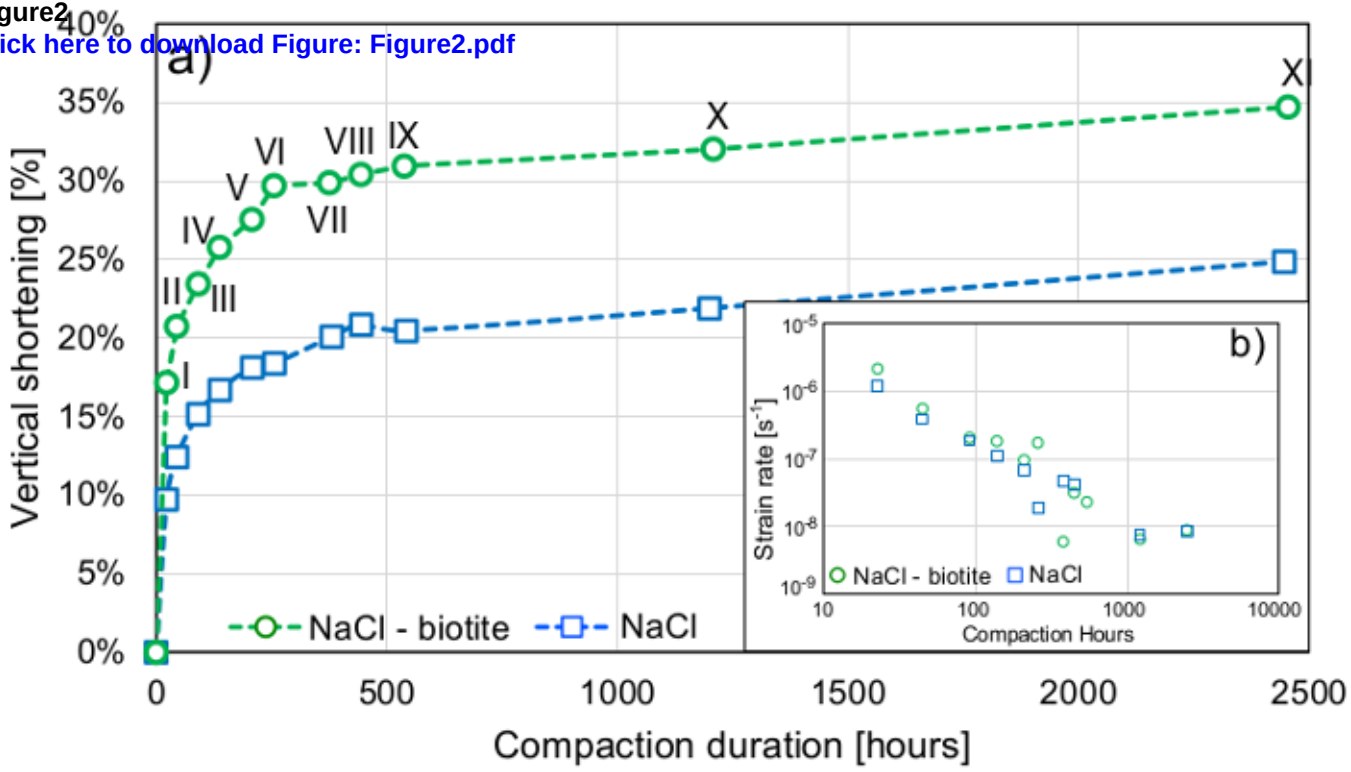

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