Dear Editor,

As requested, I have reviewed the manuscript titled "Rapid hydration and weakening of anhydrite under stress: Implications for natural hydration in the Earth's crust and mantle" by Heeb et al., please find my general and specific comments below.

Heeb et al. present data from a series of deformation experiments run on anhydrite dominated samples that come from the Òdena Gypsum Formation and one reference experiment run on Volterra gypsum. The main result of the work is to show, for the first time, that a non-hydrostatic stress state influenced the hydration reaction both in timing and in extent. These results are then brought into a geological context and discussed to give the reader explicit understanding of why the experiments are meaningful.

## General comments:

The contribution from Heeb et al. fills a gap in our understanding of deforming and reacting rocks, in particular hydrating rocks. The work is well written and the figures are generally very good at conveying the results with clarity. The science has been carefully conducted and is well detailed, which translates into clear results and a convincing narrative. It is great to see the recording of the threshold segmentation as has been done in supplementary material, it makes it very easy to assess visually what the authors have done. I particularly like the final discussion section and how it nicely captures a necessary extension of the model for décollement formation.

I have two minor comments that I would like the authors to address and one recommendation for future work:

- The first, is that the authors don't really discuss the alignment of the crystallography of gypsum with respect to the largest principal stress in detail. I find it a fascinating result that the planes in gypsum that contain the water molecules, {010}, form in the orientation of maximum shear stress. In the context of your mechanical-chemical coupling would you not want to discuss this further? These planes are also surely the weakest in the crystal structure, do you see any evidence of gypsum accommodating deformation along these planes? I know you discuss how gypsum in fractures ultimately act as locally weak regions for further shear fracturing, brecciation and eventual brittle failure, but I think you might want to make a stronger link to your crystallographic results that you have as they are probably pertinent to this argument. To be clear I am not suggesting more data are required, only that you think about linking what you already have to your existing text.
- My second comment is that you should add location data for your samples. You mention that they were collected from the field and others might want to replicate these experiments in future and collect similar samples. It would be useful to the long term reproducibility of your study to have these locations reported.
- My last comment, which I want to make clear is not something I think the authors should action in this work, is that I recommend that they move away from using simple thresholding. While their bimodal histograms clearly justify the approach, fig. S18 highlights the limitation of the method. There are clearly regions that are grain boundaries that are being identified as gypsum that aren't. If the authors wish to use the area, or volume, values that they get to say something more quantitative about the reaction progress in future then I would recommend using more advanced segmentation methods as the uncertainties cannot really be referred to as 'minor' as the authors write in the text, largely because the uncertainties are not known. As a starter, check out the machine learning feature WEKA in Fiji. Regardless of this comment, I want to reiterate that I think that the authors are justified in their methods in the work under review and do an excellent job of recording and reporting their methods.

I have some specific comments below but otherwise congratulate Heeb et al. on an excellent piece of science that was a pleasure to read.

Best wishes,

James Gilgannon

## **Specific comments:**

Line 99:

Two recent papers that stand out as missing for me here are Schrank et al. (2021) [<u>https://doi.org/10.1038/s43246-021-00156-9</u>] and Marti et al. (2020) [<u>https://doi.org/10.1016/j.epsl.2020.116679</u>].

Line 145:

'The anhydrite rocks have a minor natural gypsum content.'

Would you be able to give an estimate fraction? Even from previous studies if not from your own analysis. I think this would be good to report if you have it or access to it.

Line 241: ..., viscous layer.'

I don't think this is the appropriate descriptive word for solid reaction products. For me viscous would only work if it was a liquid that you were describing. This description is also used on line 519 and I would change it there too.

Line 327:

'... compaction contrast ...'

Do you mean that the regions have different amounts of compaction? And by this do you mean amount of porosity? I personally find compaction a confusing term here because you don't have access to the variable of compaction to compare. You only have microstructural descriptions like area of porosity.

Line 380:

'This suggests that there is an intrinsic link (or links) between the application of a non-hydrostatic stress field and the rate of the hydration reaction.'

I agree with Referee 1 (Sergio Llana-Funez) here that you would want to include a little more emphasis on permeability in your discussion. I say this because in a generic sense a low differential stress would not yield the results you have described because if the rock remained elastic and intact you would not have allowed as much access for water to allow hydration to proceed. Therefore, while a differential stress clearly has an effect, it must be partially through how it alters the microstructure of the rock. I am uncertain if one can claim that the link is differential stress -> rate of reaction from the results you present but rather, differential stress -> microstructural change -> change in permeability -> rate of reaction.

Line 517:

'The resulting lateral cheeks are either not faulted or extremely faulted, compared to the dry and 'wet' test samples.'

I found this a non-intuitive phrasing as I am not entirely sure what you mean by cheeks.

Figure 7 caption:

I might have missed it but I think the colours in b) aren't explained anywhere.