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Time-resolved grain-scale 3D imaging of hydrofracturing in halite layers induced by gypsum dehydration and pore fluid pressure buildup

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Abstract

Fluid release from dehydration reactions is considered to have significant effects on the strength and dynamics of tectonic faults at convergent plate boundaries. It is classically assumed that the production of fluid leads to increased pore fluid pressures that perturb a fault's stress state and thereby facilitates and enhances deformation. This important assumption has never been supported by direct microstructural observations. Here, we investigate the role of gypsum dehydration in the deformation of evaporitic rocks using synchrotron-based time-resolved X-ray computed microtomography (4D) imaging. This approach enables the documentation of coupled chemical, hydraulic and mechanical processes on the grain scale. In our experiments with deforming halite-gypsum-halite sandwiches we observe that the fluid released by dehydrating gypsum accumulates at the gypsum-halite interface before a distributed hydraulic failure of the halite layer drains the fluid. From our observations we conclude that perceivedly impermeable halite layers in evaporites are unlikely to trap overpressured fluid, e.g., in thin-skinned tectonic detachment horizons. Moreover, as the hydraulic failure is diffuse and not localized, our

experiments suggest that dehydration reactions alone may not explain intermediate depth seismicity in subduction zones. Our data demonstrate the significant potential that in-situ 4D imaging has for the grain-scale investigation of fundamental tectonic processes.

1. Introduction

The importance of mineral reactions in tectonics is evidenced by geophysical, experimental and field-based observations (e.g., Obara, 2002; Arkwright et al., 2007; De Paola et al., 2007; Fagereng and Diener, 2011). At convergent plate boundaries hydrous minerals undergo prograde metamorphism and dehydrate (Hacker et al., 2003, Peacock, 2001). The associated reduction in mineral volumes and fluid release leads to the build-up of a high pore fluid pressure (Pf) in a newly-formed porosity (e.g., Llana-Funez et al., 2012, Leclère et al., 2018). It is the generation of this high pore fluid pressure that is considered as critical for the preferential formation of tectonic detachment faults in evaporites (Hubert & Rubey, 1959) and the emergence of intermediate-depth seismic phenomena in subduction zones (e.g., Raleigh & Paterson, 1965; Hacker et al., 2003; Fagereng and Diener, 2011). Several models have been used to explain the interaction between dehydration and mechanical instabilities (e.g., Miller et al., 2003, Hacker et al., 2003, Jung et al., 2004, Chernak & Hirth, 2010, 2011, Brantut et al., 2012, Ferrand et al., 2017, Plümpner et al., 2017). These models all involve complex feedback loops that arise through the coupling of chemical, hydraulic and mechanical processes.

In these feedback loops, porosity and permeability are expected to be key variables (e.g., Llana-Funez et al., 2012, Leclère et al., 2018); they (i) define the drainage capacity, controlling pore fluid pressures and thus the effective pressure, (ii) they control where the reaction progresses and thereby influence the rock's mechanical properties and (iii) they provide chemical transport pathways through the rock, supporting fluid-mediated processes such as pressure solution creep and chemical mass transfer. These parameters and processes, all of which have been

shown to affect dehydrating, deforming rocks, are assumed to co-evolve and dynamically feed back on each other (Miller et al., 2003, Hacker et al., 2003, Jung et al., 2004, Chernak & Hirth, 2010, 2011, Brantut et al., 2012, Ferrand et al., 2017, Plümpner et al., 2017). In order to evaluate the individual significance of these feedbacks, a detailed characterisation is necessary. However, classical laboratory experiments and field-based studies can only infer active processes through indirect observations or post-mortem studies and thus do not, and cannot, observe the evolution of chemical-hydraulic-mechanical coupling at the microscale.

We report on experiments that overcome this limitation and provide robust experimental observations that (i) visualise the chemical-hydraulic-mechanical feedbacks operating during the dehydration and deformation of evaporites and (ii) quantify the microscale evolution of transport properties using time-resolved 3D x-ray microtomography (4D μ CT) data. We present data from experiments on layered halite-gypsum samples, simultaneously deformed and dehydrated in an x-ray transparent triaxial deformation rig installed at the Swiss Light Source's TOMCAT beamline. Our 4D μ CT data show that gypsum dehydration affects the capacity of the halite layers to drain the liberated fluids. The reaction itself generates the pore fluid pressure to create permeability in the halite layers through, what we interpret as, hydraulic fracturing. This potentially constitutes the first direct observation of hydraulic fracturing induced by mineral dehydration. The resulting increased permeability facilitates the transport and precipitation of solutes, supporting the dehydration reaction, and contributes to the further micromechanical evolution of the rock. Our findings help us to better understand the rheological complexities of reacting and deforming rocks. These results have repercussions for long-standing assumptions about the significance of dehydration reactions in convergent tectonic settings, in particular in thin-skinned tectonic detachments.

2. Materials and Methods

2.1 Sample preparation and Experimental setup:

Experiments were performed on layered gypsum (Volterra Alabaster) - halite samples in the form of cylinders with diameters of ~ 3.15 mm and heights of $\sim 8 - 10$ mm. Either one or two gypsum layers were chosen (Figure 1a, Table 1). Details on sample fabrications are given in Appendix A1.

The X-ray transparent, triaxial deformation rig “Mjölñir” (Figure 1b, c; Butler et al. submitted; Marone et al., 2020) was used, with 15 MPa confining pressure (P_c), a constant differential stress ($\Delta\sigma$) of 8 MPa, and sample temperatures (T) of 110 °C, 103 °C or 96 °C in different experiments, respectively (Table 1).