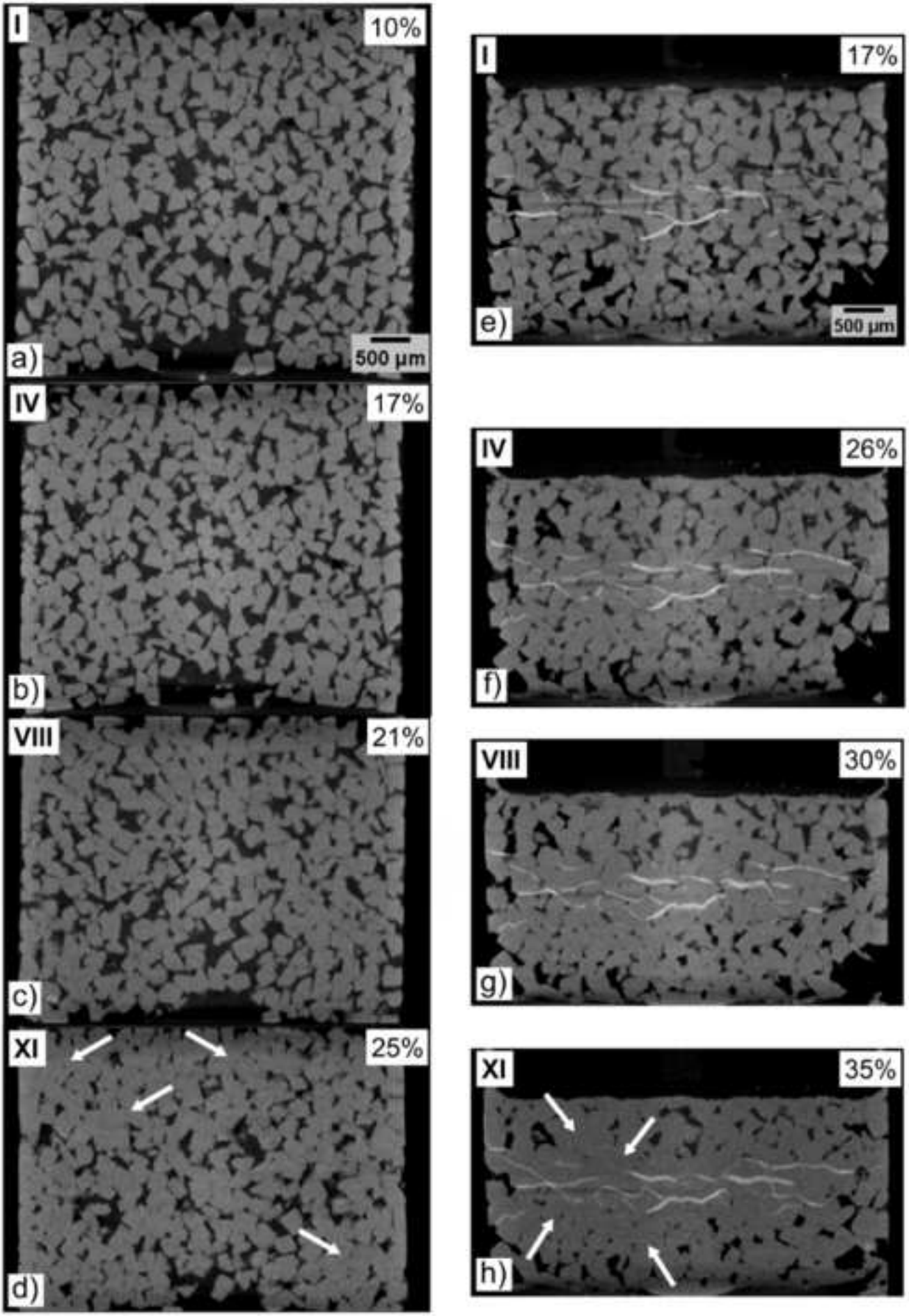

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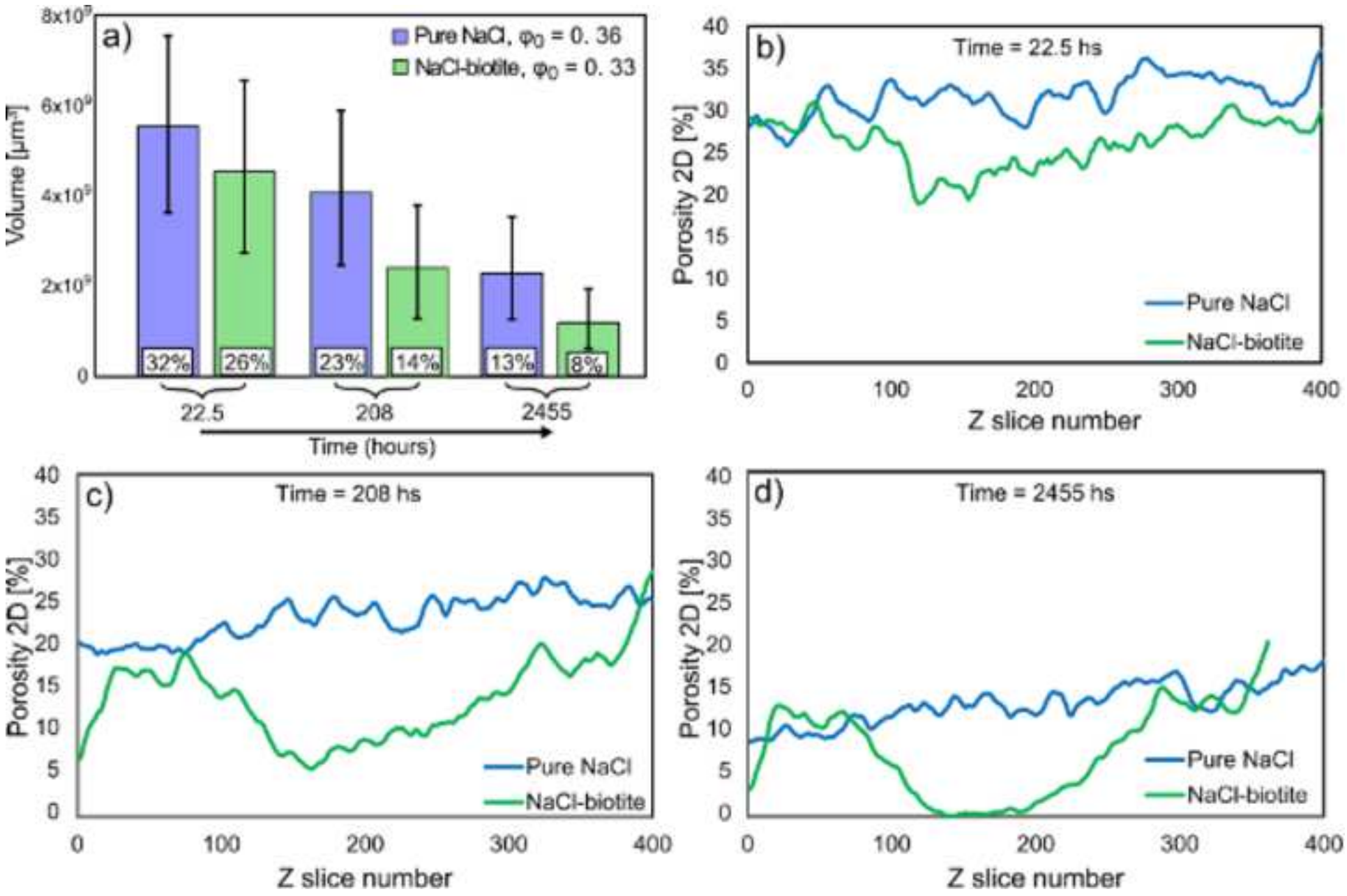

