



# Sealing, Healing and Fluid Flow in Clay Rocks: Insights on Episodic Flow Events in Fault Zones

Ville Nenoen, Pierre Dick, Juuso Sammaljärvi, Bo Johansson, Mikko Voutilainen, Marja Siitari-Kauppi

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Auteurs dans l'ordre de la publication :

**1**, NENOEN, Ville

Univ Helsinki, Dept Chem, FI-00014 Helsinki, Finland, Finlande

**2**, DICK, Pierre, REFERENT , CORRESPONDING AUTHOR

Institut de radioprotection et sûreté nucléaire, PSE-ENV/SEDRE/LETIS, Fontenay aux Roses, France

**3**, SAMMALJÄRVI, Juuso

University Helsinki, France

**4**, JOHANSSON, Bo

Geological Survey of Finland, P.O. Box 96, 02151, Espoo, Finland, Finlande

**5**, VOUTILAINEN, Mikko

Université de Jyväskylä, Department of physics, Jyväskylä, Finlande

**6**, SIITARI-KAUPPI, Marja

University Helsinki, France

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**Résumé d'auteur :** The porosity distribution and mineralogical changes in a clay-rich fault core from the Tournemire underground research laboratory are analyzed to determine the mechano-chemical processes in a small-scale vertical strike-slip fault. The results display significant spatial variations in porosity and mineralogy along different gouge zones due to a polyphased tectonic history combined with complex paleo-fluid migrations. Porosity values increase from the center of the gouges to their borders indicating diffusive sealing/healing effects and past hydrothermal activities. The healing and thus the strengthening of the fault is marked by an increase of calcium content, which is concurrent with lower porosities around the gouge zone. Chemical mapping in the gouges reveal clay alteration, iron zonality and the presence of zinc sulphide as well as barium sulphate inside the gouge, further suggesting past hydrothermal activity. Finally, even though the observed porosity variations only occur in subcentimeter-thick gouge bands, the higher porosity sections are pathways for fluid flow during fault activity.

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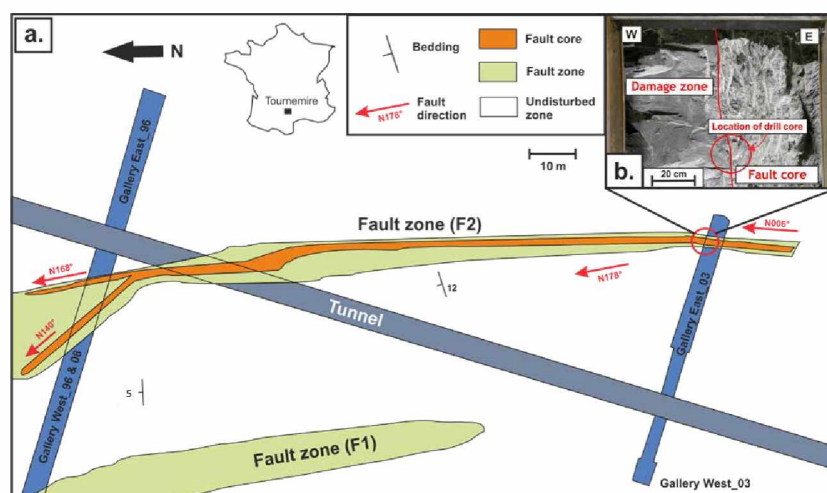
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## Introduction

Sedimentary clay rocks are being considered in various countries as a suitable host rock for the deep geological disposal of radioactive waste due to their favourable properties such as low permeability and high sorption capacity for radionuclides. However, faults and fractures in these clay rock formations can compromise their seal effect by acting as a preferential flow paths for fluid through the otherwise impermeable structure. Therefore, understanding the behaviour and fault-sealing properties of faults is essential in order to ensure the long-term safety of a deep geological repository.

