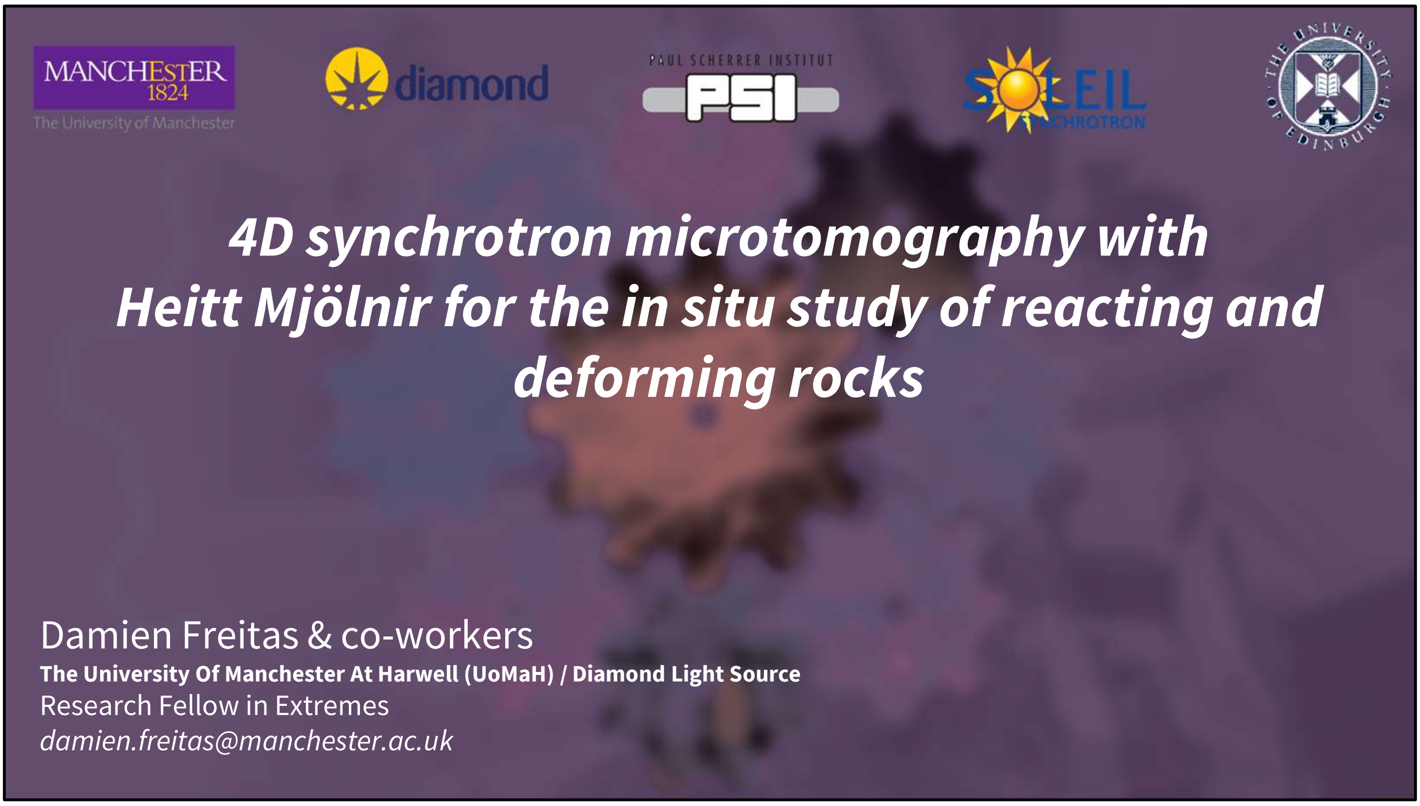


Hello everyone,

The notes commented with these presentation aim to help the reader to follow the progression of the talk and link the various elements presented. For any questions, feel free to contact me at the address provided above.

This presentation will venture into the complex and vast world of 4D synchrotron microtomography (understand here 3D imaging at a relatively fast imaging frequency) by presenting a new tool Heitt Mjolnir (we will come back on the name later on), a device design to study in situ reacting and deforming rocks.

Presentation Outline

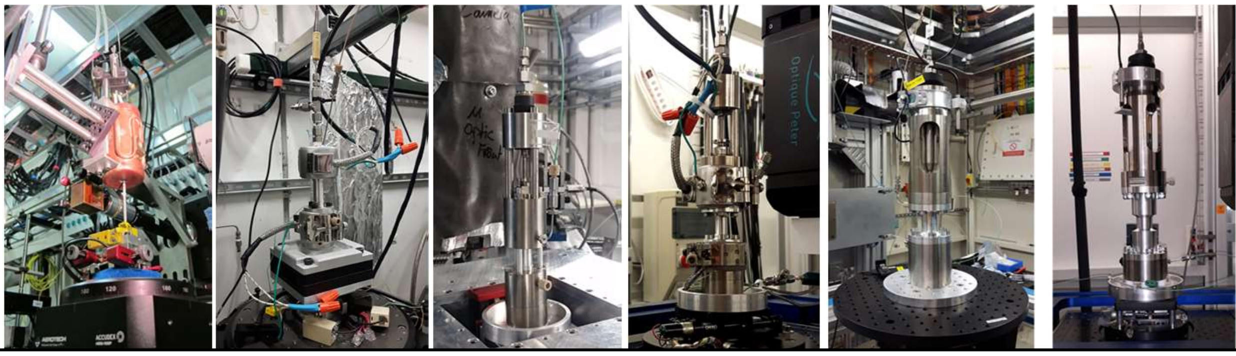
- Who, what and where?
- Context: extreme conditions and *in situ/operando* characterisations
- The Norse arsenal for 4D µCT and Heitt Mjölnir
- Examples of experiments at synchrotron radiation facilities

This presentation will follow a simple progression going from the type of science we are interested in, the numerous coworkers involved and where such research is possible with where rigs are located and facilities to use them.
We will then briefly give some context to justify why we need these new rigs and in situ/operando characterisations.
The main focus of this presentation will be showcasing the rigs, explaining their fundamental principles and their current capacity. Following their design principles, we will review together basic concepts for design of X-ray translucent pressure vessels.
Finally, some examples of experiments at different beamlines will be given to illustrate the versatility of these devices and what they could bring to our research.

Who, what and where?

Development of **open-design rigs** for ***in situ*** **experiments** with

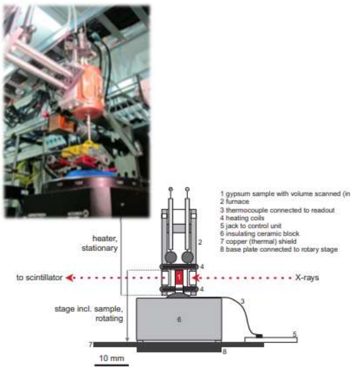
synchrotron X-ray 4D µCT



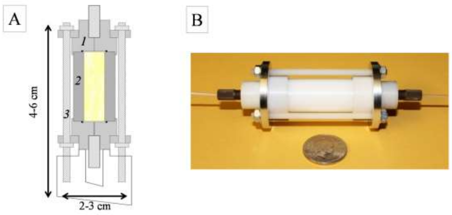
Before acknowledging the coworkers and parties involved in this wide program, I think is a simple definition of what this is all about is first need.
Hence, we will once start with the what.
Heitt Mjolnir is the last member of a large family of rigs developed around principle: building open design and portable rigs for in situ experiments with synchrotron Xray 4D microCT. We will detail the philosophy here after, but the idea is to make new tools for a relatively new technique (the 4D tomography) and to increase the using groups, we aim to have simple and open rig design.

<u>Who</u>, what and <u>where</u>?

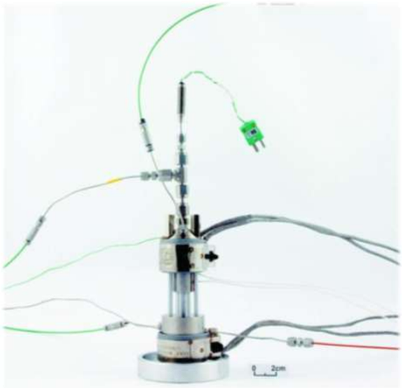
A story started at the APS & UoE (lab CT and synchrotron)



Fusseis et al., 2011 (Solid Earth)

