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# Calcite veins as an indicator of fracture dilatancy and connectivity during strike-slip faulting in Toarcian shale (Tournemire tunnel, Southern France)

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## Highlights

- Cathode-luminescence observations indicate that two phases of vein formation occurred.
- Fluids circulations mainly off the shear surfaces in centimeter scale dilatant volumes.
- Relationships between calcite concentration and strain partition around the fault main slip surface.

## Abstract

The reactivation of faults induced by natural/human induced [fluid pressure](#) increases is a major concern to explain subsurface fluid migration and to estimate the risk of losing the integrity of reservoir/seal systems. This study focusses on paleo-fluid migration in a [strike slip fault](#) with >100 m long, affecting a [Toarcian shale](#) (Causses Basin, France). A high [calcite](#) concentration is observed in a 5 cm thick zone at the boundary between the fault core and damage zone. Cumulated displacements in this zone are of millimeter-to-centimeter-scale offsets and different dilatant deformation textures are observed. The zone is affected by thin slip planes containing gouge. Cathodo-luminescence observations indicate that two phases of vein formation occurred. The first phase coincides with the fluid migration along this centimeter thick dilatant zone. The second one is associated to re-shear along the millimeter thick slip planes that results in more localized mineralization, but also in a better hydrologic connection through the shale formation. These results show that in shales fluids may migrate off a slipping surface in centimeter scale dilatant volumes, at first controlled by the intact

shale [anisotropy](#) related to bedding and then favored by brecciating, structures re-orientation and strengthening processes induced by calcite sealing effects.

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## Keywords

Shale

Fault activation

Paleo-fluids migration

Seal

Vein geochemistry

## 1. Introduction

Fluid pressurization effects on the re-activation of faults affecting [shale](#) formations are a crucial process to understand fluid migration through seal layers, eventually producing loss of the reservoir/seal systems integrity. The reservoir/seal system integrity is of key prime importance for many applied domains (oil and gas extraction, storage site of radioactive waste/CO<sub>2</sub> geothermal projects...).

It is commonly admitted that a fault cannot be considered as a single continuous surface but corresponds to a rock volume having various deformation structures on different scales of space and intensity ([Gudmundsson et al., 2001](#), [Constantin et al., 2004](#), [Faulkner et al., 2010](#), [Gudmundsson et al., 2001](#)). Typically, a fault zone is composed of a core surrounded by a damage zone consisting of a transitional zone between the core and the intact [host rock](#) ([Caine et al., 1996](#), [Chester et al., 1993](#), [Wibberley et al., 2008](#), [Faulkner et al., 2010](#)) or multiple fault cores ([Faulkner et al., 2003](#)). The fault core/damage zone boundary sometimes is gradual, but more often appears as a sharp interface interpreted as a [fault plane](#). The fault core is composed of various structures such as slip surface planes, [breccia](#), gouge, [cataclasite](#), ultra-cataclasites, mylonites and ultra-mylonite ([Sibson, 1977](#), [Chester and Logan, 1986](#), [Chester et al., 1993](#), [Bruhn et al., 1994](#), [Caine et al., 1996](#), [Clausen et al., 2003](#), [Kim et al., 2004](#)). Structures observed in the damage zone are fractures, subsidiary faults, veins, folds and cleavages ([Chester and Logan, 1986](#), [Chester et al., 1993](#), [Caine et al., 1996](#), [Hesthammer et al., 2000](#), [Jourde et al., 2002](#), [Berg and Skar, 2005](#)).

Such structural heterogeneity of fault zones induces strongly heterogeneous [fluid flow](#) in the fault zone ([Evans et al., 1997](#), [Mandl et al., 1977](#), [Manzocchi et al., 1999](#), [Yielding](#)