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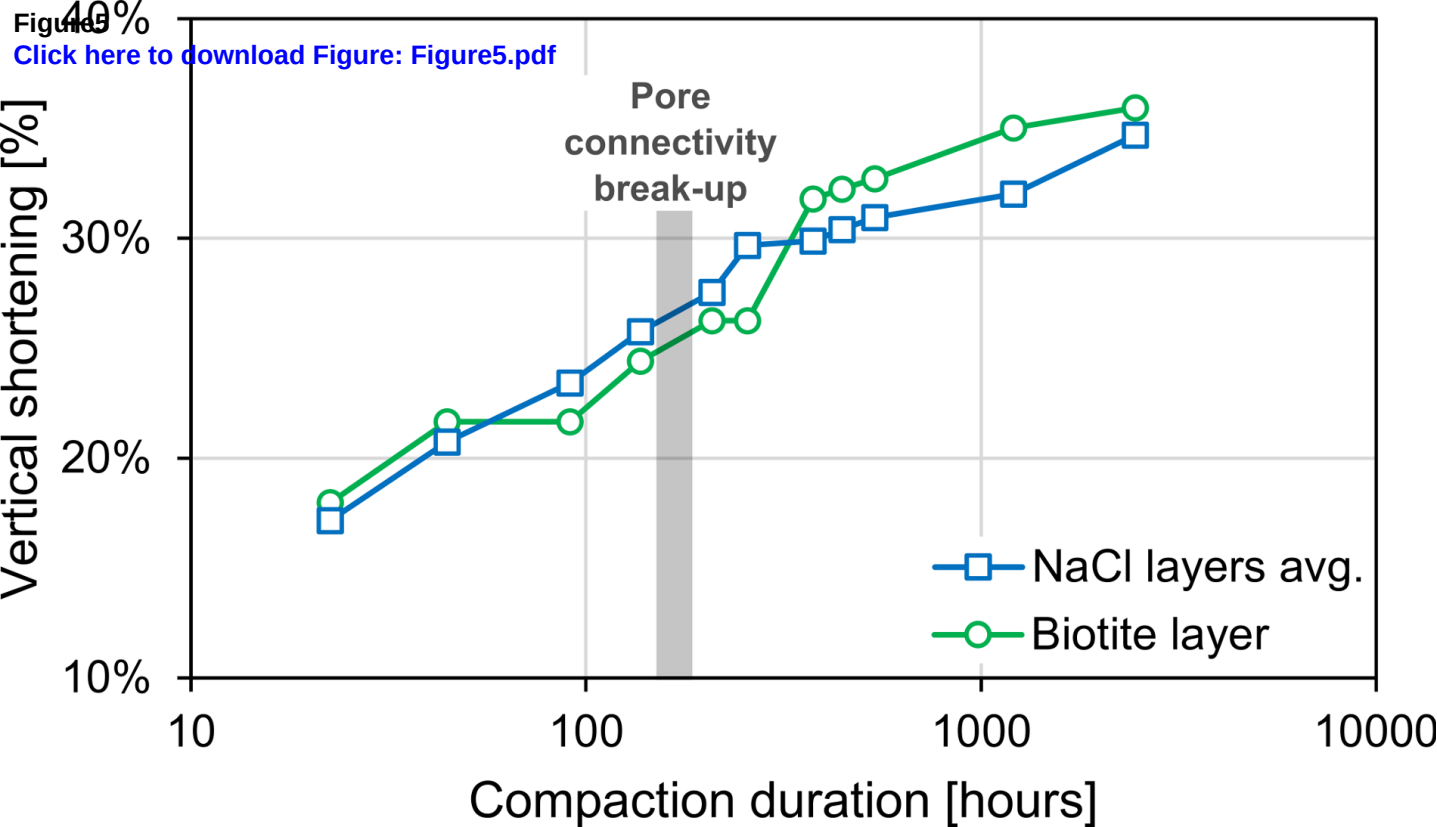