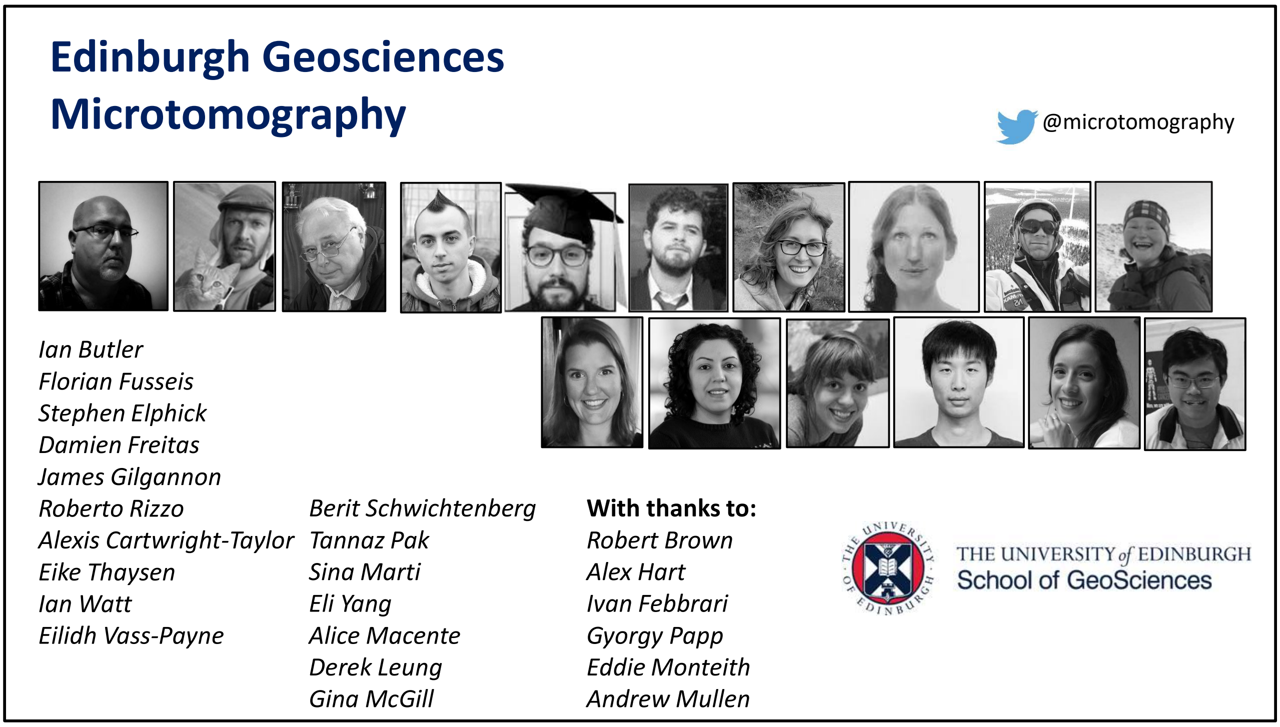


Pak et al., 2015



Fusseis et al., 2014 (JSR)

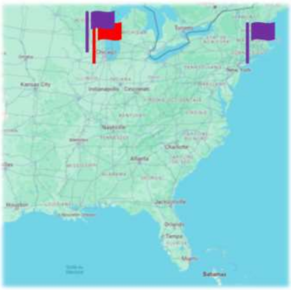
Like every journey, the starting point of "our" group was motivated by a collaboration led by F. Fusseis early 2010 at APS and pioneer experiments at UoE using our open & homemade CT scanner. Joining these two forces, the first experiments were performed on gypsum dehydration, a system that you will hear a lot about today, were done on X-ray transparent furnace (static) and first generation of flow cells.

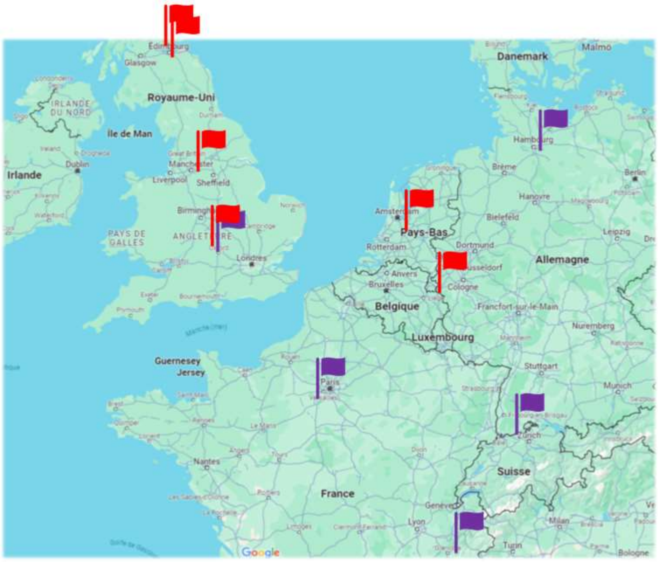


From this first contributions the story significantly expended at the university of Edinburgh. This presentation takes joined effort of a significant group developed around 3 academics Ian Butler, Florian Fusseis and the emeritus professor Stephen Elphick and a vast amount of PGR and PDRA.
The spread of these ECS to new institutions, mostly now having permanent jobs around microCT and development of collaborations is now contributing to the spread of the technology.

Who, what and where?

Now expanding across Europe and the pond…





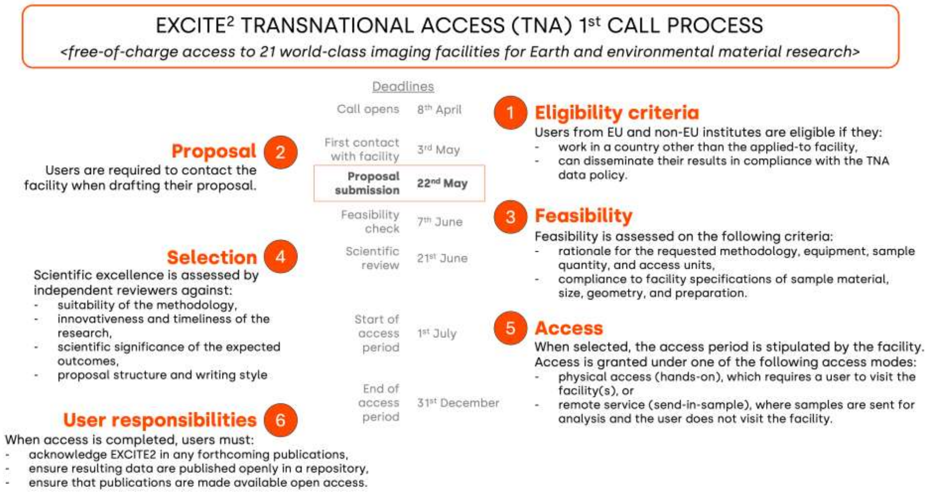
UoE
HWU
UoM (aH)
RWTG
U. Utrecht
U. Chicago/APS

DLS
SLS
SOLEIL
APS
NSLS II
ESRF/ILL
& more

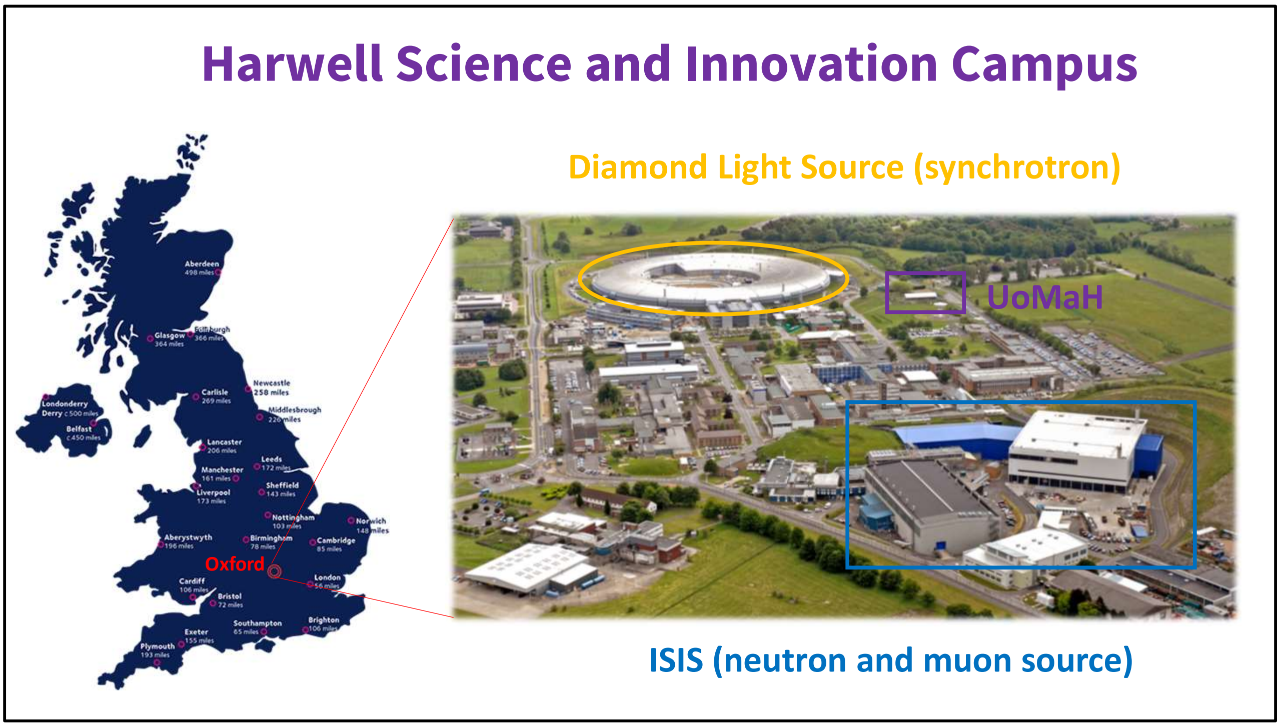
As a consequence, you can now find the devices presented today in several institutes (in red) spreading over Europe and USA and have been now/soon deployed at large number of synchrotron facilities. All of that to say, it is an expanding field, and we are obviously not the only group working on similar setups.

In EXCITE^2 network

https://excite-network.eu/



One great recent news, is that heitt Mjolnir are parts of the Eu funded Excite 2 network. Excite aims to provide on proposal based access to scientists to Cutting edge equipment like SEM, CT scanners etc. all free of charge if selected. I will not venture into the details to much here, because the procedure is being finalised but it can be discussed afterward and I can put you in contact with relevant people if your have any query. For more info, please visit their website!



Now, for slightly more personal aspects. I am now research fellow at the UoMaH, division of Manchester based at Harwell campus. Our mission is to develop and promote via collaborations and support various type of in situ experiments at UK national facilities. Our advantage is being located onsite at I13 beamline and working closely with these facilities for new developments and deployment.

For example, we have a series of in situ furnaces, mechanical testing devices of all kind… often dedicated to work with specific/dangerous materials. These are listed on our website and can be available for projects at radiation facilities.