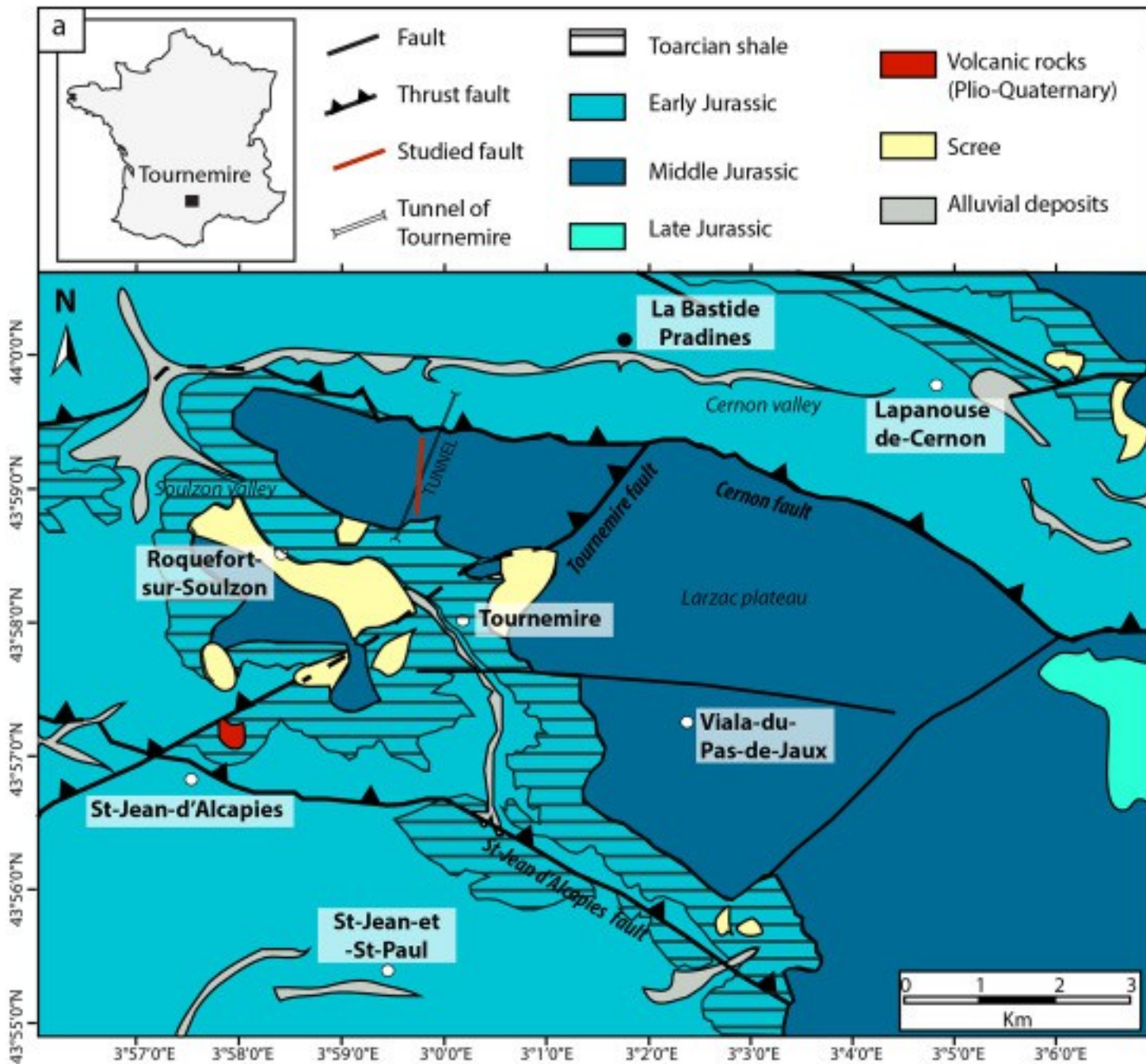
[et al., 1999](#)). The intensification of the [fracture network](#) in the damage zone may lead to an increased permeability with respect to less fractured zone ([Caine et al., 1996](#), [Faulkner et al., 2010](#), [Gudmunson et al., 2010](#)). Fault core, contain specific [grain size](#) reducing structures ([Billi et al., 2003](#)) that generally result in decreasing permeability. On the other hand, during shear movement on the fault, dilatant effects may favor permeability increase, which in return may influence the evolution of [fault slip](#) ([Rice and Rudnicki, 1979](#)).