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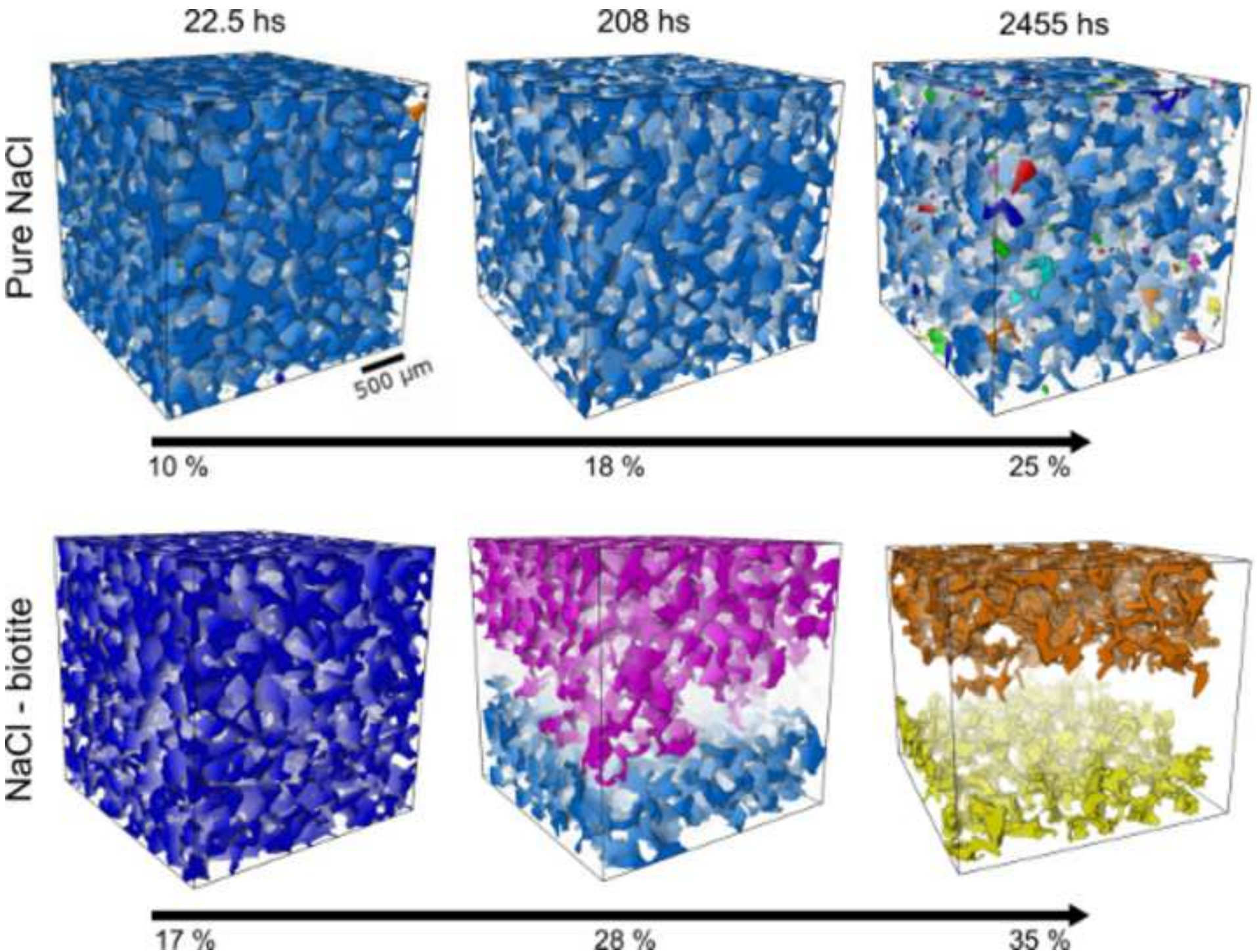