Geological and geophysical observations have revealed that fault zones are typically heterogeneous and variable over short distances and that their fault sealing assessment must involve a quantitative appraisal of the petrophysical properties of fault rocks. Moreover, fault rock permeability is related to the microstructures within the fault zone, which in turn reflects the deformation mechanisms of dynamic recrystallization operative under different faulting conditions. Grain scale deformation in clay rich formations is generally dominated by pressure solution mechanisms, grain-scale fracturing, resulting in cataclastic fabrics along shear zones and an increase of porosity within the damaged fabric due to the dissolution of carbonates or a combination of these processes, depending upon their environment (stress level, temperature). Fault seal analysis thus involves assessing the distribution of such fabrics over individual faults and estimating their associated properties.

The aim of this study is to obtain a better understanding on spatial and temporal changes in fault gouge development and their implication to fluid flow within a clay rich formation. Results are presented from microstructural studies of natural samples collected from the Tournemire Underground Research Laboratory (URL) in Southwestern France. The Tournemire URL is a research laboratory managed by the French public expert in nuclear and radiological risks (IRSN). It is a fundamental tool for IRSN to study the confining properties of clay rock and the performance of certain engineered barrier systems, in order to ensure an independent safety expertise of the French National Radioactive Waste Management Agency's (ANDRA) Cigeo (industrial geological disposal facility) project. The URL tunnel crosses 250 m thick Toarcian-Domerian shale formation, which is interbedded by two limestone aquifers (Dick et al. 2016, Matray et al. 2007). The Tournemire shale shares similarities with the Callovo-Oxfordian argillaceous formation at Andra's Meuse/Haute-Marne URL but the presence of a hydraulically active regional fault and small-scale local faults, constitute extreme conditions (compared to the ANDRA site where such discontinuities have not been identified) for the evaluation of the long-term confinement properties of a clay medium.



**Fig. 1 a.:** Structural map of the Tournemire URL and the location of the URL in France. The fault zone that was under interest (F2) crosscuts the URL tunnel and galleries in several locations. Drill cores used in this work were cored from East Gallery 03. **b.:** The contact between the damage zone of the host rock and the core zone. The fault core exhibits a high strain area with sub-vertical cleavages (Dick et al. 2016).



In this work,  $^{14}\text{C}$ -Polymethylmethacrylate ( $^{14}\text{C}$ -PMMA) autoradiography was used on samples from the Tournemire strike-slip fault zone to achieve high resolution maps of porosity. These samples were then investigated by SEM-EDS to characterize element and mineral distribution within the samples. One of these samples taken from the fault core and containing a well-developed fault gouge is presented in this paper.

## Methods

The clay-gouge samples were taken from a single drill core that was drilled parallel to a small scale, strike-slip fault zone that crosses the Toarcian shale formation and the over- and under-laying limestone units of the Tournemire URL. The internal structure of the fault consists of three main architectural elements: a fault damage zone, a central high strained fault core and an adjacent damage zone-fault core transition. The fault core is bounded on its western side by the principal fault surface and on its eastern side it gradually fades into the damage zone through a transition zone. The fault core (FC) is 50 cm thick and consists of three thin dark bands (ranging from grey to black) of 1-10 mm thick gouge, cataclastic and brecciated rock as well as subvertical schistosity planes and folds. The damage zone-fault core transition zone (DZ-FCT) is 50 to 80 cm thick and consists of spaced schistosity planes dipping towards the south and cross cutting the north dipping bedding planes. The schistosity planes are genetically related to the fault activity and the development of a crenulation cleavage. The damage zone is 2 to 5 meters wide, it bounds the FC to the west and the DZ-FCT to the East, and consists of a dense network of small faults, fractures and veins sealed by crystalline calcite.

The drilled core was oriented perpendicular to the fault plane to reveal the fault gouges' structure and kinematics. The sample piece containing the fault gouges was coated with epoxy resin to prevent it from fracturing.

$^{14}\text{C}$ -PMMA autoradiography is an imaging technique based on intruding an organic monomer liquid methylmethacrylate (MMA) labelled with a radioactive tracer  $^{14}\text{C}$ , into a sample (Hellmuth et al. 1993). The monomer liquid has very low a viscosity and thus an ability to intrude even micro- and nanoscale porosities. The spatial distribution of  $\beta$ -radiation emitted by the tracer is characterized either by film or a digital imaging plate (Sardini et al. 2015, Sammaljärvi et al. 2012).