“Extreme” conditions and *in situ* characterizations

“in high-pressure science, your working conditions are always at the extreme of someone’s else”

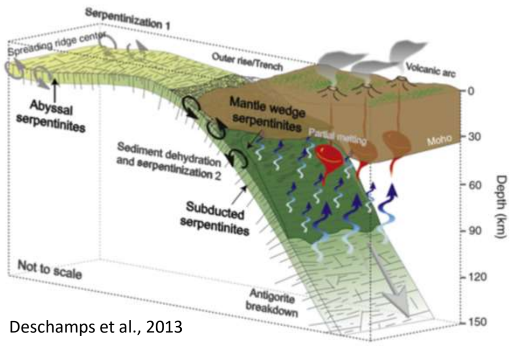
<u>In this presentation, we will only consider the upper crustal conditions</u>

Why?

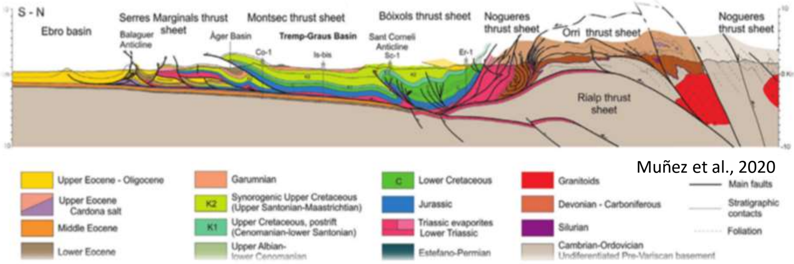
-Geo-resources

-thin-skinned tectonics

-metamorphic reactions



Deschamps et al., 2013



Muñez et al., 2020

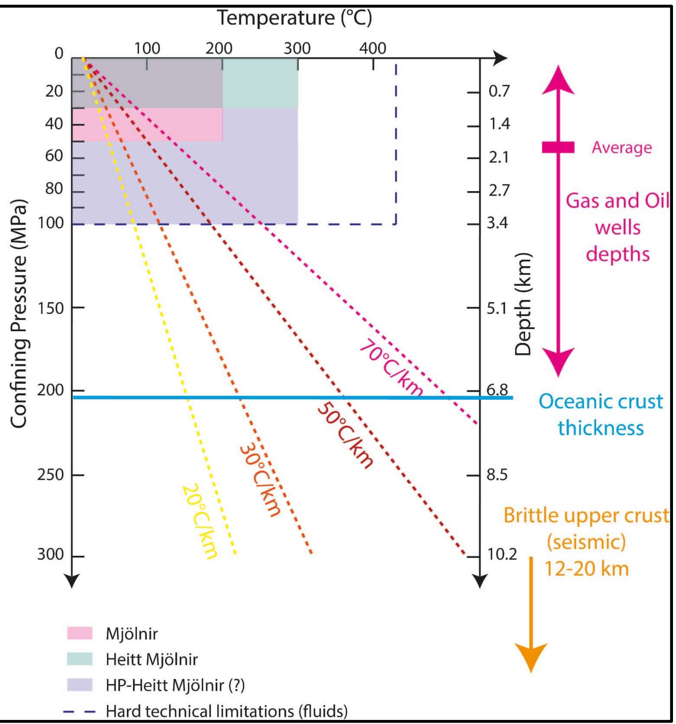
Now that we have seen who, what and where. Lets look at our motivations. We aim to understand the condensed matter at extreme conditions, these extreme having various definitions depending on our scientific interest. Here we will mainly focused on the upper crustal conditions. Why, because as you will see later, we face several constraints limiting our accessible P-T range for microCT. The upper crust is a zone of great interest for all our georesource: oil, gas storge, CSS etc., one important focus of structural geology with thin-skinned tectonics and also lower grade metamorphic reactions.

Crustal/Subsurface's P-T

One pressure-depth path…
but various geothermal gradients !

Craton-Subduction:
20 to 30 K/km

Volcanic (Hydrothermal/ geothermal):
>50 K/km



Lots of interest for geologists are in upper crust, but which conditions are we talking about?
Roughly, in earth crust we have one pressure depth path (obviously depending on rock densities, which aren't hugely varying), for the first kilometres of the crust we talk about 100's of MPa. At the opposite, temperature and geothermal gradients reported are varying in a greater extent from low cratonic and subuction gradients to high volcanic, rift, geothermal one where values can be more than double. This lead to a ideal P-T range we need to cover between 0-200 MPa and T from 0 to >400C.

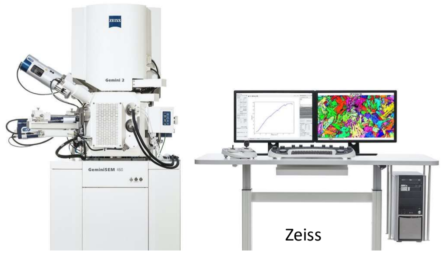
On the necessity of *in situ & operando* characterisations:

-Physical access to **extreme conditions** (subsurface, offshore or space) is **limited and expensive**

-(Geo)physical methods need **calibration**

-Models require **ground truth**

-Classical experiments can't offer **direct visualisation**



Finally, why do we need in situ characterisations?
First, access to extreme conditions remains costly and limited, even for the georesource sector.
Second, our geophysical method requires calibrations: lost of techniques require inversions, such as seismic velocities or EMT, and a ray of possible solutions may arise which can be precised with appropriate parameters measured in situ.
Our models, even sophisticated requires testing and ground truth and going back and forth between the two is necessary.
Finally, most of classical lab experiments can't offer direct visualisation of the transient processes.

Why designing new rigs ?

- Visualisation and quantification of **transient processes**
- **Triaxial devices – flow cells – thermal reactors**
- Commercial options often **not optimised for Geosciences**

1) X-ray transparent/translucent to allow 4D µCT
2) Miniature/portable (whole kit) → from labs to synchrotrons
3) Modular → cater for various experiments
4) Cheap, simple & open → wide community, H&S etc.

Naturally comes our interest of accessing to the visualisation and the quantification of transient geological processes.
We have seen for the context that we want to be able to control finely the pressure and perform deformation via triaxial devices, fluid flow through samples and temperature testing at conditions of the upper crust.
When looking at solutions on the market, the commercial options that are available are often not optimised for geological investigations, and are often driven by a more lucrative materials science/engineering market, and that research may be more interested in tensile testing than in compression, fluid flow etc.
Our objective is then to design rigs that are adapted to the needs of geoscientists. These rigs should be reasonably transparent or translucent (because they alter the transmitted beam) to enable 4D microCT. They should be portable, to be deployed at various light sources. Modular for the various type of process we want to investigate and ideally cheap and simple in order to have impact by reaching multiple communicates.

How to design an X-ray translucent pressure vessel ?

Or the art of the compromise…

1) **Type of experiment and sample targeted**

→ Research question

→ Samples - REV

→ Conditions – variables controlled/fixed

→ Type of characterization needed (3D and/or 4D imaging, XRD, etc.)

→ Duration of investigated processes

Now, I will give a non exhaustive guide for pressure vessel design based on the workflow used to design our rigs. This is more to help understanding what are the constraints used to designed a rig and a reflexion on what matters for their design.

First part of the design, and the most important, is the definition of the research question. This defines the type of experiments, characterisation and the way to find the answers we look for. This defines the sample nature and size necessary for being representative (for example working with granite may require larger samples than a sandstone etc.). The conditions will arise from the research questions and what can be fixed or needs to be varied + the conditions may be dictated as much by the available materials and sealing solutions as by the research question. (hard limits defined by the materials,. the temperature limits for o-rings, or the glass transition of polymers, etc.).

One important thing to consider is also the duration of the process to be investigated. Mechanical loading is imposed, so we can control that rate. Catastrophic failure of a material is emergent, and needs special approaches to control (see Stor Mjolnir), chemical reaction kinetics cannot be controlled, except, perhaps, by raising the temperature. (which might change the mechanism), and in general happen in their own timing.

PV design

Because it is likely we can't have it all… we better pick our limitations

2) Establish the priority list

→ X-ray transparency (spatial & temporal resolution @ given BL)

→ Sample size/nature

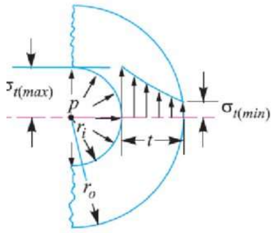
→ Conditions

The second point is then from this from the "wish"/requirements list, establish a priority list. What is important and what can be sacrificed or at least changed with minimal impact on the research question.
Again, it can be discussed for several axes with questions like: do I really need fast scans? Can I use other samples, like a microgranite or fine grained sandstone to reduce my sample size? Do I need to be at the max conditions or having less is still ok?

PV design

3 & 4) Guideline calculations

For cylindrical PV → thick-walled cylinder model

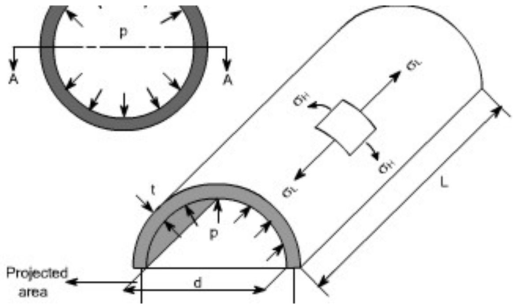


Wibawa et al., 2020

Axial + radial + max

circumferential (hoop) stresses

<< yield strength of the material



Precise + complex shapes → FE modeling

Campbell-Sevey

Then, from these we can start working on the design itself. This is an iterative process between steps 3 and 4.
The design of the pressure vessel is first dictated by the geometry of the vessel. For uCT and cylindrical samples, thick walled cylinder model works reasonably well. One rule of thumb is to add the axial, radial and hoop stresses. Via taking the max hoop stress (e.g at the inner surface of the vessel), the sum of the stresses should fall below half of the yield strength of the vessel material.

