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Comment on egusphere-2022-824

Johanna Heeb (Referee)

Referee comment on "Insights into the interaction of a shale with CO₂" by Eleni Stavropoulou and Lyesse Laloui, EGU sphere,
<https://doi.org/10.5194/egusphere-2022-824-RC1>, 2022

Manuscript title: Insights into the interaction of a shale with CO₂

Author(s): Eleni Stavropoulou and Lyesse Laloui

Journal: EGU sphere

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Key aspects:

- Does the paper address relevant scientific questions within the scope of SE? *Yes*
- Does the paper present novel concepts, ideas, tools, or data? *New data is presented that adds to a field of research that is immensely relevant at this point in time.*
- Are substantial conclusions reached? *Yes. The discussion of what the findings mean in context of the 'big picture' is lacking in the current version and would greatly improve the manuscript.*
- Are the scientific methods and assumptions valid and clearly outlined? *Commonly, assumptions are made but it is not explained on what basis they are made. Overall, the scientific methods seem valid.*
- Are the results sufficient to support the interpretations and conclusions? *Yes.*
- Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)? *Yes.*

- Do the authors give proper credit to related work and clearly indicate their own new/original contribution? *In the majority of cases, they do. But in some instances, it is not clear whose work was described and in other places references are missing. Improving the style of writing (shorter sentences, etc.), and a different structure of the manuscript (separate results, discussion, and conclusions sections) should also clarify these issues.*
- Does the title clearly reflect the contents of the paper? *No. The title is not specific enough. It should clearly state what this study is about: experiments with one sample type (Opalinus Clay shale) exposed to CO₂ and studied using x-ray tomography.*
- Does the abstract provide a concise and complete summary? *The Abstract provides a short, concise summary of the study. It is intelligible to the general reader without reference to the text. The key points of the article are covered. Possible directions for prospective research could be added.*
- Is the overall presentation well-structured and clear? *Results and discussion are merged in Chapters 3 and 4. Chapter 5 is named Discussion and Conclusions but mainly presents Conclusions. I strongly suggest separating the discussion to have a clear distinction of findings vs. discussion/interpretation of results in context to the work of others vs. conclusions. This would improve the structure significantly.*
- Is the language fluent and precise? *There is much room for improvement in terms of language, style of scientific writing, and precision. The quality of the manuscript, also in terms of understanding the value of the science presented is impacted by this. Significant improvement can be achieved by following the suggestions made in the section on Technical Corrections.*
- Are mathematical formulae, symbols, abbreviations, and units correctly defined and used? *Generally, yes.*
- Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated? *Figures 4 and 5 could be merged to make it easier to compare.*
- Are the number and quality of references appropriate? *Generally, yes.*
- Is the amount and quality of supplementary material appropriate? *Yes.*

General Comments:

This manuscript presents new live x-ray tomography 2D and 3D data from long-term injection tests of liquid CO₂ into Opalinus Clay shale samples. The overall objective is to study the capacity of shale as caprock for CO₂ storage under realistic pressure and temperature conditions. The study sets out to reveal potential chemo-mechanical processes and improve the understanding of localised Thermo-Hydro-Chemo-Mechanical (THCM) interactions in the shale under natural conditions.

The manuscript focuses on microstructural variations and kinematics of Opalinus Clay shale when exposed to CO₂ in two different test modes, studied via 3D image analysis of real-time x-ray tomography. For long-term testing, liquid CO₂ (8 MPa) was injected into samples and held under confined conditions (10 MPa) for 9 months under constant volume conditions in a PEEKcell. For the second line of testing, samples were exposed to supercritical CO₂ (P = 10 MPa, T = 34°C) for up to 56 days. A combination of SEM-EDX mapping and x-ray tomography was used to analyse changes in mineralogical composition and structural evolution of the samples before, during, and after testing. The 3D volumetric response was analysed via digital volume correlation (DVC). Normalised grey level values (GV) x-ray imaging visualised and ultimately quantified CO₂ penetration into

the material.

Long-term injection of liquid CO₂ resulted in re-arrangement of the pre-existing micro-fissures in the clay matrix and significant fissuring of calcite-rich zones that were for the first time visualised and quantified from x-ray images. Tests with exposure to supercritical CO₂ showed initial swelling at pre-cracked zones and new micro-fissures in areas of direct contact to CO₂. Advanced 3D image analysis showed an increasing CO₂ uptake with time and potential CO₂ trapping in the material.