In the experimental setup, P_c was controlled via an external automatic high pressure syringe pump, using deionized water as a confining medium. The sample in the pressure vessel was protected from the confining medium with a silicone tube jacket (Figure 1b). Load was applied via a hydraulic actuator and controlled via an automatic high pressure syringe pump. No pore fluid pressure was applied to the sample and samples were drained to atmospheric pressure (P_{atm}) at their ends. Both sample configurations have halite layers at the top and bottom of the sample, to “seal” the gypsum layers from the drainage holes at P_{atm} in the pistons.

Samples were brought to experimental conditions by first applying and increasing an isotropic confining pressure to the desired 15 MPa (Figure 2). Subsequently the temperature was increased from room temperature to 55 °C and samples were allowed to equilibrate for ~ 1.5 h. The samples were then loaded to a constant 8 MPa differential stress, before sample heating to experimental condition was initiated. Control experiments with pure gypsum samples (Table 1) were run at the Advanced Photon Source (USA) using the same sample setup.

Table 1: List of experiments

	duration of	reaction	time until reaction
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105	Sample nr.	Setup	T(°C)	experiment	extent	extent > 98vol.%
106	SHP20	halite-gypsum d.l.	110	~ 97 min	> 98 vol.%	~ 18 min
107	SHP15	halite-gypsum s.l.	103	~ 196 min	> 98 vol.%	~ 111 min
108	SHP19	halite-gypsum d.l.	96	~ 168 min	> 98 vol.%	~ 131 min
109						
110	Gy-core 2	Volterra Alabaster	96	~ 123 min	<< 1 vol.%	-
111	Gy-core1-1APS	Volterra Alabaster	100	~ 110 min	<< 1 vol.%	-
112	Gy-core1-2APS	Volterra Alabaster	115	~ 220 min	> 98 vol.%	~ 220 min

113

114 d.l. = double layer setup, with two gypsum layers sandwiched between halite layers. s.l. = single layer
115 setup, with one gypsum layer sandwiched between halite layers.

116 APS control experiments run at the Advanced Photon Source (USA) with $P_c = 15$ Mpa, differential stress
117 = 23 MPa. Gy-Core1-1,2 i.e. experiment started at $T = 100$ °C; after 110 min, T was raised to 115 °C.

118

119

120 *2.2 Data acquisition and processing:*

121 The experimental setup was installed at the x-ray microtomography beamline TOMCAT at the
122 Swiss Light Source (SLS) synchrotron facility (Marone et al. 2020). We imaged a field of view of
123 ~ 5 mm width x 3.3 mm height at a voxel size of $2.75 \mu\text{m}^3$. The scan time for one full CT scan
124 was ~ 2 sec. At the start of an experiment, scan intervals were chosen to be at either every 0.5
125 or 1 min. In later stages of the experiment, where the rate of microstructural change slowed
126 down, acquisition intervals were reduced to every 5 - 10 min. Pressure values derived from the
127 P_c and load syringes were logged at 1 Hz. Temperature was logged every time a CT scan was
128 acquired (Figure 2b). Projection data were reconstructed to tomographic volume using a direct
129 Fourier method and an efficient data reconstruction pipeline (Marone et al., 2017). Data was
130 corrected to reduce ring-artefacts.

131

132 *2.3 Image segmentation and 3D visualization and pore volume analysis*

133 ImageJ's 3D Weka segmentation plugin was used to train a random forest segmentation
134 algorithm to create probability maps for the phases gypsum, bassanite and porosity. From the
135 probability maps, porosity was segmented using a 76% threshold. In a few cases, ring artefacts
136 occurring in the reconstructed CT scans caused artefacts in the segmentations. These were
137 removed manually.

138 The software Avizo was used for 3D visualization. Segmented pores were analysed for
139 connectivity and labeled to connected pore volumes. Pore volumes of labeled pores were
140 extracted with Avizo's "Label Analysis" tool. The minimum size of a pore was set to > 12 voxel
141 (pore volume > 249.6 μm^3 , i.e. equivalent pore diameter of > 7.8 μm).

142

143 *2.4 Theoretical background on gypsum dehydration:*

144 At the chosen experimental conditions (15 MPa P_c , 8 MPa $\Delta\sigma$, $T = 96 - 110\text{ }^\circ\text{C}$), gypsum
145 dehydrates according to:

147 The reaction is accompanied by a solid volume reduction of ~ 29 %, which causes the formation
148 of porosity around bassanite grains (Bedford et al., 2017). However, the total volume of the
149 reaction is positive with +8 %, due to the large volume of the released water. Where the water
150 cannot drain, pore fluid pressure will build up as the dehydration reaction progresses.

151

152

153 **3. Results**

154 *3.1. Overview microstructures:*

155 The processes observed in the experiments were the same at all three temperatures, but
156 microstructural changes advanced more slowly at lower temperatures. In the following, an

overview of the observed processes is given for sample SHP20 (dehydrated at 110 °C, Table 1).

The FOV for sample SHP20 included a gypsum layer sandwiched between two halite layers (Figure 3a). Initially, the gypsum layer is largely non-porous except for a few micro fractures that formed during sample preparation. Prior to the experiment, the halite layer shows a porosity of 2 - 4 volume %, in the form of isolated pores.

During heating, the dehydration reaction initiated at the halite - gypsum interfaces before the desired sample temperature of 110 °C was reached (Figure 3d - f). Bassanite formed, surrounded by 'moats' of porosity (Figure 3f, i; e.g., Bedford et al., 2017; Leclère et al., 2018). It was striking how fast the dehydration reaction occurred compared to control experiments on pure gypsum aggregate samples (Table 1). The dehydration reaction in the halite-gypsum sample progressed with a relatively well-defined dehydration front advancing from both the upper and lower halite - gypsum interfaces into the gypsum layer. From time step $t_0+6\text{min}$ onward, a significant amount of (new) fracture porosity formed within the gypsum layer (Figure 3h,i). These fractures subsequently grew and broadened. Additionally, along these fractures, dehydration to bassanite was observed ahead of the main dehydration front in some places. No reaction front was seen to travel from the outer vertical sample boundaries inwards. The gypsum layer was fully dehydrated (reaction extent > 98 %) after ~ 18 min.