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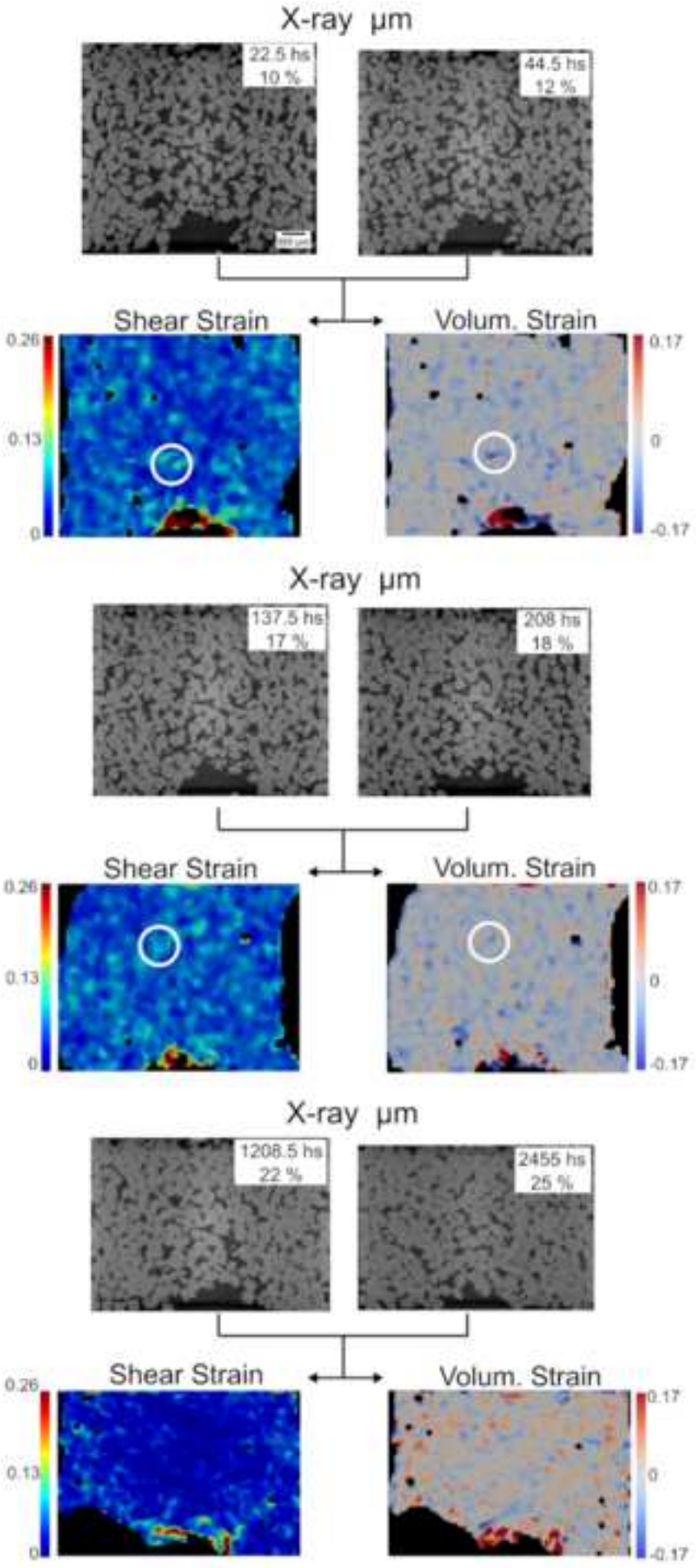