The most significant findings include: successfully proving the increasing CO₂ uptake and potential CO₂ trapping in the Opalinus Clay shale. The method used as well as the results will need further work and will have importance for evaluating and monitoring the integrity and stability of shale caprocks in CO₂ storage.

I found that the manuscript contains a variety of useful data that has the potential to add to what is known about CO₂ storage regarding shaly rocks. However, I also identify many issues that need to be addressed before the manuscript can be considered for publication in EGU sphere or any other Journal. My assessment is that, once the raised issues have been adequately addressed (if possible), the manuscript could be acceptable for publication. In my view, the extent of the changes required amounts to a major revision.

Specific Comments:

Issues range in significance and include: a lack of articulation of the significance of the study in regards of the 'big picture', sub-optimal structure of the manuscript (particularly the results and discussion), and generally poor quality of writing (See section on Technical Corrections and line-by-line comments).

1 - The study sets out to reveal potential chemo-mechanical processes and improve the understanding of localised Thermo-Hydro-Chemo-Mechanical (THCM) interactions in the shale under natural conditions. I recommend that the author elaborates more on how the results of their study fit into the bigger context, potentially by adding a sub-section to a possible Discussion section.

2 - Structural changes to improve readability:

a) Separate observations from interpretations and methods. Description of results is often blended with interpretations of the data, methods applied and comparisons with published literature (which should be done more often). Please could the author revise the results so that all interpretations of the data and comparisons with published literature are moved to

a new, separate discussion or new sub-chapters.

b) Long sentences with lots of sub-sections make it harder to understand the precise meaning of sentences. They also divert the attention away from the main message of a sentence and distract the reader. Please could the author consider shortening sentences in order to be more precise on what they want to say.

Technical Corrections:

The manuscript is riddled with major writing style idiosyncrasies and needs a lot of revision. Detailed suggestions on how the text can be improved can be found in a line-by-line comment section. The following issues and others (I recommend also consulting the author guidelines) repeatedly appear throughout the text:

- Avoid words/expressions like 'unveil', 'embrace', 'thanks to', as they are very casual.
- Avoid using possessive nouns (the 's). For example, instead of 'the caprock's response', use 'the response of the caprock material'.
- Avoid ';' in regular sentences, see lines 75, 187, and throughout. They are preventing the writing to be more fluid.
- Commas are frequently missing or in the wrong position, see Lines 89, 91, 93 and throughout the document.
- *Italic* font may be used for emphasis, although this should be used sparingly (see journal guidelines). See Lines 384, 444, ...
- If the author's name is part of the sentence structure only the year is put in parentheses ("As we can see in the work of Prakash et al. (2022) the precipitation has increased"). If the author's name is not part of the sentence, name and year are put in parentheses ("Precipitation increase was observed (Smith, 2009)") – per journal guidelines
- Use metaphoric terms instead of clear objective terms ('opening the door' instead of 'possibly', 'on the other hand' instead of 'conversely')
- Avoid incorporation of superfluous words/phrases (e.g., 'indeed', 'will be discussed in section xx', 'as discussed above')
- Avoid erroneous usage of temporal terms instead of comparative terms where no time frame is implied (e.g., 'while' instead of 'whereas', 'occasionally' or 'sometimes' instead of 'rarely' or 'less commonly', 'often' instead of 'commonly')
- Use precise or quantifiable language instead of vague and/or relative terms (e.g., 'relatively high/low', 'a very long time', ...)
- As per the journal guidelines on capitalization: only the first word is capitalized in headers (in addition to proper nouns) – per journal guidelines
- From the journal guidelines: The abbreviation "Fig." should be used when it appears in running text and should be followed by a number unless it comes at the beginning of a sentence, e.g.: "The results are depicted in Fig. 5. Figure 9 reveals that...".
- Be consistent throughout the manuscript. For example:
 - Opalinus samples, Opalinus Clay shale, Opalinus Clay samples
 - 8 % and 8% (spaces must be included between number and unit, e.g., 1 %, 1 m)
 - In situ and in-situ (Latin phrases should not be hyphenated (e.g., "in situ", not "in-situ"))

Figures:

Figure 1

This figure is very self-explanatory. Caption: please use 'grey values' instead of 'greyvalues'.

Figure 3

Visually separate the columns and/or separate the headlines by a different font or boldness to make it clear at first view that the bottom sections are also from the initial / after 9 months material.

Figure 6

Please add to the caption explanations: what type of sections, what type of imaging, what the red dotted lines are marking, ... also consider adding (a), (b), (c), (d) to mark the panels.