Porosity in the gypsum layer evolved in line with the advancing dehydration reaction, with moats around bassanite grains forming an interconnected porous network in reacted domains. The salt layer also showed a porosity increase during the experiment (Figure 3d, f, h), with the formation of an interconnected network of tabular pores. Pores within the salt layer interconnected mainly in the vertical direction (e.g. Figure 3l).

The log for the ram syringe and the temperature log for sample SHP20 (Figure 4) show that during the time period where the dehydration reaction occurred, an accelerated outflow from the ram syringe into the load ram indicates accelerated sample shortening. Although the

dehydration reaction itself is a volume positive reaction, axial sample shortening can occur if the rock sample deforms in response to the imposed differential load. For the cylindrical-shaped samples in our case, no localisation of deformation has been observed and samples deform homogeneously, leading to barrelling of the sample (see e.g., Figure 3). The same trend of increased piston advancement during the main period of dehydration reaction is also observed in the other two experiments, SHP15 and SHP19. A notable difference is that in SHP20 (Figure 4b), the piston movement slowed down before dehydration reaction came to a full completion, whereas for sample SHP19, the reaction completed before accelerated piston movement ceased.

3.2. Porosity evolution:

To quantify the microstructural developments, porosity in sample SHP19 was segmented for 14 time-steps, in a subvolume of 1056 x 973.5 x 1600 μm (x y z) that includes the halite - gypsum interface (Figure 5). Limited by the voxel resolution of the μCT scans, but aided by a phase contrast effect, pores can be identified when they reach a size of approximately $\geq 2.753 \mu\text{m}^3$. At the start of the experiment (t_0), resolvable porosity in the gypsum layer is negligible, and porosity in the halite layer is around 1 - 2% (area % of analysed area per slice, Figure 5). There is a small peak in porosity at the halite - gypsum interface with ~ 4 area %. From then on, the increase in porosity within the gypsum layer is due to the progress of the dehydration reaction and, to a lesser degree, due to fracturing (see e.g., Supplementary Figure 3). Porosity values within the dehydrating (and deforming) part of the original gypsum layer increase to a maximum of $\sim 25 - 28 \%$ (Figure 5). This is within the theoretically expected range of solid volume loss in the gypsum dehydration reaction (e.g., Llana-Funez et al., 2012) and points to limited pore compaction.

As the dehydration reaction initiates, an irregular pore layer forms at the halite - gypsum interface (Figure 5, Figure 6a, c). This interface pore layer is seen to persist for a while in the earlier stages of the experiment but collapses eventually (Figure 6b). Simultaneously with the disappearance of the interface pore layer, porosity formation is observed in the halite layer, where tabular pores are seen to follow grain boundaries and do so preferentially in directions close to the vertical (Figure 6a, b). This evolution can also be followed in the porosity distribution plots of Figure 5. Initially, the highest amount of porosity occurs at the halite-gypsum layer interface (Figure 5b). The porosity peak however moves away from the interface into the dehydrated part of the original gypsum layer, simultaneously with a marked increase of porosity in the halite layer (Figure 5b, t0+52 min and later). The porosity within the halite then stabilizes around 5 - 7 %.

An analysis of the 3D connectivity of the pore space shows that within the well-defined dehydrated gypsum-bassanite layer a fully interconnected pore network forms early on (Figure 7a, t0+31 min). Isolated pores observed further into the gypsum layer are associated with bassanite formation ahead of the main dehydration front and some micro fracturing. In comparison, pores within the halite layer initially are small and isolated (Figure 7a,b t0+31 min). Thirty minutes into the experiment, a significant fraction of the total pore volume within the halite is made up of small pores with volumes $< 2000 \mu\text{m}^3$ (equivalent sphere diameter of $< \sim 16 \mu\text{m}$). For both halite and gypsum layers, the proportion of larger pores then increases with time (Figure 8a-d), as does the overall volume of the largest interconnected pore space. In the halite layer, the latter increases non-linearly relative to the slowly and steadily growing total pore volume in the halite layer (Figure 8 a, c). This indicates that pores “grow” not only by the formation of new pores, but predominantly by the merging of smaller pores into larger ones. This is particularly evident at t0+52 min, continued by an even more marked increase after t0+58 min. The total number of pores reaches a maximum at t0+52 min, after which the number stagnates and even slightly decreases (Figure 8e).

Porosity is a product of the gypsum dehydration reaction caused by the reaction's solid volume decrease. If compaction is limited, porosity values can be used as a proxy for dehydration extent. As previously mentioned, the reached 25 – 28 volume % porosity within the analysed volume of sample SHP19 is very close to the theoretically predicted solid volume loss and thus point to limited pore compaction in the sample. On this basis, we suggest that the linear increase in pore volume (Figure 8a, "vol.% pore" and "vol. of largest pore") indicates a relatively constant dehydration rate over the observed time period of 50 min.