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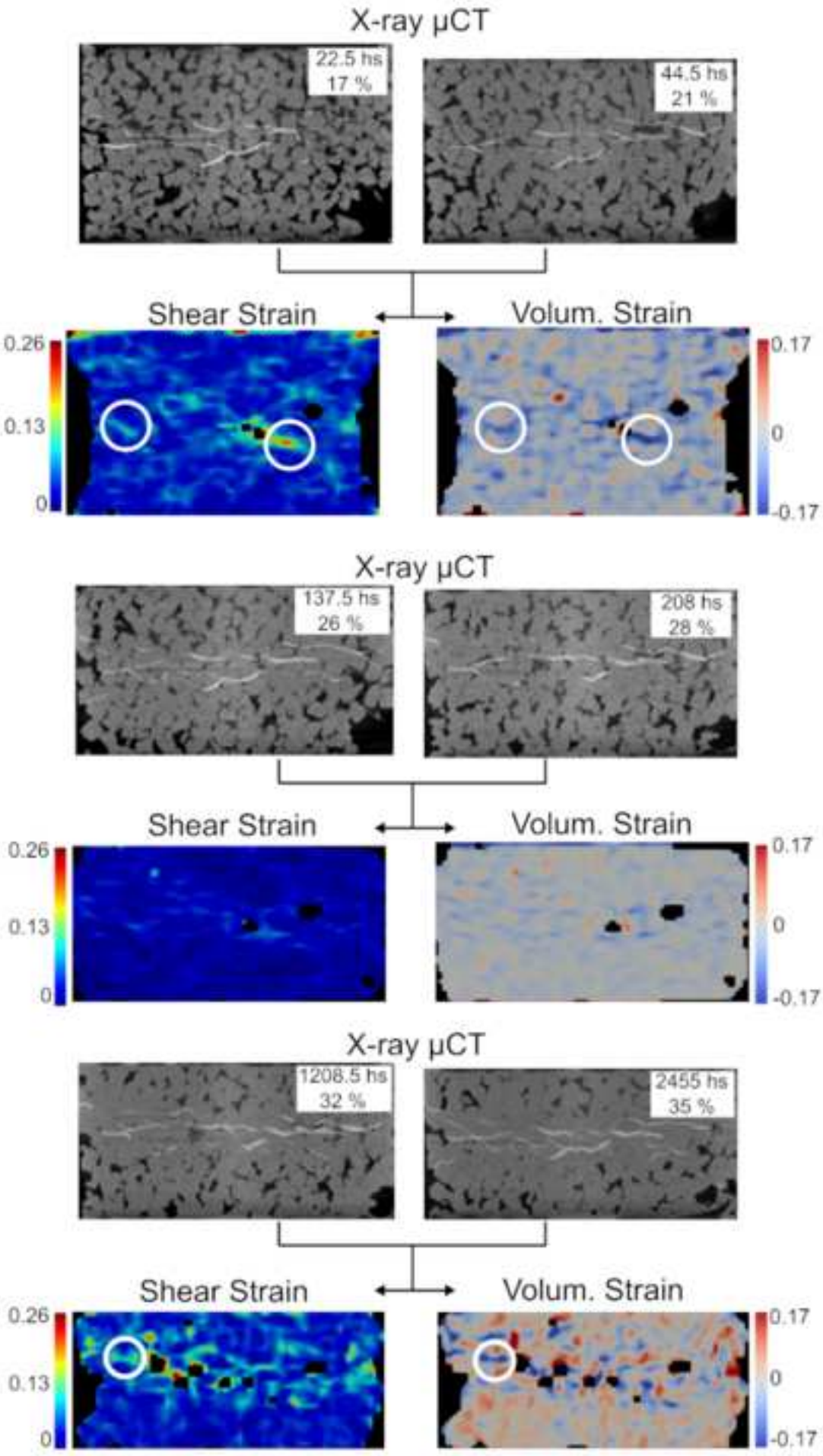

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