Figure 7

Name the columns and describe them in the figure caption. What type of imaging was used? Named panels make it easier to highlight significant features throughout the text. Add arrows to show there is a progression.

Figure 8

Consider naming the panels (a), (b), (c), (d), (e). An arrow or several arrows would indicate to the reader that there is a progression. Add a scale and legend. Mark the significant areas in some way to make it easier for the reader to know what you are discussing specifically.

Figure 9

Please consider naming the rows (panels), add arrow(s), mark the most significant feature(s), and consider making the images of the second row larger, as it is difficult to see features.

Figure 10

Overall, this figure presents a great summary of the work. I suggest making the subsets larger, as the current size makes it difficult to see what is going on.

Line-by-line:

Line 11

... the long-term integrity of the caprock.

Line 12

Please be more specific, what is a 'very long time'? Compared to what?

Lines 15-16

Please revise/split this sentence for better understanding.

Line 19

... resulted in significant fissuring ...

Line 21

'a re-arrangement ... was observed'

Lines 24-25

The meaning of this sentence is not clear. Please revise.

Line 30

Please be more specific. Relatively high compared to what?

Line 50

Replace 'in' with 'on'

Line 53

Please specify what type of anisotropy. Mechanic, seismic, acoustic, ...? Heterogeneous in terms of what? Mineral composition, fabrics, ...?

Line 59

Consider replacing 'thanks to' with 'because of' OR: ... due to the fundamental mechanical/physical properties of the material, such as ...

Line 64

Please explain what is meant by 'representative boundaries'. Critical limitations for testing, perhaps?

Line 86

What are 'large scale experiments'?

Line 71

Please consider revising this sentence.

Line 72

... on that scale.

Line 74

... on the fundamental material properties ...; Consider what properties: i.e., physical, mechanical, ...

Line 75

Replace ';' with ',' and consider moving the cited authors: ... injection tests in Opalinus samples (references) do not show evidence ...; The references could be moved to the end of the sentence.

Lines 77-78

Please clarify what this statement is based on.

Line 82

This sentence could be improved by making clear in what regard the measurements not enough. Also: consider replacing 'not enough' with 'not sufficient' or something similar.

Line 88

Consider replacing 'on' with 'about': ... conclusions about their impact on the structural properties ...

Line 89

Add a comma: ... and hence, transport and mechanical response.

Line 91

Add a comma: Flow is extremely slow, resulting in ...

Line 93

Add commas: ... since injection pressure, and therefore effective stress, has an important

...

Line 95

Please state what has slow transport properties and add ';' before (v) to be consistent. Consider revising (v), as it does not seem to fit into the context of (i-iv). Give more details about the scale?

Line 99

non-destructive

Lines 101-104

Commas?

Line 109

Please change the headline to: ... Analytical Principles

Line 117

Consider using another term instead of 'exploration'. For example: 'development'.

Line 126

Please consider: ... in order to improve the temporal resolution ... OR ... in order to increase the temporal resolution ...

This could be split into two sentences instead of using ()

Line 129

... subject. The pore size ...

Line 131

Be more specific, for example CO₂ injection / exposure / ...

Line 135

Please specify what type of heterogeneity and anisotropy

Line 136

Microstructures

Line 141

... to identify the heterogeneity of the specimen in 3D ... What type of heterogeneity?

Line 142

To be more specific you could change the beginning of this sentence: X-ray Tomography is a very ...

Line 145

Please consider specifying what type of platform this is?

Line 149

Replace 'of' with 'at'

Line 150

Explain what PEEK is / stands for

Line 157

... is placed on both ends (). OR 'Two pressure transducers are in position at the top and the bottom of the cell to monitor pressure levels during the scans.' Sides are also left, right, front, back.

Line 158

This sentence could be improved by not using a ';'. For example: The samples ... mm by cutting rectangular pieces with a saw, followed by ...

Line 177

'a pressure drop' OR specify how it drops: successive/constant/rapid at the end/beginning/middle, ...

Line 187

... are reflected ...

Line 187

Replace ';' with '.' and start a new sentence.

Line 197

Make sure all variables are explained.

Line 202

Please take a look at the citation style / grammar of this sentence and amend accordingly.

...

Line 206

... monthly ... ALSO consider revising this sentence and/or split it into two.

Line 215

Please check the grammar of this sentence. ... based on a single ... for the entire image ...

Line 226

Consider using 'Furthermore' or something similar instead of 'On the other hand'.

Line 235

Please replace 'is' with 'are'

Line 237

What is meant by 'demonstrate elements'? Show the presence of elements perhaps?