This is a rule of thumb, and for precise estimates, complex vessel shapes Finite element modelling may be required.

PV design

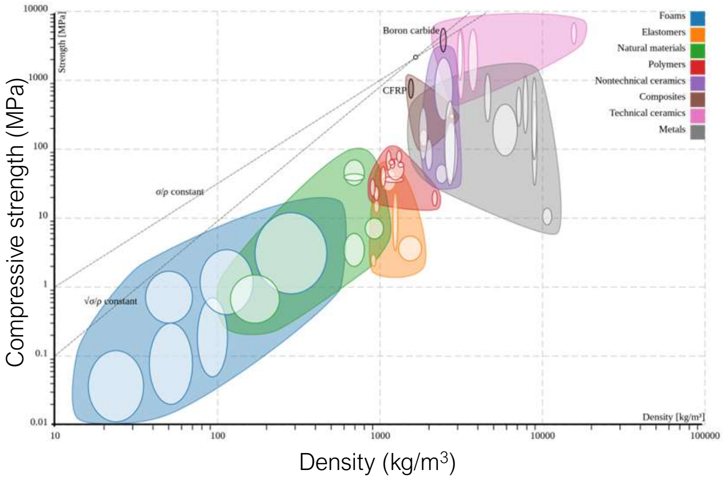
3 & 4) Choice of the material(s)

Compromise between strength (s)

(max P & T conditions)

and density

(X-ray transparency)



Example of Ashby chart (wikipedia)

As stated earlier, the pressure vessel material is a key component and the material selection for its make is critical. It should be selected based on a compromise between strength (e.g the mechanical properties desired) and the density of the material, which directly translates into x-ray attenuation. Using Ashby charts like the one presented, can help selecting family of materials for the most interesting grades and within the family of materials.

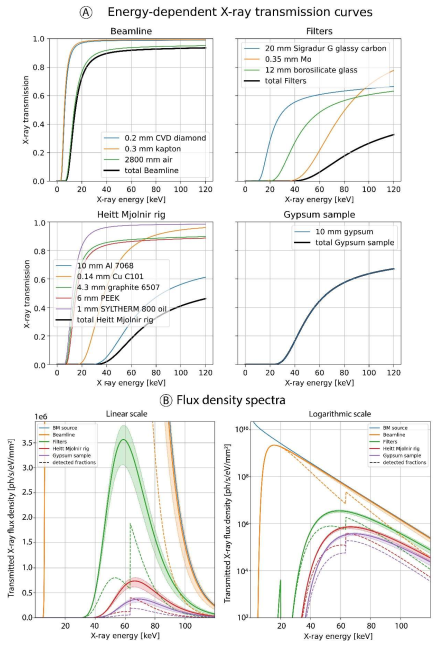
PV design

5) Beam transmission simulations

→ Beamline specific:

Beam type, energy, optics, filters, rigs and detector…

→ Key to define experimental plans (scan frequency for 4D imaging)



Freitas et al., 2024 JSR

For estimating the beam transmission through the rig, different type of simulations can be done, like simple calculations using beer Lambert law with material attenuation coefficient. We have recently implemented a code, using a bit more information with the beamline setup, energy optics and filters. This allows precise simulation of the effect of all the rig components and simulate for each beamline what is the imaging quality and the frequency of imaging could be.

PV design

6) Be H&S compliant

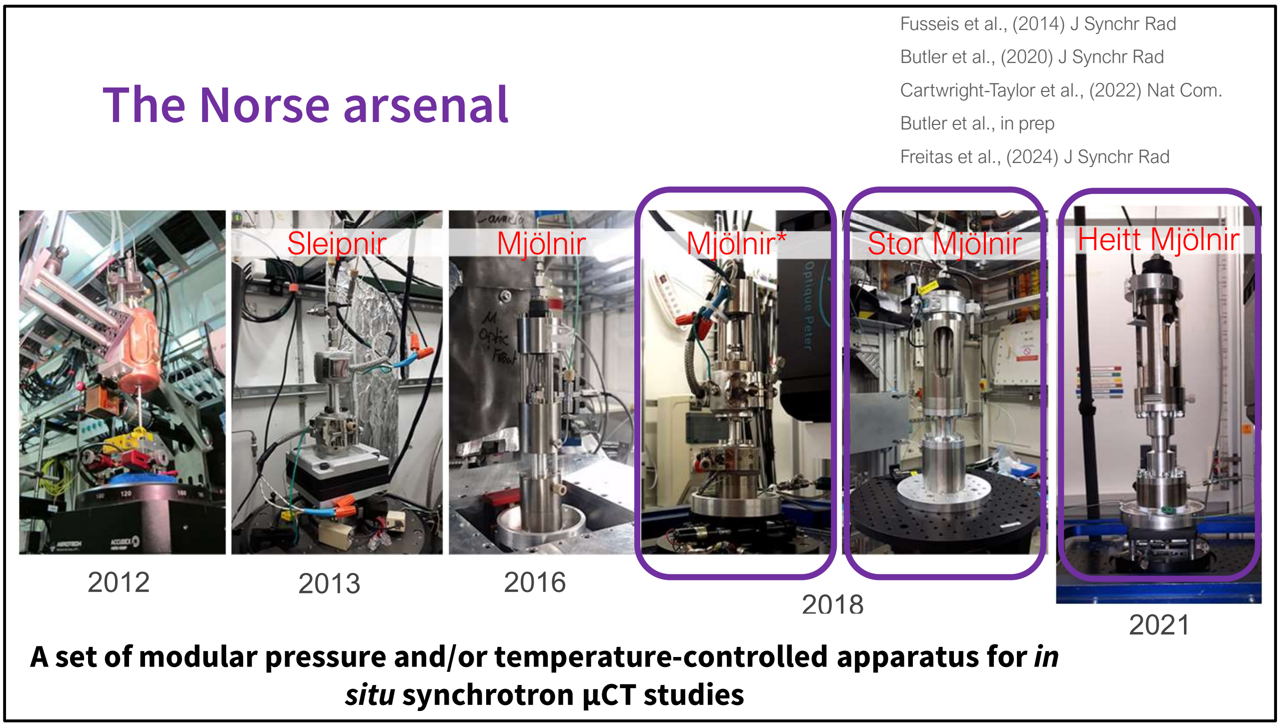
→ UK PSSR 2000 provides comprehensive guidelines

→ A rig deployed at different institutions may face different procedures

7) Make a dummy and safe testing above operational limits

→ Adjust and refine the design (iterations from 3-7)

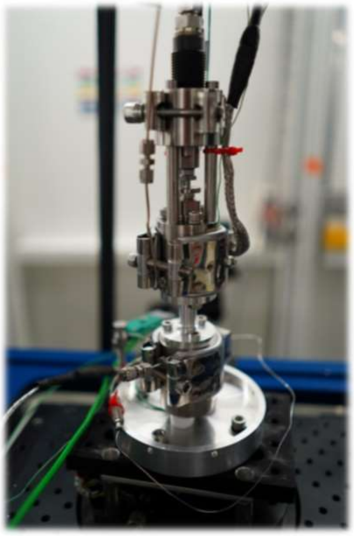
Never forget to be Health and safety compliant and think about where you want to deploy the vessel. Some national regulation give comprehensive guides on what the procedure should be even if these rigs fall below the pressure system thresholds. And finally, nothing replace building a dummy rig and testing above designed range, all of that in a safe way. From that testing phase, all flaws of design may be adjusted and refined. Drawing must be then updated and re-iterations of the former steps should be taken.



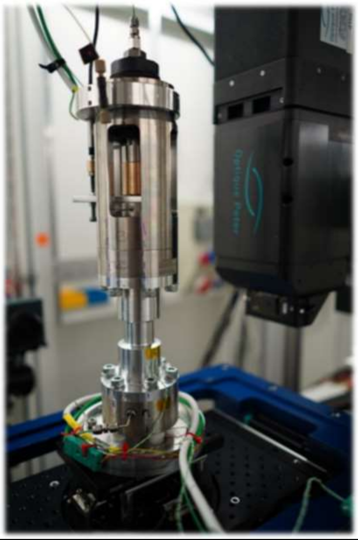
Now that we have seen, the design principles. We will detail how the rigs works. Today we will focus on the triaxial rig suites from our group. The Norse arsenal name comes from Nordic mythology: what is better than an hammer to break some rocks? Most of these rig design are now published or soon to be in open access journals and you can refer to them for any technical details.

How do they work?