3.3. Proxy for dehydration front advance rate

The mineral reaction in the tomographic images is characterized by a change in grey values due to the different absorption contrasts of bassanite and porosity compared to halite and gypsum. That is, the increased occurrence of porosity appearing in dark, and bassanite appearing in light grey broadens the initially narrow grey value distribution of the sample. We utilized this grey value change to trace the dehydration reaction and porosity changes in the time-resolved data set and extracted approximate advance rates of the dehydration front (see Supplementary Figures 1 and 2 and supplementary material for a detailed description of the procedure). For the low temperature sample SHP19, dehydration front advance rates range from ~ 9 to 15 $\mu\text{m}/\text{min}$ (Supplementary Figure 2), with the values being similar for the dehydration fronts advancing from the upper and the lower halite - gypsum interface, respectively.

4. Discussion

Our data capture and visualise the microscale evolution of dehydrating, deforming layered gypsum-halite samples, allowing us to analyse the effects of halite on the coupled chemical,

mechanical and hydraulic processes during the experiments. From our data, we infer that the halite layer initially trapped and retained the pressurized pore fluid liberated in the dehydration reaction. At the interface with the gypsum layer, dissolved halite enhanced the rate of the dehydration reaction, likely through lowering the water activity by increasing ionic strength of the brine. Starting from the halite-gypsum interface, the dehydration reaction generated a stable, well-connected pore space. Eventually, the increasing pressure of the fluid trapped in this pore space caused dilatant opening of pores along halite grain boundaries at a small angle to the imposed loading direction (which determines the largest principal stress σ_1 in the sample). We interpret this dilation in response to high pore fluid pressures as hydraulic fracturing, which allowed the pore fluid to be drained and the overpressure to be released.

No obvious acceleration of the dehydration reaction rate was observed after the pore fluid overpressure release (as would have been expected after Llana-Funez et al., 2012). However, we attribute this lack of reaction rate increase to the fact that the strongly reaction-enhancing effects of the (dissolved) halite (Bock, 1961; Hardie, 1967; Blount and Dickson, 1973; Ossorio et al., 2014) largely exceed potential rate changes inflicted from fluid P variations. We see the latter as subordinate and below detection at our relatively “low” experimental temperatures of 95 - 100 °C.

4.1 Hydro-mechanical feedbacks: Hydrofracturing limits the trapping capacity of halite layers

The tectonic significance of dehydration reactions is ultimately determined by the fluid transport properties, the pore fluid pressure and the reaction rate, which are coupled and feed back on each other (e.g., Llana-Funez et al., 2012; Leclère et al., 2018). Evidently, these feedbacks revolve around the evolving transport properties of the rock. In our experiments with layered samples, these transport properties are controlled by the advance of a well-defined dehydration front in polycrystalline gypsum, which creates a highly interconnected pore space (Figure 7; also cf. Füsseis et al., 2012; Bedford et al., 2017). By allowing for the accumulation of a growing

286 volume of increasingly pressurized fluid while drainage through the halite layer is retarded, this
287 evolving pore space controls the early hydro-mechanical evolution of the sample.

288 Our image data allow a detailed reconstruction of this process. The pore space in the gypsum
289 layer evolves from the gypsum-halite interface, where fluid released from dehydrating gypsum
290 accumulates in a prominent fluid layer (Figure 6a, c). This interfacial fluid layer is sustained for
291 several minutes, before it vanishes simultaneously with the formation of new tabular pores at
292 low angles to σ_1 in the halite layer (e.g., Figure 6b). We interpret this to show dilatancy due to
293 hydrofracturing of the halite in response to high pore fluid pressures in the fluid layer. The
294 hydrofracturing and associated pore opening in the halite then allows the fluid to be drained and
295 pore pressures to relax.

296 Published mechanical data from polycrystalline halite allow some insights into the mechanical
297 evolution of the sample in the build-up to hydrofracturing. Applying findings by Popp et al.
298 (2001) on the dilatant behaviour of halite to our experimental conditions, dilatancy should occur
299 where effective pressures are approximately $< 2.5 \pm 1.5$ MPa. This is in good agreement with
300 the observations of Peach and Spiers (1995), which would suggest effective pressures of < 5
301 MPa for dilatancy within the halite layers of our samples. Effective pressures of ≤ 4 MPa
302 suggest that the pore fluid pressure from the dehydration reaction in our experiments exceeded
303 11 MPa. These data point to a limited trapping capacity of the halite layers under tectonic
304 loading and a strong hydro-mechanical response during dehydration.

305 The importance of dehydration in the gypsum for the deformation of halite is further
306 corroborated by the observation that in SHP19, porosity in the halite layers does not increase in
307 the initial 31 minutes of the experiment, when the sample experiences both mechanical and
308 thermal loads (Figure 5b, t_0 to t_0+31 min). This indicates that differential stress and temperature
309 alone are not sufficient to cause the salt to fracture and dilate. Only the fluid pressure build-up
310 caused by the ongoing dehydration reaction and the associated changes in the effective
311 pressure eventually causes dilatancy in the halite layer.

In addition to the fractures within the halite, dilatant fractures also open within the gypsum layer (e.g. Figure 3h-i , Supplementary Figure 3a (II)). We consider these fractures to form as a response to the dehydration reaction, since the process only commences after the dehydration reaction initiated (Figure 3g-i). However, in contrast to the halite layer, fracturing in the gypsum layer should be promoted not only by the increased pore fluid pressure, but also by stress concentrations around bassanite grains and their associated porosity (cf. Bedford et al., 2017). Hydrofracturing along halite grain boundaries opens fluid pathways and at least partly drains the released water from the gypsum dehydration reaction. Once established, this porosity consolidates and matures. Porosity in the halite layer increases and becomes increasingly more interconnected (Figure 7), facilitating drainage of pore fluid and pore fluid pressure release as gypsum dehydration advances. This interpretation is supported by findings of Peach and Spears (1995), who showed that minor dilatancy (< 0.2 vol.%) during plastic deformation of salt rock can lead to very large increases in permeability. Despite its efficiency in draining the metamorphic fluid, the extent of pore interconnectivity within the halite layers is much lower than within the dehydrated part of the gypsum layer (as evidenced by the proportion of the largest interconnected pore cluster in the total porosity, cf. Figure 8). This is the consequence of different dominant pore-forming mechanisms in the two domains: reactive solid volume reduction in the gypsum layer versus dilatant hydrofracturing and subsequent dissolution in the halite.