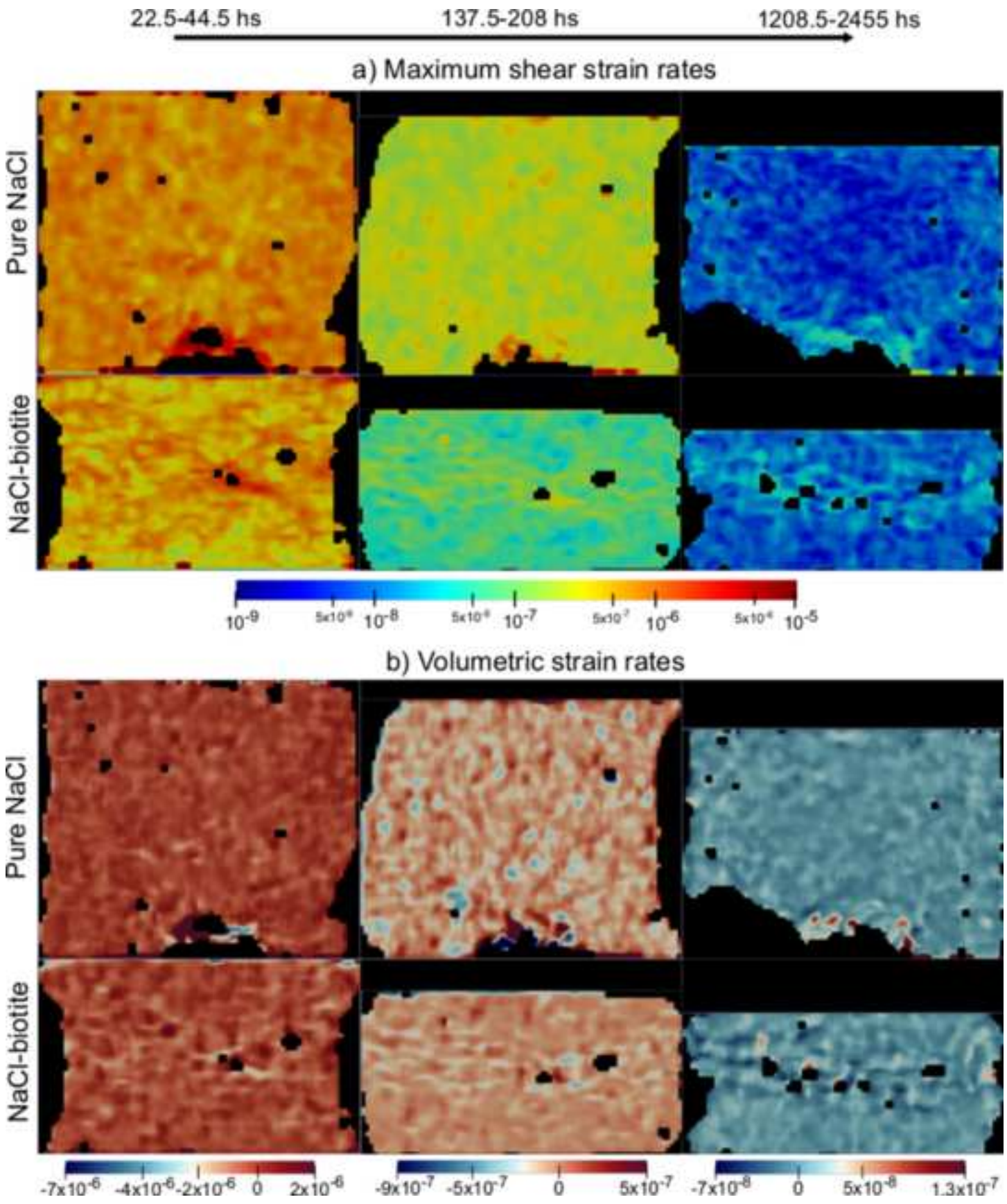

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