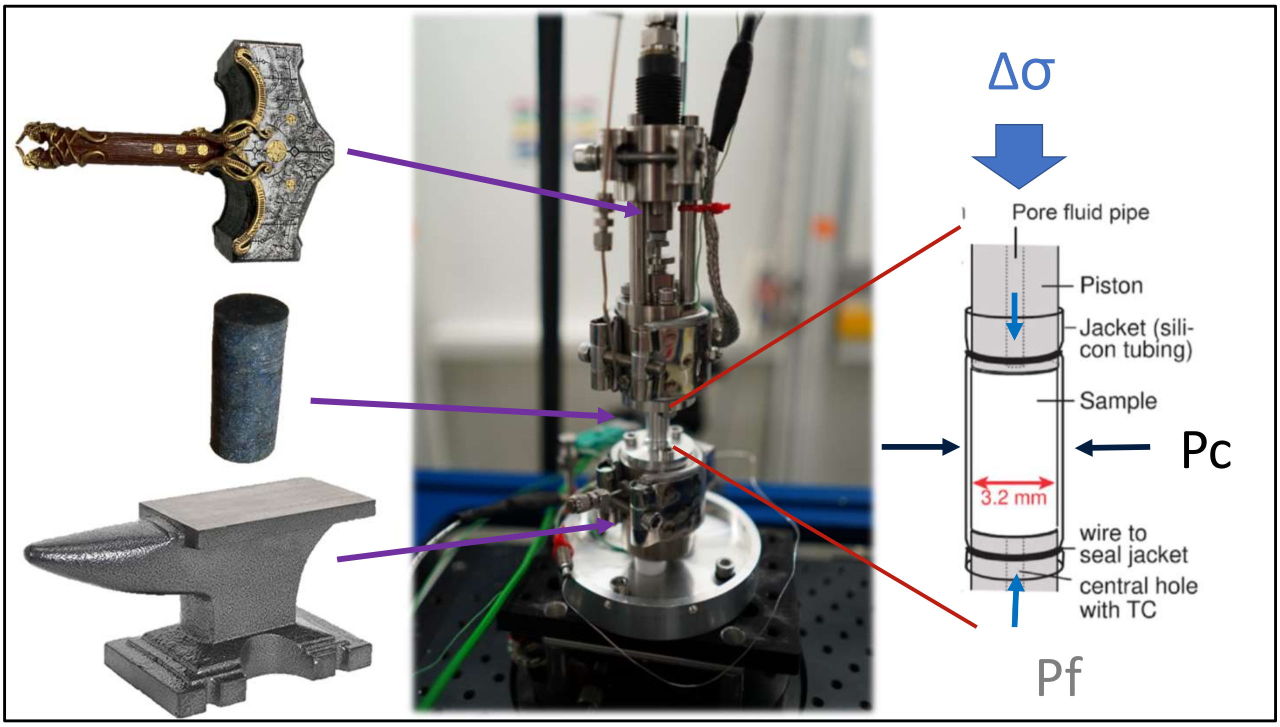
Mjölnir



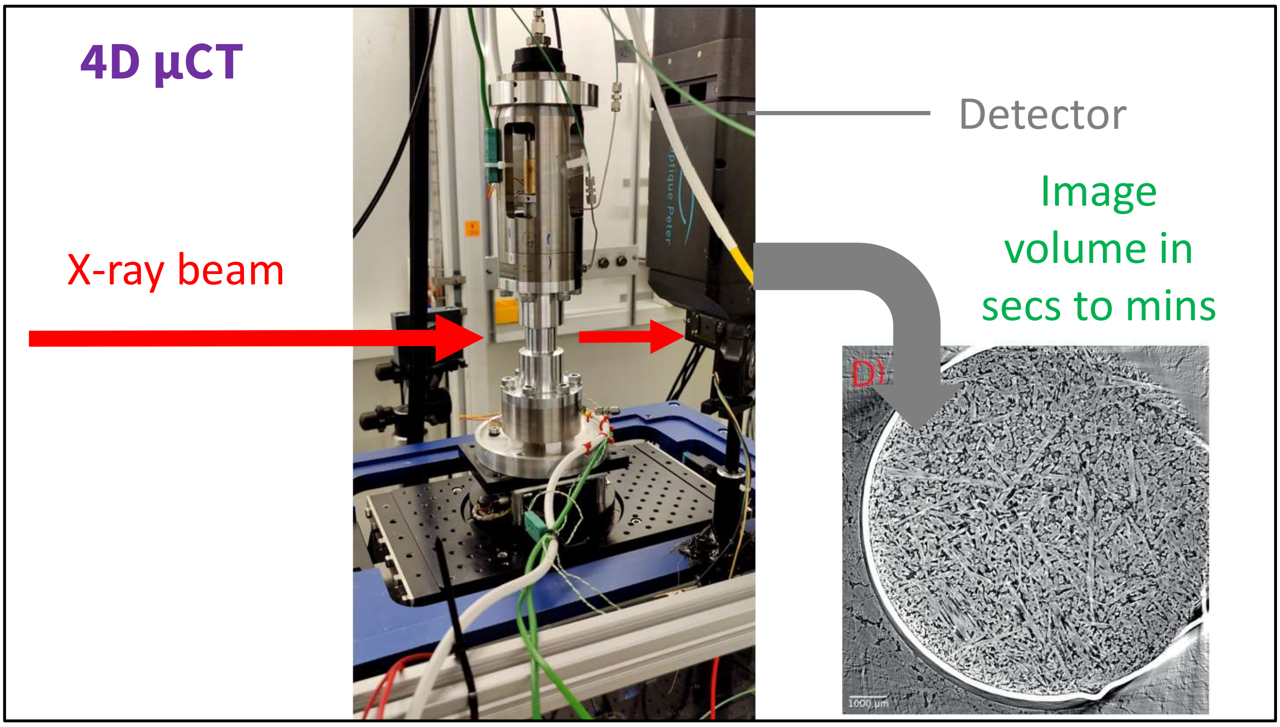
Heitt Mjölnir



Lets now review together some fundamental aspect of triaxial rigs. Mjolnir and Heitt Mjolnir are triaxial rigs build on the same principle. They are hydraulic based apparatus, but unlike the furby example here described. The feature confining pressure which prevent the extrusion of the material.



The base of the rig, is fixed to the rotation plate: it is the anvil on which the sample is placed. The sample site in the pressure, vessel, the thinnest part of the rig in which the beam passes through. The tope of the rig is the dynamic part, which will allow via the hydraulic actuator to apply the additional axial load.
To sum up, we have the radial/confining pressure applied via an hydraulic fluid. The sample is isolated from this fluid via a jacket. The differential stress is applied via the hydraulic actuator on the top side and via using pistons with holes, we can access the sample volume with fluids, to control the pore fluid pressure.



The setup for the micro CT, if the following. The rig is small enough (a few kilos) be on the beamline rotary table. The beam passes through the full rig in its finest part of the PV and the detector is located on the other side. Via rotation of the stage and accumulation of the projection, we can reconstruct 3D volumes. At the synchrotron the flux and energy of the beam allow this process to be performed in seconds to minutes.

Design process

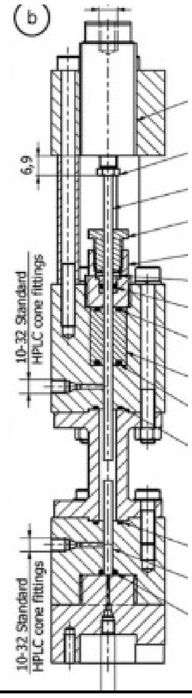
1) Objectives:

Rock deformation + T reactions (<300°C)

2) Priority list:

- Miniature (low weight)
- Fast tomo (s/mins) on 3rd gen Synchrotrons
- Samples up to granite → $\Delta\sigma > 500$ MPa
- Temperature

→ Compromise (sample size, max P, max T)



If we take the design method detailed earlier and go through the design process for these rigs: the objective was to design rock deformation rigs with temperature capacity to also perform reactions.
The priority list was constrained with size, to notably on the table of tomcat beamline <6 kg. Tomography were set to be fast with Mjolnir (s) and relatively fast for heitt Mjolnir (min). The differential stress had to be greater the 500 MPa to be able to break rocks up to granite. Temperature was also an important priority.
The compromise for this design was clear limitation on sample size especially for Mjolnir (very light rig) and maximum P-T we could reach.

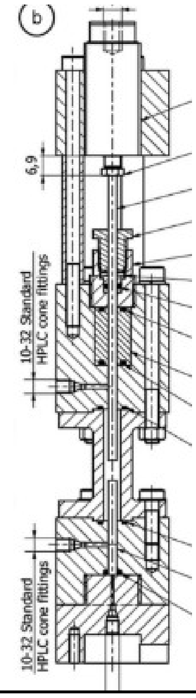
Design process

3 & 4) Calculations for aimed conditions + material choice

PV: Total stress 157 MPa << YS Al (6061) @150C

Outside beam path: titanium or steel

LT: elastomeric jacket / HT: metal jacket



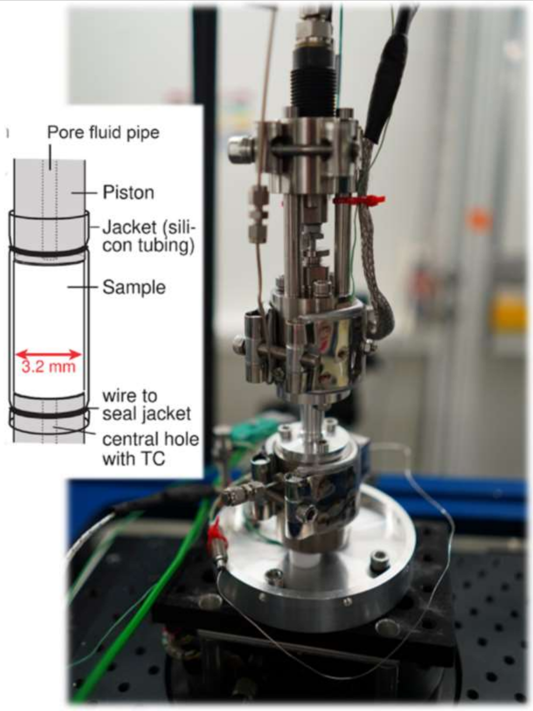
For Mjolnir calculations round showed that with 3.2 mm diameter sample total stress while at maximum operating conditions we well compatible with an aluminium pressure vessel and for temperature up to 150C. Aluminium offers a decent strength for light weight, however it is sensitive to temperature.
Materials outside the beam were mainly titanium (light) and jacketing options were elastomeric jacket for LT while metal jacket have to be considered for t>150C.

Mjölnir

Mjölnir specs:

Samples = Φ3.2 X 10 mm
Pc = 50 MPa
Pf = 50 MPa
Δσ up to 550 MPa
T < 150°C
+ LVDT

Rock deformation
LT reactions



A quick sum up on Mjolnir. The rig is mainly dedicated to rock deformation and LT reactions.