Line 238

Consider replacing 'by means' with 'by studying'

Line 243

Consider putting the evaluation at the end of the sentence / section. 'quite approximate' is vague, be more specific, for example by citing numbers.

Line 247

The meaning of this sentence is unclear due to grammatical and structural issues. Please revise.

Line 248

Consider moving this before presenting the Sample A results.

Line 249

analytical

Line 252

... Figure 2-a, using sandpaper.

Line 254

Be consistent: Figure 2-a OR Figure 2 (a).

Line 256

Consider starting the sentence like this: There are two sets of ...

Line 257

Consider rewriting this sentence, for example: 'This becomes more significant by taking into account that a second peak representing the lower GB inclusions is visible when a bilateral filter is applied (Figure 2 (b)).'

Line 261

'lighter' instead of 'whiter'

Line 263

The meaning of this sentence is not clear. Please revise.

Line 268

Replace 'let' with 'held' or 'subjected to' or similar.

Line 271

Please be more specific about the significance of this! How significant was the pressure loss? What is the time scale, etc.?

Line 271

Why is there no significant impact? What leads to this statement? If this is based on the results from this study, it should be moved to the discussion.

Line 282

Please clarify: is the explanation self-sealing followed by precipitation or are both explanations independent from each other?

Line 284

See comment Line 202: a better start of this sentence would be: Prakash et al. (2022) ...

Line 295

Elaborate – what is the basis for this statement?

Line 297

minimum instead of min.

Line 309

Please clarify – does this refer to the initial sample?

Line 337

Citation style: These results confirm the findings of Minardi et al. (2021) from carbonate rich Opalinus Clay shale.

Line 338

Please clarify if these are results from this study or from Minardi et al. (2021).

Line 349

... are applied. CO₂ is introduced into the PEEKcell when the P-T conditions are stable.

Line 351

... 56 days (scan 03) of CO₂ exposure. A final scan is performed after the release of pressure and temperature (scan 04).

Line 359

These ... with an initial maximum aperture of ...

Line 362

Name the middle slice in the figure to make it easier for the reader to connect what you describe to what it is you see. In this instance, it is not clear at once what part of the figure is referred to.

Line 376

. A distinct pattern is absent in the rest of the material, but the calculated volumetric ...

Line 384

Consider regular, not italic

Line 387

... suggests that the sample is not completely saturated.

Line 390

... at full saturation, which means a decrease of free water and increase in CO₂. Is this correct? Please be more specific here

Line 395

... has been discussed little in the ...

Line 395

Add references

Line 403

Whose calculation?

Line 410

Please elaborate what mechanisms may be active and discuss why these mechanisms.

Line 417

Consider deleting the first sentence of this section. Start with:

Analysis of the evolution of GV from x-ray images after correction for volumetric strain (Stavropoulou et al., 2020) is used to visualise and quantify the CO₂ penetration into the material.

Line 420

Consider using 'difficult' instead of 'ambitious'. OR say something like the method is not particularly sensitive for the small scale of density variations due to supercritical CO₂ invasion. Also, give details why this statement is made. Are there other studies that show the 'slight density variations'? Are these results from this study? Give more details.

Line 423

This whole section describes a method and should be moved.

Line 430

After Stavropoulou et al. (2020) the attenuation ...

Line 432

Explain $\Delta\mu$ as well.

Line 439

Move the explanation of the legend to the figure caption.

Line 444

Consider using regular instead of italic

Line 448

Please clarify: ... around and in crack locations or either around or in crack locations.

Line 451

... fissures. Therefore, the density will always be decreased in the direct vicinity of the fissures.

Line 452

Please consider: ... of the material increases homogeneously ... otherwise words like 'eventually' and 'relatively' are vague terms that make the statement highly speculative.

Line 461

'the' instead of 'their'

Line 465

For this work... Please mention briefly why the interaction (...) was studied.

Line 476

... of a shaly material ... OR ... of Opalinus Clay shale

Line 477

What are those conditions? Give numbers.

Line 478

... that develop fissures after 9 months of exposure.

Line 486

... as shown in Stavropoulou and Laloui (2022) but do not re-appear 9 months later, after pressure release.

Line 488

Capacity; ..., the x-ray scan after 9 months of CO₂ exposure shows new micro-fissures in the ...

Line 498

It is significant that after CO₂ release....

Line 501

Consider replacing 'sums up' with 'highlights'.

Line 504

Non-destructive

Line 506

Please consider naming the phenomena here to underline the significance of the contribution.

Kind regards,
Johanna Heeb