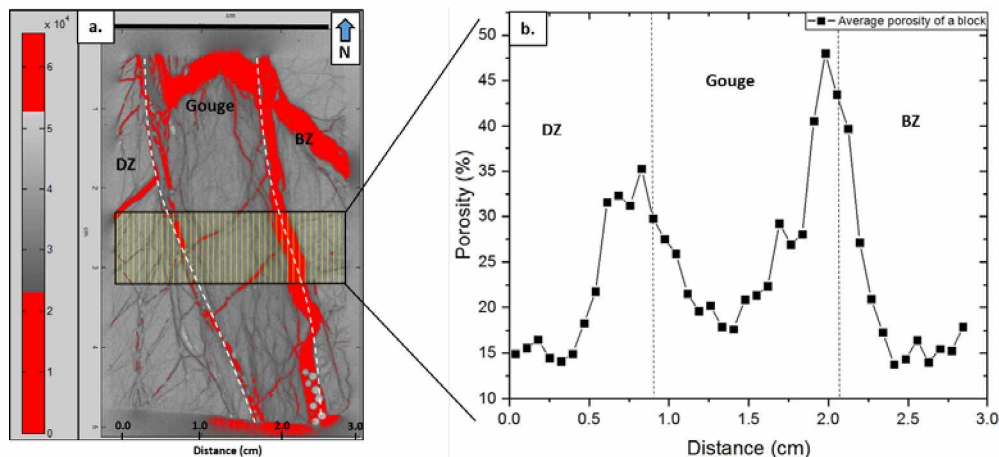
Samples in this work were impregnated in a cylinder vacuum for one month with  $^{14}\text{C}$ -labelled MMA resin in the Radiochemistry unit, University of Helsinki. The used tracer activity was 82 kBq/ml. The samples were then irradiated using  $\gamma$ -radiation with a  $^{60}\text{Co}$  source to polymerize MMA into PMMA in Scandinavian Clinics OÜ, Estonia. The polymerized samples were then sawed to reveal an even surface. Next, these surfaces were polished and exposed for 72 h on FujiTR2025 FLA digital imaging plates and scanned using Fuji 5100 FLA scanner to obtain 16-bit digital autoradiograms. By scanning the sample, a spatial distribution of activity was obtained, presented as an intensity of 16-bit grey scale values. The greyscale intensity values can be converted to activity using calibration series of known activity. The quantitative porosity calculation tools were achieved with the Matlab Image Analysing Toolbox. The results of autoradiography are presented here as grey-scale autoradiograms and porosity maps made by matching porosity values with colours for better contrast. This approach can give clearer images on the local differences in porosity, although grey-scale autoradiogram sometimes shows discrete features more clearly. The uncertainties in  $^{14}\text{C}$ -PMMA autoradiography arise from errors in calibration parameters and optical density measurements (Sammaljärvi et al. 2012). Furthermore, the porosities provided by autoradiography are absolute porosity values.

The element distribution in the sample was studied with a Scanning Electron Microscope with Energy Dispersive X-ray spectroscopy (SEM-EDS). The SEM used in the analysis was JEOL TM JSM-7100F FE-SEM with Inca X-sight EDS system at the Geological Survey of Finland. Moreover, the Backscattering Electron Mode (BSE) was used to distinguish different element compositions.