Stor Mjölnir

Stor Mjölnir specs:

Samples = Φ10 X 25 mm
Pc = 50 MPa
Δσ up to 510 MPa
+ LVDT
+ 2 Acoustic emission (AE) sensors
+control on deformation based on AE rate + stroke (LVDT)

Rock deformation

→ More acoustic sensors coming (Butler et al., UoE)

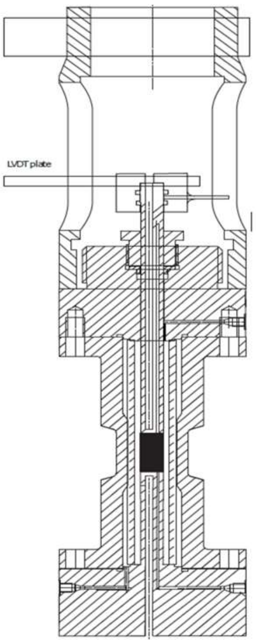
From that design, we have built storr Mjolnir. A rig designed to operate at similar conditions with a larger sample. The temperature capacity has been sacrificed to implement acoustic emission sensors and a capacity to control deformation based on AE rate.

Why designing Heitt Mjölnir ?

Mjölnir is a reliable, cheap and easy-to-operate device, but its simplicity comes with **some limitations**:

- **Sample size** is limited: Φ3.2*10 mm (for some rocks, we need a larger volume to be **representative**)
- **Temperature** is limited to 150°C
- Other designs are either 1) own privately, 2) not fully open, 3) not portable, 4) not H&S friendly, 5) difficult to adapt etc.

From that, this was already a good platform but we realized Mjolnir, despite being a very easy rig to work with, had some limitations which were interesting to overcome. The interest to gain in sample volume size and temperature to access more of the crustal conditions motivated the design of a new rig. This has also been mostly motivated by the absence of other design on the market to do what we exactly aimed at with several limitations.

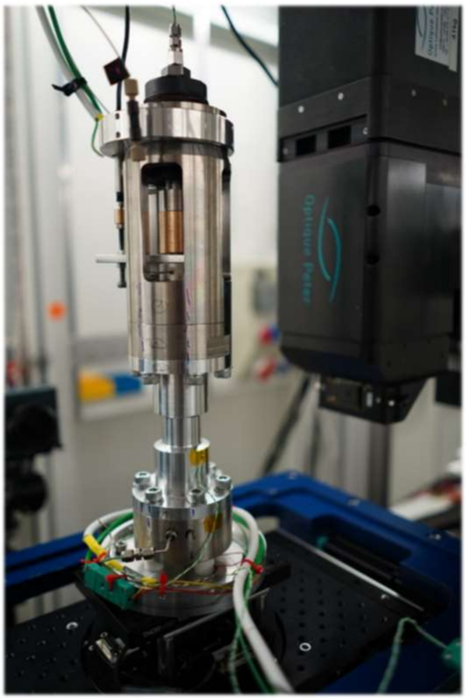


Heitt Mjölnir

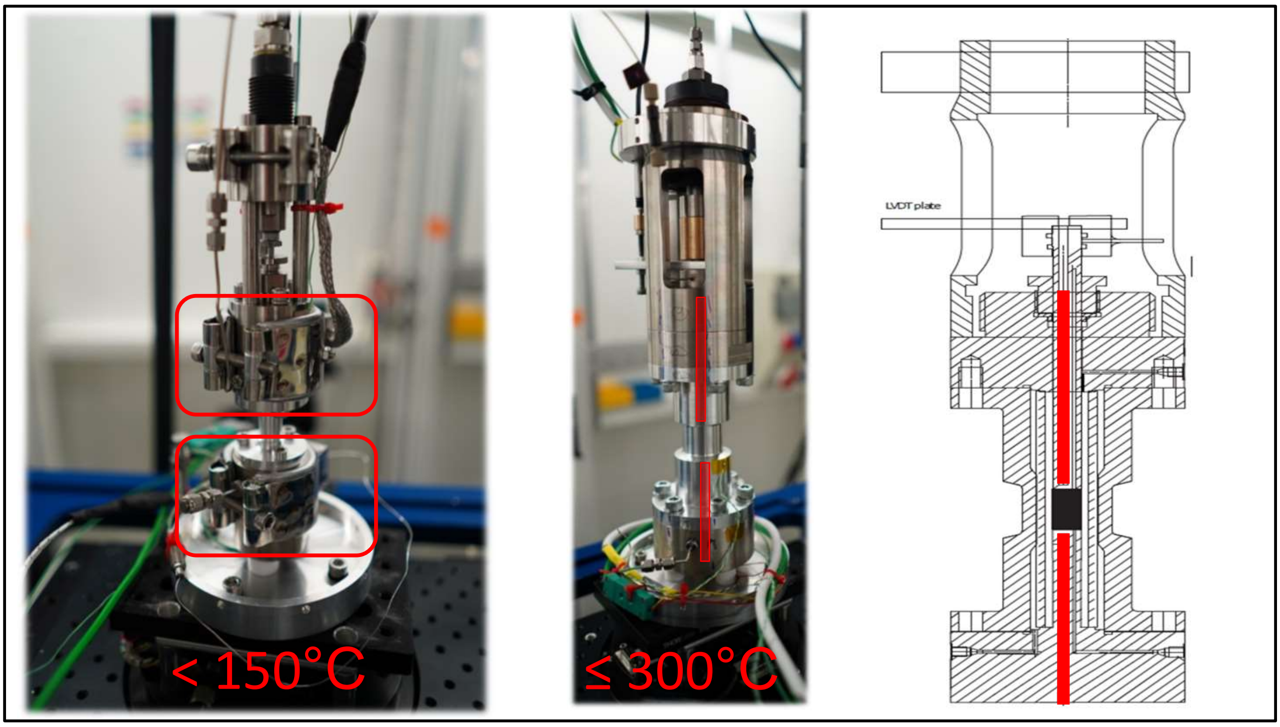
<u>Heitt Mjölnir specs:</u>

Samples = Φ10 X 20 mm
Pc = 30 MPa
Pf = 30 MPa
Δσ up to 510 MPa
$\boxed{T < 300\ ^{\circ}C}$ + low thermal gradient
+ LVDT

Metamorphic reactions
Hydro/geothermal reactions
→ Improvements in P capacity coming

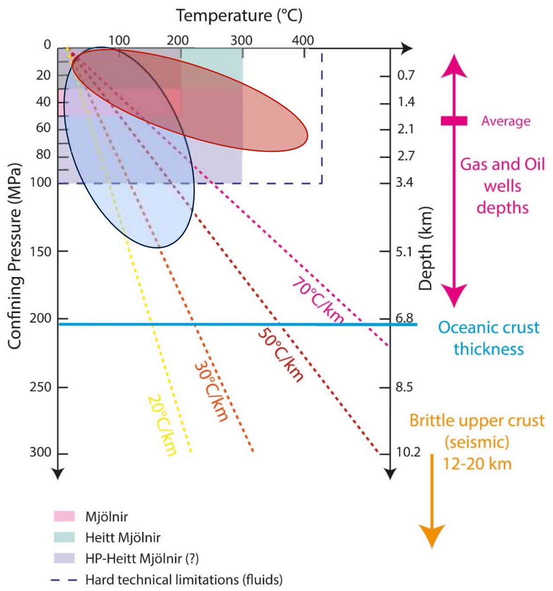


Heitt Mjolnir can now operated with 10*20mm samples. At conditions close to Mjolnir but doubling the temperature capacity. This makes it very much adapted to low grade metamorphic reaction study and various amount of fluid/rock interactions of the upper crust.



The whole difficulty of the design process for this rig was shifting the heating system for external to internal. This is due to the limitation imposed by the aluminium pressure vessel being very sensitive to temperature.

Capacities and relevance



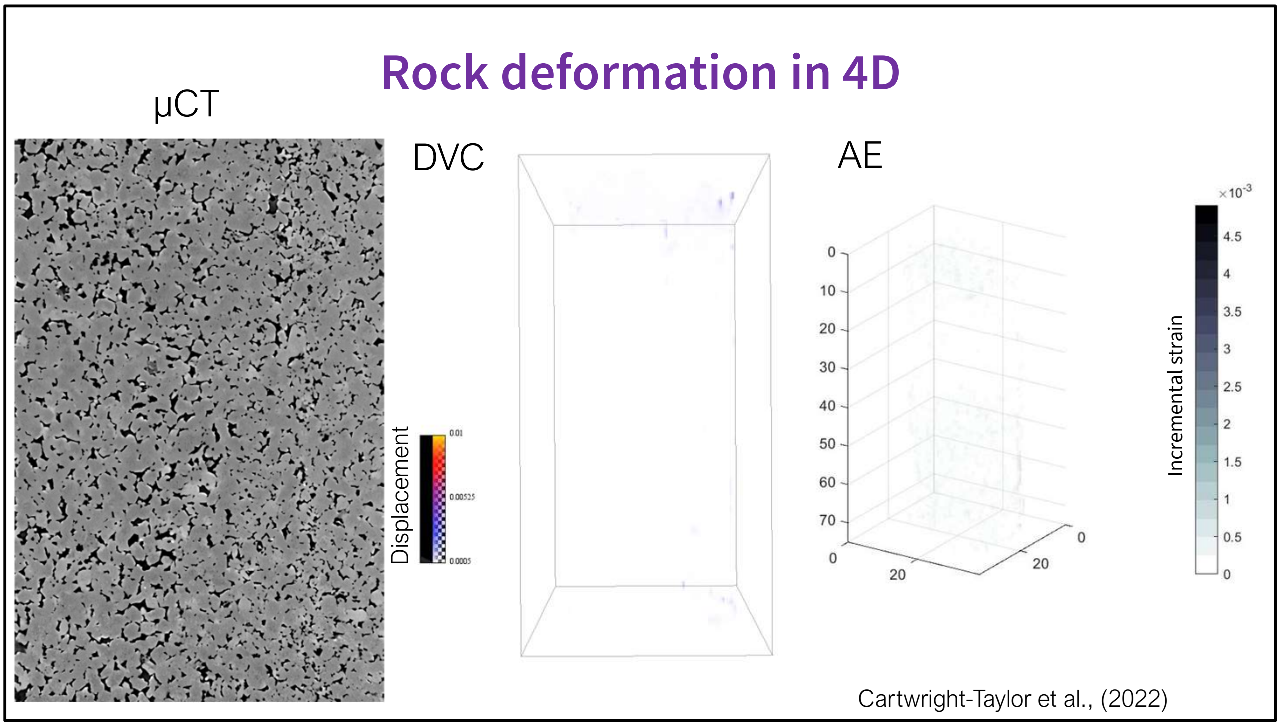
	Mjölnir	Heitt Mjölnir
PC (MPa)	50	30
Pf (MPa)	50	30
Δσ (MPa)	550	510
T (°C)	<150	≤300
Sample size (Φ*h mm)	3.2*10	10*20