4.2 Influence of halite on gypsum dehydration:

Control experiments on pure polycrystalline gypsum samples (i.e., Volterra alabaster cores, Table 1, see also Bedford et al., 2017) allow us to constrain how halite influences the dehydration reaction. We find a two-fold effect of the presence of halite in our samples:

1) Compared to control experiments, gypsum dehydrates significantly faster (Table 1) in the presence of halite. This is consistent with published studies which demonstrate that the gypsum to anhydrite (i.e. dehydration) transition temperature is lowered for water activities reduced by dissolved ionic species (e.g., Bock, 1961; Hardie, 1967; Blount and Dickson, 1973). Similarly, the stability range of bassanite was also found to be increased by the presence of dissolved halite (Ossorio et al, 2014).

2) The presence of the halite directly influences the spatial advance of the reaction; in all experiments we found that the halite-gypsum interface formed the nucleus of the dehydration front, and the reaction always advances normal to the interface into the gypsum layer (Figure 3, Figure 9). No reaction front was seen to travel from the gypsum layer's outer boundaries (in contact with the sample jacket) inwards, indicating that the dehydration reaction is not simply localized by any structural interface.

Once established, the synreactive porosity in the gypsum layer also provides well-connected diffusion pathways that enable halite to diffuse from the gypsum-halite interface through the dehydrated layer to the dehydration front, allowing the dehydration reaction to continue to advance at high rates.

4.3 Implications for the tectonic significance of dehydration reactions:

Our results document the grain scale processes in deforming and dehydrating rocks and therefore overcome a limitation of both, classical experimental and field based studies, which can only infer these processes through indirect measurements or post-mortem evaluation of a finite microstructure. Within their limitations, our experiments can be considered as a test for the predictions of models that link dehydration reactions with mechanical instabilities, and have direct implications for the interpretation of thin-skinned tectonic detachments. By using gypsum dehydration as an analogue for the dehydration of serpentine minerals (cf. Leclere et al., 2016,

2018), the observations may also elucidate metamorphic developments in subduction zones that are often related to intermediate-depth seismicity (e.g., Miller et al., 2003, Jung et al., 2004, Plümper et al., 2017, Dilissen et al., 2018, Clément et al., 2019).

Extrapolating the results of our experiments to nature we infer that at the depths at which gypsum dehydration can be expected in natural evaporites (~600-1000 m, Murray, 1964; Jowett et al., 1993; see also appendix section A2), and where it forms detachments in thin-skinned tectonic belts, halite layers would not be able to retain the pressurized water expelled during the gypsum dehydration reaction. Our experiments suggest that the gypsum dehydration reaction is unlikely to yield persistently high pore fluid pressures within evaporitic sequences (cf. Hubbert & Rubey, 1959, Ko et al., 1997), as even salt layers, which are considered impermeable under hydrostatic conditions, become permeable when pore fluid pressures rise under tectonic loading.

However, our experiments do show accelerated bulk sample shortening during the main phase of the dehydration reaction (Figure 4), which points to a transient destabilization of the sample caused by the dehydration reaction. We interpret this accelerated sample shortening as having been accommodated by (i) a combination of granular flow and limited cataclasis within the bassanite - pore aggregate, aided by some pore compaction (ii) grain boundary sliding and accelerated pressure solution creep in halite due to the presence of water. We suggest that similar accelerated deformation should be expected in nature after the initiation of the dehydration reaction. Now dehydrated, former gypsum layers should be susceptible to pore collapse and possibly granular flow of bassanite, whereas the water liberated in gypsum dehydration would accelerate pressure solution in salt layers and contribute to a pore-fluid pressure increase elsewhere. In combination, these processes should facilitate strain localization and thrust fault formation within evaporites.

388 In this contribution, we provide the first direct microscale observations of how dehydration and
389 fracturing coevolve, and within their limitations, these observations might be extrapolated to
390 other tectonically relevant fluid-involving reactions that lead to large volume changes.
391 Hydrofracturing as a consequence of metasomatism and dehydration has been discussed as a
392 potential source of intermediate depth seismicity and deep episodic tremor in subduction zones
393 (Jung et al., 2004, Fagereng & Diener, 2011, Chernak & Hirth, 2011, Proctor & Hirth, 2016,
394 Plümpner et al., 2017, Tarling et al., 2019). With regards to that link, our findings suggest that, on
395 the microscale: (i) Hydrofracturing is preceded by the accumulation of fluid in pores that are
396 formed by a combination of reactive volume change and dissolution. (ii) The amount of fluid that
397 accumulates in these pores before the system becomes hydraulically unstable is moderate, and
398 after the initial fracturing, the fluid supply is steady until the reaction comes to completion. (iii)
399 On the scale of our sample, the fluid does not feed into a single hydraulic fracture but
400 pervasively fractures the halite aggregate on the grain scale. While the direct applicability of
401 these results to subduction zone settings is restricted by the vastly different boundary
402 conditions, the findings do still highlight relevant aspects for future research. The most
403 provocative extension of these results is that, as dehydration does not induce localized failure in
404 our experiments, it may be that dehydration-related mechanical instabilities cannot be used to
405 singularly explain deep seismicity, and it must be a combination of factors that lead to the
406 observations preserved in the rock record (cf. Chernak and Hirth, 2011, Proctor & Hirth, 2015,
407 Plümpner et al., 2017). To this end, it would be useful to conduct further experiments to
408 understand the energy radiated by the diffuse pore fluid pressure dissipation along grain
409 boundaries and contrast this with the emissions associated with intermediate depth seismicity in
410 subduction zones. Such experiments should be conducted on serpentinites and take full
411 advantage of the enormous potential that in-situ 4D x-ray imaging holds for studies of tectonic
412 processes, including the possibility to quantitatively test models that are derived from field- and
413 experimental observations.