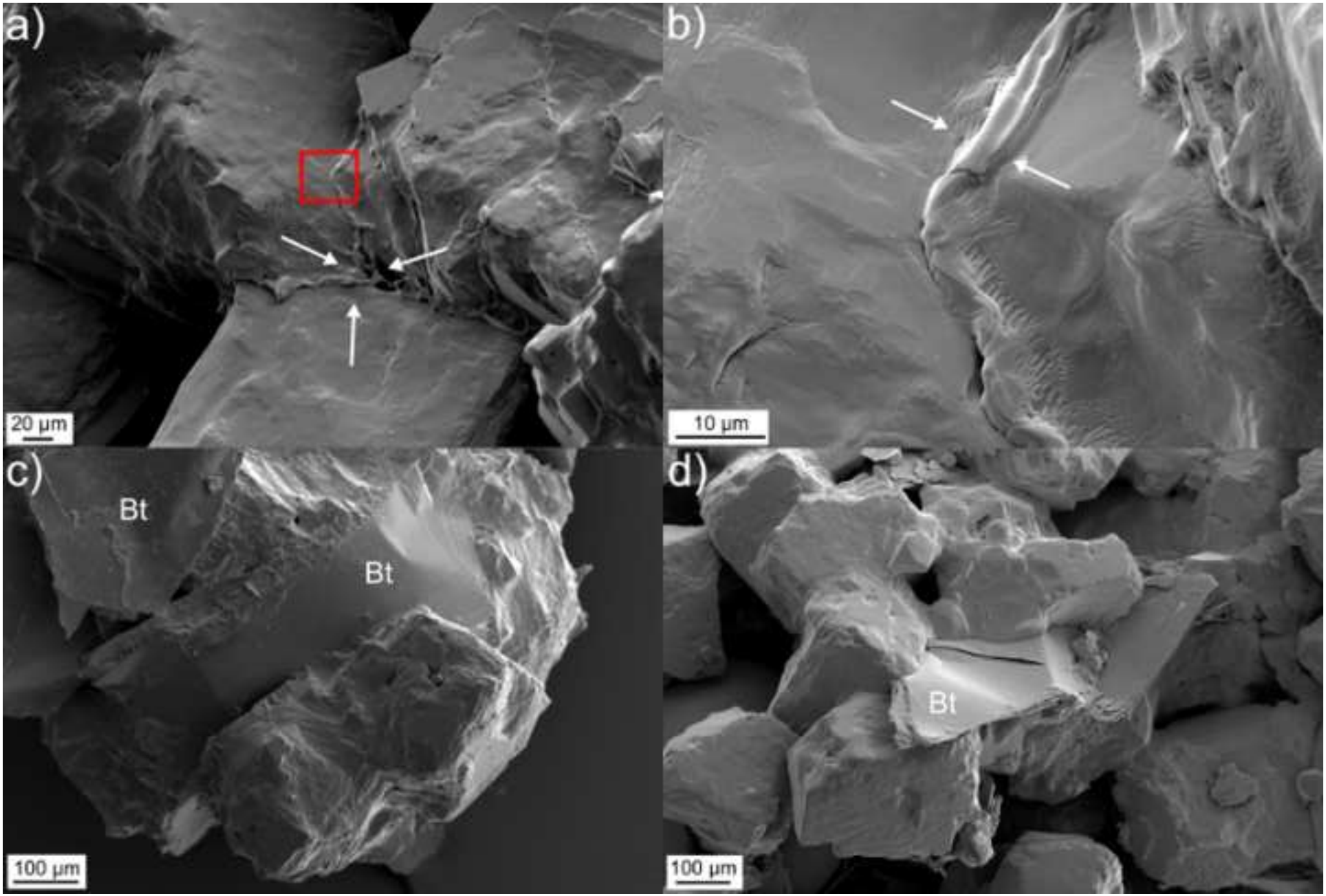

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