Sum up of the capacities of these rigs. Mjolnir style apparatus are adapted for the high pressure Low Temperature, low geothermal gradient context, while Heitt Mjolnir is more adapted to shallower and hotter context. This make these tools, with stor Mjolnir complementary and their ongoing improvement will offer more and more advances features in a near future.

Examples of applications

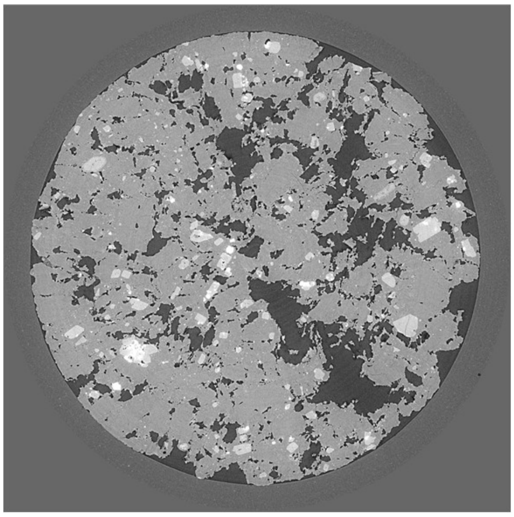
TOMCAT @ Swiss Light Source	I12- JEEP @ Diamond Light Source	PSICHÉ @ SOLEIL
		

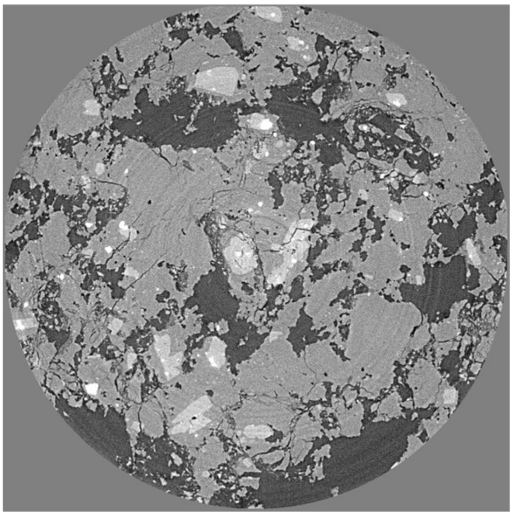
Now that we have seen the rig, I would like to finish this presentation with some example of applications we have been working on in the last years.



Brittle deformation of sandstone with AE recording and deformation control to visualize failure with unprecedented resolution. Quantification of the displacement can be achieved via using digital volume correlation techniques see Stamati et al. presentations.

Rock deformation in 4D

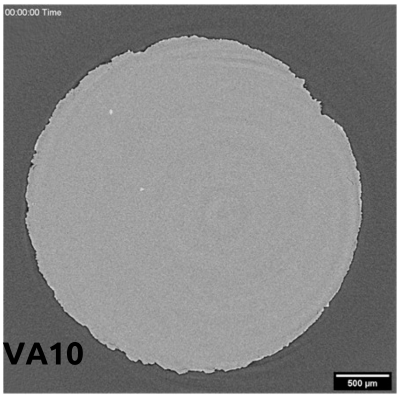


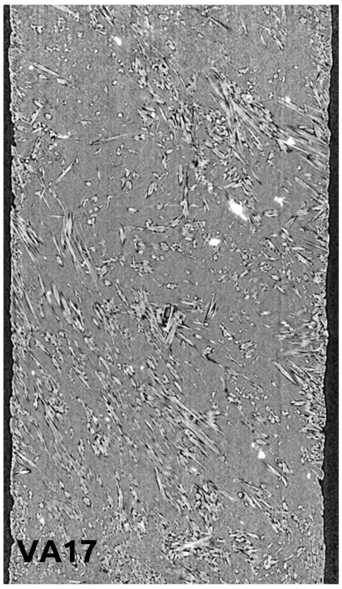


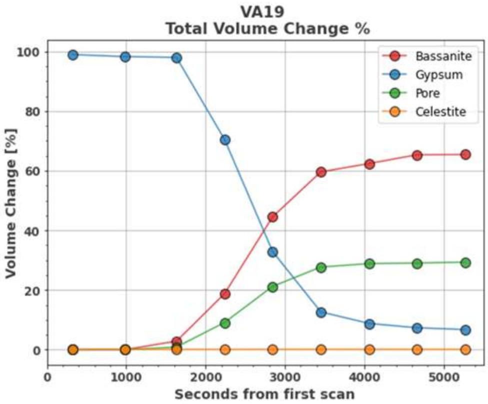
Freitas, Rizzo, Heap et al., in prep

Another very recent example, may 2024 at soleil synchrotron, is the study of compaction bands, plastic features in volcanic rocks.

Metamorphic reactions with differential stress





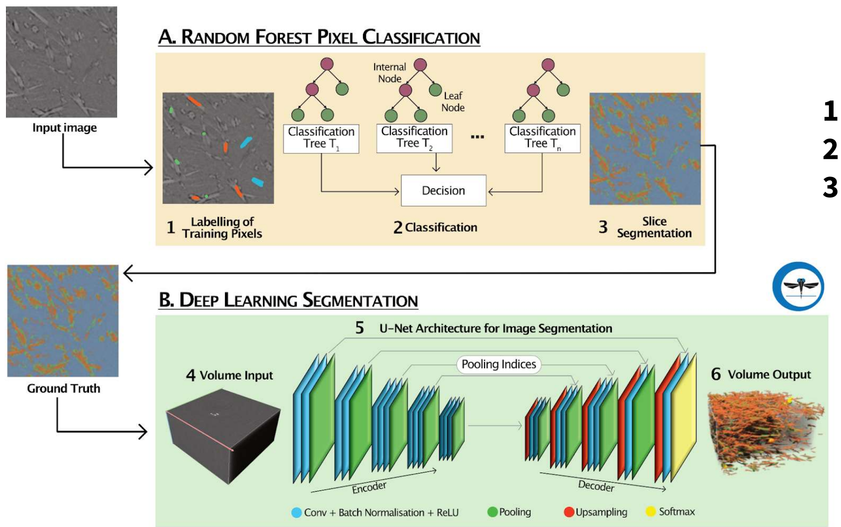


Gypsum → Bassanite + H_2O (pores)

Gilgannon et al., (2023) Geology

On the reaction with deformation, we have performed numerous studies around gypsum dehydration and the influence of stress state on the reaction fabrics. From these data, we have worked on new processing techniques able to segment accurately the large 4D data sets to produce the reaction curves here presented.

Processing of large 4D datasets

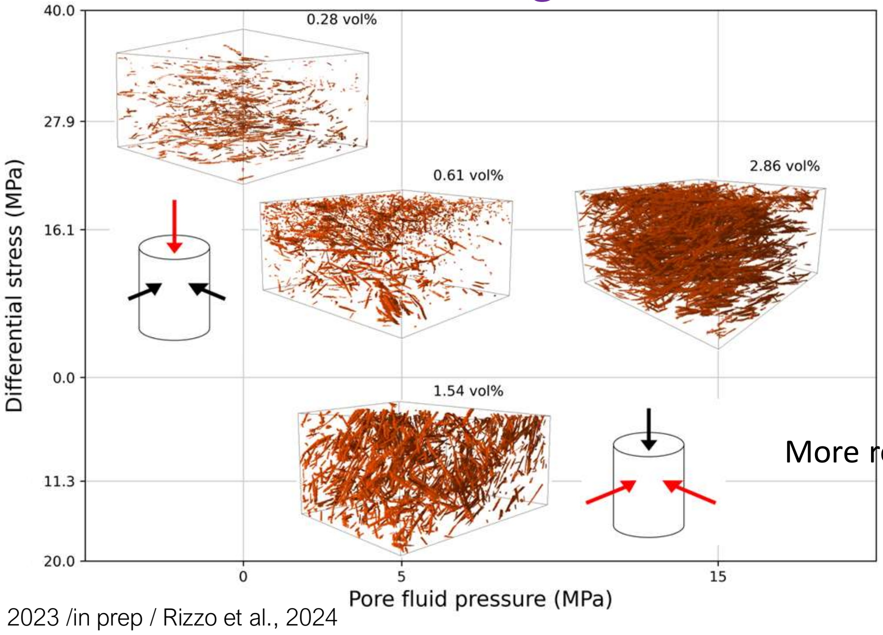


1. **Computational cost**
2. **Consistency**
3. **Accuracy**

Rizzo et al., 2024 Solid Earth

For this, we used a combination of deep learning and machine learning techniques. This allows using extensively the data sets at a moderate computational cost. It helps maintaining and checking the consistency and accuracy of our segmentation by benchmarking on the chemical balance of the system. For more information please refer to our very recent paper on the matter.