414

415 **5. Summary and Conclusions**

416 We present direct, time-resolved 3D observations of hydraulic fracturing in deforming rocks
417 containing dehydrating gypsum sandwiched between impermeable halite layers. From our
418 observations we conclude that halite is not able to act as a sealing cap rock for overpressured
419 fluid released from gypsum dehydration under tectonic loading. The water liberated by the
420 gypsum dehydration reaction can only temporarily be retained by the halite layers and
421 eventually drains through a fracture network induced by hydraulic fracturing within the halite
422 layers. The interaction between dehydration and mechanical instability is not localized, but
423 occurs distributed throughout the sample, i.e. no single hydraulic fracture forms.

424 Both of these observations have important implications for our understanding of how
425 dehydration reactions can affect fault formation or influence fault dynamics within the earth's
426 lithosphere. Concerning the role of gypsum dehydration in thin skin tectonic systems, our finding
427 suggests that a significant effect of overpressured fluid on basal thrusting (as suggested by
428 Heard & Rubey, 1966) within evaporitic detachment horizons seems unlikely. As far as gypsum
429 dehydration serves as an analogue for serpentinite dehydration in subduction zones, the lack of
430 localized failure in our experiments calls into question whether distributed hydrofracturing can
431 trigger mechanical instabilities that are linked with intermediate depth seismicity. The 4D μ CT
432 approach used in this study has proven to be a powerful tool in describing and quantifying the
433 microscale evolution in a complex system, combining dehydration with polyphase rock
434 deformation.

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Appendix

A1. Sample materials, sample preparation and experimental procedure:

Sample preparation:

Experiments were performed on layered gypsum - halite samples. The samples were fabricated in the following way: analytical grade crystalline NaCl (halite) (grain size < 75 μm) was mixed with just enough deionized water to produce a thick slurry. The wetted salt and whole-rock Volterra Alabaster (gypsum) cylinders (~ 3.2 mm diameter, $\sim 1.7 - 3$ mm height) were then filled alternately into a small pin press (Appendix Figure 1a). The samples were pressed at constant

pressure of 15.4 ± 0.3 MPa, at temperatures of $\sim 48 - 51$ °C for $\sim 16 - 18$ h (Appendix Figure 1b). No signs of dehydration were observed during the sample preparation procedure. Samples were cylinders with diameters of ~ 3.15 mm and heights of $\sim 8 - 10$ mm. Volterra gypsum has a very low starting porosity of 2% (Stretton, 1996). Porosity amounts in the NaCl layers were $\sim 1 - 2$ vol.% after sample pressing. Initial porosity in the NaCl layer was as isolated pores at grain junctions. This is in good agreement with the observations from Lewis & Holness (1996), which would suggest wetting angles of water $> 60^\circ$ for the P-T conditions of our sample preparation method.

Appendix Table 1: gypsum layer dimensions and calculated volume

Gypsum	Gypsum	Gypsum	theoretical	
Sample nr.	Layer height	layer width	cylinder vol.	H2O release
SHP20	~ 2.05 mm	~ 3.04 mm	14.68 mm^3	$\sim 5.3 \text{ }\mu\text{l}$
SHP19	~ 1.71 mm	~ 3.16 mm	12.74 mm^3	$\sim 4.6 \text{ }\mu\text{l}$

Values used in calculations: Molar volume of gypsum: $74.7 \cdot 10^{-6} \text{ m}^3/\text{mol}$; molar volume of H_2O : $18.018 \cdot 10^{-6} \text{ m}^3/\text{mol}$; density of NaCl : 2.17 g/cm^3

A2. Extrapolation of the experimental conditions to natural conditions:

The experimental pressures achieved in this study translate to about 566 - 612 m depth (calculating with either 2.5 g/cm^3 or 2.7 g/cm^3 average upper crustal density respectively). Assuming standard crustal geothermal gradients, temperatures at such depths will be about half those in our experiments ($T \text{ } 96 - 110 \text{ } ^\circ\text{C}$). However, it is known that gypsum does dehydrate at such depth. We have to run our experiments at higher temperatures to realize the dehydration reaction within available time scales (as opposed to geological time scales). Our extrapolation follows a standard procedure in experimental rock deformation that is referred to as “temperature-time trade-off”, which makes use of the fact that temperature-activated processes (such as mineral reactions) occur at higher rates for higher temperatures.

Supplementary Data

Image analysis - stacked histogram plots

Grey value distributions (GVD) of the sample were obtained from cylindrical volumes of 5 voxel height throughout the whole scanned sample height (Supplementary Figure 1). That is, each GVD represents a small cylindrical volume of $\sim 3 \text{ mm}$ diameter and $13.75 \text{ }\mu\text{m}$ height ($5 \cdot 2.75 \text{ }\mu\text{m}$ voxel size). GVD are presented as “stacked histogram plots” where the x axis represents grey value, the y axis represents the vertical position in the sample, and the z axis represents the grey value frequency (Supplementary Figure 2c, d).