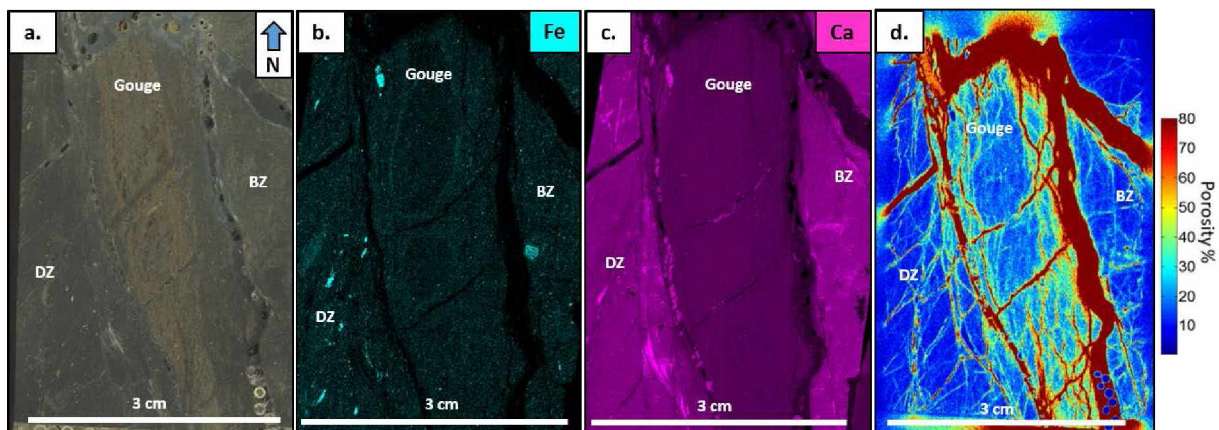
## Examples

The fractures observed from the sample coincide with brittle shear zones and associated Riedel shears within the F2 fault zone. The deformation structures observed coincide with the last Pyrenean orogenic event with dominantly sinistral shear component (Lefèvre et al. 2016).

The gouge appears to have higher tracer activities, as it exhibits darker grey colours than the surrounding DZ and BZ (**Fig. 2a**). Therefore, it can be assumed that the gouge has higher porosities. The porosity histogram (**Fig. 2b**) further indicates that the gouge is distinguishable from the adjacent zones. The porosities in the central gouge approach those of the DZ and BZ. However, the porosities on the gouge margins present significantly higher values, ranging from 30 % to 40 %. The DZ and BZ appear to contain porosities around 15 %.



**Fig.2 a.:** An autoradiogram (AG) from the surface of the gouge sample. The gouge is between a damage zone (DZ) and a brecciated zone (BZ). A yellow block profile was taken across the gouge and adjacent zones to highlight porosity variations. Activities are shown as grey colour intensities and darker grey colour levels thus indicate higher tracer activities. Therefore, higher activities express larger porosities in the sample. Red values are removed from porosity calculations because they are completely filled by PMMA and are thus an artefact from sample handling. On the left is the grey-value scale. **b.:** A porosity histogram from the yellow block profile. Each point represents an average porosity of a corresponding block/slice on the AG porosity image. The interfaces between the gouge and DZ/BZ exhibit significantly higher porosities than the central gouge or DZ/BZ matrix porosities.



**Fig. 3 a.:** A tabletop scan of the gouge sample. **b.:** An EDS-mapping image of iron. Iron exhibits a spherical concentration around the central gouge. **c.:** An EDS-mapping image of calcium. Calcium is



enriched on the DZ and BZ and depleted inside the gouge. **d.**: An AG colour map of porosity from the sample surface.

The SEM-EDS elemental mapping from the sample revealed possibly an iron diffusion halo around the central gouge (**Fig. 3b**). The structure of the halo indicates it was formed at the late stage of deformation after the strain was localized in the gouge margins. Moreover, calcite is concentrated on the DZ and BZ which could indicate fault sealing and healing (**Fig. 3c**).

## Conclusions

The microstructural analysis of the gouges suggests a polyphased tectonic history, consisting in the development of microfabrics subparallel to the main fault plane and concomitant to mineral dissolution due to fluid flow that weaken the gouge. However, simultaneously diffusion processes controlled by the pressure dissolution deformation enabled the precipitation of calcite along the gouge margins, thus strengthening the fault. The continuous shearing of the fault gouge and fluid overpressure build-up enabled the granular material to expand and rupture along the gouge margins creating clay clast aggregates and indicating unequivocal evidence of seismic faulting. Porosity results are in alignment with textural studies as the AG indicates an anisotropy in porosity along the studied gouge zone. The porosity increases towards the gouge and DZ/BZ interfaces and respectively decreases towards the central part of the gouge. The distribution of iron, revealed by SEM-EDS elemental mapping, could indicate multiphase hydrothermal fluid circulation within the fault core. Forced fluid overpressure could lead to fault reactivation and thus allow fluid circulation in the very narrow gouge margins with higher porosities.

This work provides evidence that impermeable small scale faults in clay formations can be naturally thermally pressurized and act as potential drains for fluids at shallow depths.

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