4D evolution of reacting rock textures

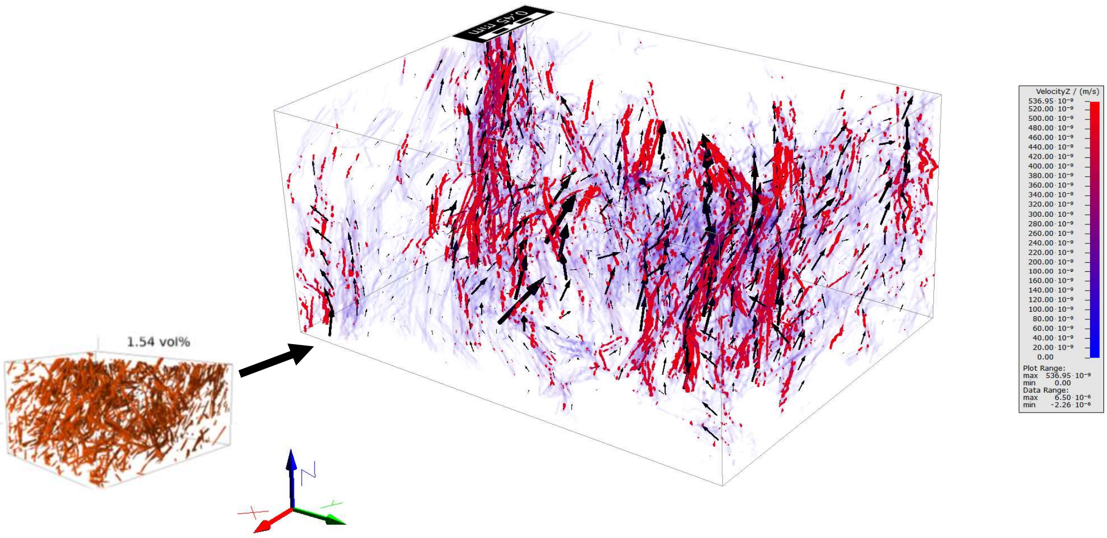


More results to come!

Gilgannon et al., 2023 /in prep / Rizzo et al., 2024

With these processing method we were able to investigate more systematically the reaction fabrics formation during metamorphic and dehydration reactions.

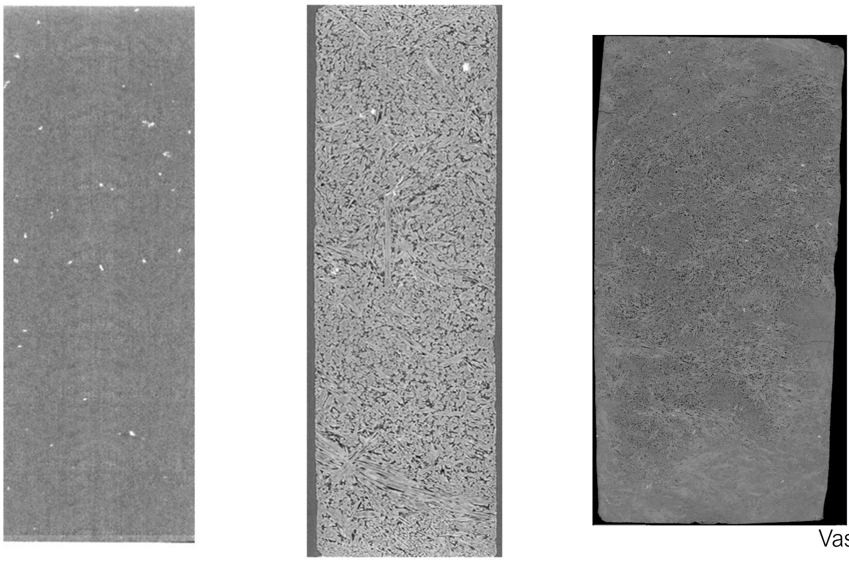
Fluid velocity modelling/ Permeability



Gilgannon et al., in prep

From segmented volume, we can now incorporate the data into digital rock physics models to for example, vusualise and model the fluid velocity and the permeability with the emergent rock texture. As you can imagine, creating texture that are anisotropic also generate oriented fluid channels and anisotropic permeability.

Dehydration and rehydration

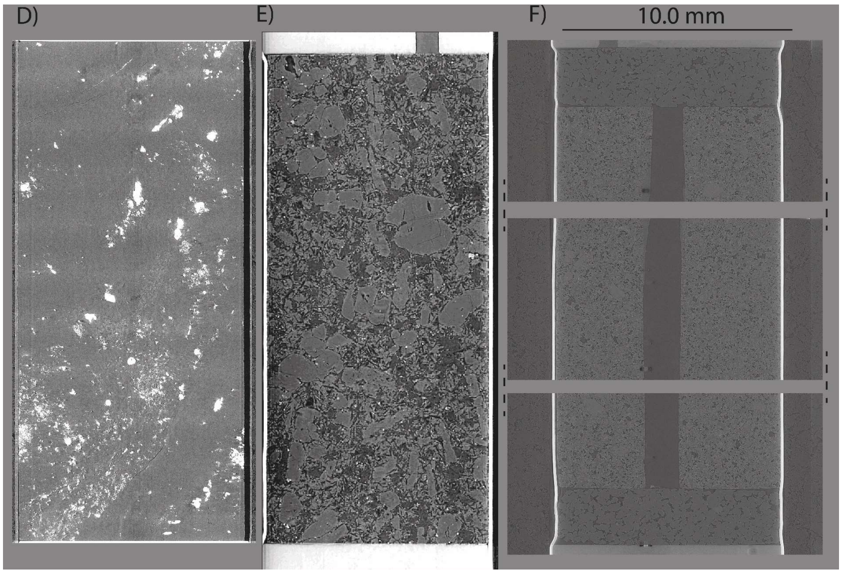


Freitas et al., 2024

Vass Payne et al., in prep

We have also started preforming dehydration and rehydration cycling into gypsum basanite to hydrite system

Fluid-rock interaction



Freitas et al. 2024
Watt et al. in prep

And finally, we are now starting to perform various types of fluid rock interactions, more in a thermal reactor style, under pressure. To investigate carbonation of volcanic rocks for example.

Access route -rigs

- Accessing an existing kit ?

→ **Collaborations and EXCITE^2**

- Replicating them?

→ All technical drawings, instructions & kit's specifications in published materials (Butler et al., 2020 and Freitas et al., 2024 @ JSR).

→ Experience and practice are mandatory requirements

→ **We are happy to answer questions**

I hope this gives you some ideas and illustrates the potential of these rigs. If you are interested to use them for any project , we are obviously open for collaborations and Excite2 will aim in this direction.
If you would like to replicate them, the design and all the details are published in the JSr papers, except storr Mjolnir which is in prep, for now. I just remind everyone to pay strong attention to safety aspects using bespoke devices and we are happy to answer any questions.

Access route – synchrotron radiation facilities

Proposals:

- **2 calls/year**
- Target **the good beamline**/light source
- 2 pages: background, **proposed research, technical feasibility/beamline setup**
- Awarded beamtime (max 6 days) is free, travel/accommodation support can be obtained

I just made a slide, for the colleagues which aren't familiar to synchrotron beamtime facilities.
The beamtime is attributed by calls and selected on proposal. They are usually targeted for a beamline with s defined research project. On the proposal you need to demonstrate the interest of the scientific question and as well the technical feasibility /capacity to carry on the proposed research.

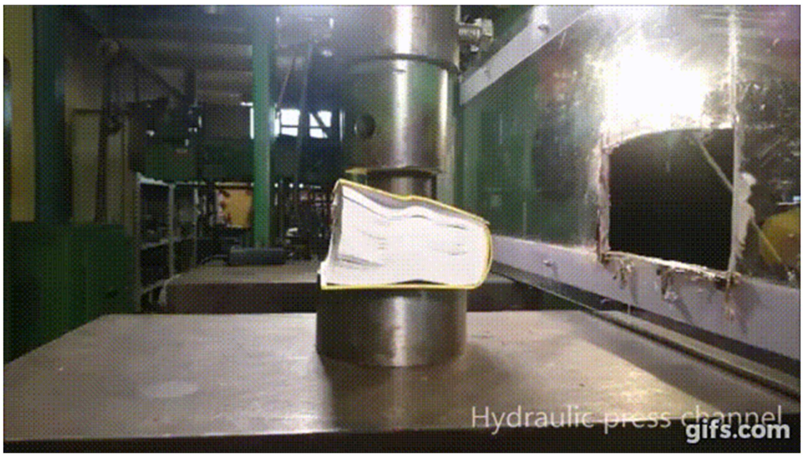
Versatile rigs for the rock deformation community

		Measurement	Classical Black Box Experiments	Post Mortem Evaluation	4D In-Situ Imaging Experiments
Sample scale		Bulk strain evolution	✓		✓
		Bulk stress evolution	✓		✓
		Acoustic emissions	✓		✓[1]
		Permeability evolution	✓ (in some experiments)		✓[3]
Grain scale	For all mineral grains and pores: **full 4D evolution** (space and time)	Proportion[2]		(✓)	✓
		Spatial distribution		(✓)	✓
		Percolation[3]		(✓)	✓
		Size[2]		(✓)	✓
		Geometry[3]		(✓)	✓
		Orientation		(✓)	✓
		Local strain[5]			✓
		Local strain rate[5]			✓

Fusseis et al.,

I hope to have convinced you, that 4D imaging experiments allow a vast access to information happening during transient processes and these are able to bridge up scales in our field.

Thanks for your attention,
& feel free to ask any questions now or later!



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