4D microstructural evolution as seen from grey value distribution changes (stacked histogram plots):

Whereas gypsum and halite have similar grey values in the CT reconstructions, porosity and bassanite introduce darker and lighter grey values respectively (e.g., Supplementary Figure 6b (II)). The microstructure, its variations in the sample and changes during the experiment can thus be investigated by means of the grey value distribution in the CT reconstructions (Supplementary Figure 1, 2).

From stacked histogram plots, the microstructural evolution can be followed in 4D (Supplementary Figure 2). Initially, the GVDs are narrow, especially within the non-porous gypsum layer (Supplementary Figure 2c, t_0). Only in the halite layer, some remaining porosity from sample preparation causes a weak broadening of the GVD towards lower grey values. Where the dehydration reaction initiates, the growth of lighter grey bassanite and darker grey porosity cause a detectable broadening of the GVD within the gypsum layer (Supplementary Figure 2c, t_0+31 min onwards). Where the amount of bassanite starts to dominate over gypsum, the GVD peak swaps from the initial gypsum peak position to lighter grey values corresponding to the bassanite peak (Supplementary Figure 2c, t_0+64 min and onwards). The increased number of slices showing dominance of the bassanite grey value peak traces the advance of the front where gypsum has dominantly dehydrated to bassanite. This front is seen to travel into the gypsum layer from both the upper and lower halite-gypsum interface. This GVD peak swap which traces the dehydration reaction was used to calculate the rate of dehydration front advance. It is noted here that the position of the GVD peak swap is behind the outermost gypsum to bassanite dehydration front. The GVD peak swap position has been chosen as it is an easily and unambiguously traceable feature. The values that are therefrom determined for the advance of the dehydration front range between $\sim 9 - 15 \mu\text{m}/\text{min}$ (Supplementary Figure 2d) and are comparable for the dehydration front advancing from the upper and the lower halite -

gypsum interface. Advance rates seem to increase with time. Due to the limited data volume it is however not clear if this trend is significant.

Opening of new porosity in the halite layer can also be seen by a broadening of the GVD to lower grey values in the halite layer (Supplementary Figure 2). Looking at the microstructure in CT slices, newly formed pores within the halite layer appear to follow along boundaries (Supplementary Figure 3b (l)) - no indication for cataclasis of halite grains was found.

Figure 1: a) Sketch of samples. Either a single gypsum (volterra alabaster) layer or double gypsum layers were used, with the gypsum layers encased between halite layers. b) Schematic setup of the deformation rig “Mjöltnir”. The sample sits within the X-ray transparent pressure vessel (green). The upper load piston and the lower piston both have a central drainage hole. Temperatures are controlled by two externally mounted band heaters. c) Mjöltnir mounted at the TOMCAT beamline

Figure 2: Pump data and temperature log from sample SHP15. The different stages of an experiment are indicated by red lines and numbers. 1) Pressurization to 15 MPa hydrostatic pressure and pre-heated to 55 $^{\circ}\text{C}$ followed by 1.5 h equilibration time. 2) Loading to 8 MPa differential stress. 3) Temperature increase to desired experimental T . 4) Main part of the experiment. 5) End of experiment. Quenching and de-pressurization. a) Confining pressure (P_c) and differential stress log. b) Temperature log. Blue crosses indicate scan acquisition. c) Fill level of ram syringe. d) Flow rate into the load ram. Positive flow rate indicates ram advance in response to sample shortening/deformation.

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576 Figure 3: Sample SHP20 (dehydrated at 110 °C) - microstructural evolution during the
 577 experiment shown on a central slice through microtomographic datasets, intensities

corresponding to x-ray attenuation. Images in left column are coloured with the “lajolla” lookup table (LUT) of Crameri (2018), images in middle and right columns have a grey scale LUT. a) Sketch of sample. Red square marks the field of view in panels b, e, h and k. b) - d) Sample before experiment start, $T = 99\text{ }^{\circ}\text{C}$. b) Black dotted lines mark NaCl - gypsum interface. c) White arrows mark halite - gypsum interface. e) - g) Microstructure at experiment start, $T = 108\text{ }^{\circ}\text{C}$. Gypsum dehydration initiates with first bassanite (bas) grains forming in proximity to the halite interface. f) White arrows mark halite - original gypsum interface. Some micro fractures become visible (lower right). g) Bassanite grains are typically seen with surrounding pore space. A pore layer forms at the halite- original gypsum interface (white arrows). h) - j) 6 min after the start of the experiment. Dehydration reaction continued with a well defined reaction front from the halite interface into the gypsum layer. i) - j) Abundant microfractures are visible. k) - m) 12 min after the start of the experiment. The gypsum layer is almost fully dehydrated. l) - m) The pore layer at the halite - original gypsum layer interface has vanished. Marked increase in fracture porosity within the halite layer compared to the start of the experiment.

Figure 4: Temperature and Ram syringe flow rate log: Green dots mark the onset and end of accelerated outflow from the ram syringe as a response of piston advancement upon sample shortening.

a) Data for sample SHP20. Blue stippled lines indicate time of acquisition of scans shown in Figure 3b), e) h) and k). b) Data for sample SHP19. Blue stippled lines indicate timing of scans shown in Supplementary Figure 2. At around min 20, a leak in the hydraulic system caused fast fluid outflow from the load. The leak was brought under control within ~ 10 min and the experiment continued normally.

Figure 5: Porosity evolution in sample SHP19 during dehydration at 96 °C. As the halite - gypsum interface is not fully planar, the red shaded “range of salt-gypsum interface” indicates the vertical position of the upper and lower bound of the interface. a) Porosity % values for 14 time steps, showing porosity evolution within the halite and the gypsum layers. Sample sketch to the right indicates the approximate position of the analyzed volume from which the data is derived from. b) Zoomed-in vertical range and selected time steps from the data shown in a). (1)

Representation of transient porosity peak seen at the interface. (2) indicates the shift of the porosity peak to positions outside the interface range. Sample sketch to the right indicates the approximate position of the analyzed volume from which the data is derived from.

628 Figure 6: a) and b) Sample SHP20, vertical slice view. Detail of halite - gypsum interface a)
629 shortly after dehydration reaction initiation, and b) at a time step 12 min later. a) Formation of a
630 pore layer at the interface (white arrows) b) The interface pore layer has vanished (white
631 arrows), simultaneously with the formation of new porosity within the halite layer. c) Sample
632 SHP19, horizontal slice view shows irregular pore layer formed at the gypsum - halite interface
633 shortly after dehydration reaction initiation. As the interface is not fully planar, the horizontal
634 slice view “cuts” through both, the gypsum and the halite layer, with the pore layer in between.
635 Bright, needle-shaped objects are bassanite grains.

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Figure 7: Pore analysis a) Subvolume with segmented porosity rendered in 3D, sample SHP19, vertical section view (only parts of the total analyzed subvolume are shown for more clarity of the 3D structure). Each individual pore is assigned an individual color from a limited color range (i.e. colours repeat) . The subvolume contains part of a halite layer at the top, a gypsum layer at the bottom and a dehydration front traveling from the original halite – gypsum interface downwards into the gypsum layer. The large connected pore volume of the dehydrated layer shows how well porosity is connected behind the dehydration front. b) 3D rendering of the four largest pores in the halite layer at time step t_0+58'' .

Figure 8: Pore analysis in the halite and gypsum layers from subvolume shown in Figure 7. a) and b) Gypsum layer, c – e) halite layer. a) and c) The volume % of pores with respect to the total analysed volume (“pore vol. % of analysed layer”) shows how pore volume percentages increase with ongoing experiment. The volume fraction (in %) of the maximum pore to the total pore volume (“vol.% of max pore to tot. pore vol.”) gives a measure for the degree of pore

“growth” (usually by interconnection of several pores). The larger the value, the more the largest pore is permeating the sampled volume, contributing to effective drainage. b) and d) present the volume of the largest pore in mm^3 . e) Total nr. of pores in the halite layer of the analysed sample sub-volume.

Figure 9: Schematic summary of the succession of processes/events during our experiments. a) t0: starting condition. Gypsum dehydration initiates at the interface to the halite and progresses in a well defined dehydration front (t1, arrow (i)). Porosity within the dehydrated layer is widely interconnected but the halite layer at this stage is largely impermeable and a fluid layer is

building up at the interfaces to the halite (arrow (ii)). The zoom-in image in b) shows the pore fluid pressure distributions within the dehydrated layer and the halite. Microfracturing in the gypsum can be observed (iii), probably linked to high pore fluid pressures and stress concentrations around bassanite grains due to the solid volume reduction. At t_2 , the salt eventually fractures along grain boundaries (arrow (v)) and the interface fluid layer drains (arrow (iv)). Pore fluid pressures decrease (c).

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698 Appendix Figure 1: a) Sketch of sample pressing setup. Pin press with loaded sample material.

699 b) The pin press with the loaded sample material is placed in a pressing frame (red) with a

700 pneumatic actuator (green). The pressure in the actuator is controlled and kept constant via a

701 compressed air system (blue).

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Supplementary Figure 1: Stacked histogram plots. a) Grey value distributions are measured on horizontal slices, avoiding the external rim of the sample. b) Vertical section of the sample. c) Stacked histogram as 3D plot. Gypsum and halite have similar grey values, whereas the dehydration reaction causes the formation of dark porosity (lower grey values) and lighter grey bassanite. The dehydration reaction is seen in the histograms by a broadening of the grey value

722 distribution. d) Same data as in c) but plotted as image. Color codes for grey value frequency.

723 White stippled line indicates the position of the halite - gypsum interface.

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Supplementary Figure 2: 4D evolution of sample SHP19 as seen from stacked histogram plots.

a) Schematic overview of Field of View analyzed in c). b) Explanation on graphs shown in c). c) Stacked histogram plots revealing reaction advance and porosity changes with time. (I) and (II) at t_0+64 min and t_0+111 min indicate slice positions of microstructures shown in Supplementary Figure 3. Stippled grey lines indicate the range of the halite-gypsum interface (as the interface is not fully planar). Initially at t_0 , the grey value distribution is narrow in both the halite and the gypsum layer, showing almost exclusively gypsum and halite (any occurrence of porosity or the dehydration product bassanite would broaden the distribution). At t_0+31 min, the onset of the dehydration reaction can be seen by the broadening of the grey value distribution in the gypsum layer at the contact to the halite layer. As the dominant phases however are still halite and gypsum, the peak of the histogram stays unchanged. A weak broadening towards lower grey values in the halite layer indicates growing proportion of porosity within the halite layer. This broadening of the grey value distribution can be seen especially well when comparing t_0 with $\sim t_0+51$ min and later. At t_0+64 min, red arrow (iii) marks a shift in max. peak position, indicating the dominance of bassanite in this area. I.e. gypsum at this part of the sample has dehydrated largely to bassanite. Upward shift of halite-former gypsum interface positions (e.g. between t_0+69 and $+74$) is due to upward shifting of the sample stage / sample position. d) Dehydration front advance rates as calculated from data in c).

Supplementary Figure 3: Microstructures of sample SHP19 - horizontal slice ("on-top" view). Slice position in sample (I) and (II) are marked in Supplementary Figure 2 for the respective time t0+41 min and t0+111 min. Bas = Bassanite, por = porosity.

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