In fault zones affecting shale formations, self-healing of fractures and sealing with [calcite](#) cement, limits the permeability increase of both the damage and core zones ([Constantin et al., 2004](#), [Nussbaum et al., 2011](#)). Other processes than grain size [reduction](#) can influence the permeability of the fault core, such as mineral reorientation ([Charpentier et al., 2003](#), [Constantin et al., 2007](#), [Arch and Maltman, 1990](#)). [Laboratory experiments](#) on clay rich fault cores show that, for a given [porosity](#) and [effective stress](#), shearing generally results in permeability decrease both along and across the [shear zone](#) ([Wibberley et al., 2008](#)). For these reasons, shale faults are considered to have a low permeability in the same order as that of the intact rock ([Nussbaum and Bossart, 2004](#)). However, porosity and permeability can increase when the stress path applied to the sample results in a dilatant behavior ([Zhang and Cox, 2000](#)). Moreover, the concentration of calcite veins in fault zones suggest that fluid migration can occur during fault activation or movement. The key question is under which stress/strain processes the permeability does increase during faulting in shales. This study investigates the relationships between structural deformations and calcite crystallizations close to the main slipping surface at the interface between the damage zone and the fault core, of a pluri-hectometre reverse-strike slip fault affecting the [Toarcian](#) shales in the Tournemire Underground [Research Laboratory](#) (URL, France). The aim is to identify processes causing permeability increase during faulting in shale that include [dilatancy](#) occurring off the main slipping plane.

## 2. Fault geology in the Tournemire URL and paleo-fluid circulations

The Tournemire Underground [Research Laboratory](#) (URL) located in the [Mesozoic sedimentary basin](#) of the French Causses (Massif Central, France, [Fig. 1](#)), is operated by the French Institute for Radiological Protection and [Nuclear Safety](#) (IRSN), to study the confining properties of an argillaceous formation ([Cabrera et al., 1999](#), [Boisson et al., 2001](#)). The URL is nested in a monoclinical structure tilted 5°–10° to the North ([Constantin et al., 2004](#)) in a 250 m thick [shale](#) formation respectively composed of 50 m thick Domerian and 200 m thick [Toarcian](#) series intercalated

between two 300–500 m thick dolomitic [limestone](#) layers of Bathonien-Bajocian-Aalenian ages and Carixian-Sinemurian ages ([Cabrera et al., 1999](#)) ([Fig. 1](#)). The shale's mineral composition is relatively homogeneous within the upper Toarcian section, with more than 50 wt.% [clay minerals](#), dominantly [illite](#) and illite/smectite, 10–20 wt.% [calcite](#), and 10–20 wt.% [quartz](#). Other components (less than 10 wt.%) include detrital micas, [feldspars](#), [pyrite](#) and Fe-dolomite, and organic matter ([Boisson et al., 2001](#), [Tremosa et al., 2012](#)). The shale formation has a [porosity](#) of 10–13%, ([Dick et al., 2015](#)) a water content of 4–6 wt.% and a hydraulic permeability of  $10^{-14}$ – $10^{-15}$  m/s ( $10^{-21}$ – $10^{-22}$  m<sup>2</sup>) in the laboratory and of  $10^{-11}$ – $10^{-14}$  m/s ( $10^{-18}$ – $10^{-21}$  m<sup>2</sup>) in situ ([Bonin, 1998](#), [Boisson et al., 2001](#), [Cabrera et al., 1999](#), [Courbet et al., 2013](#)).

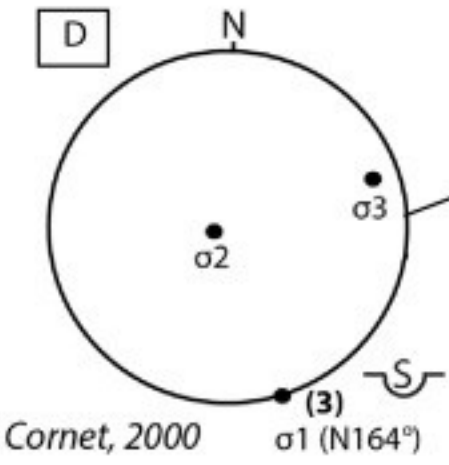
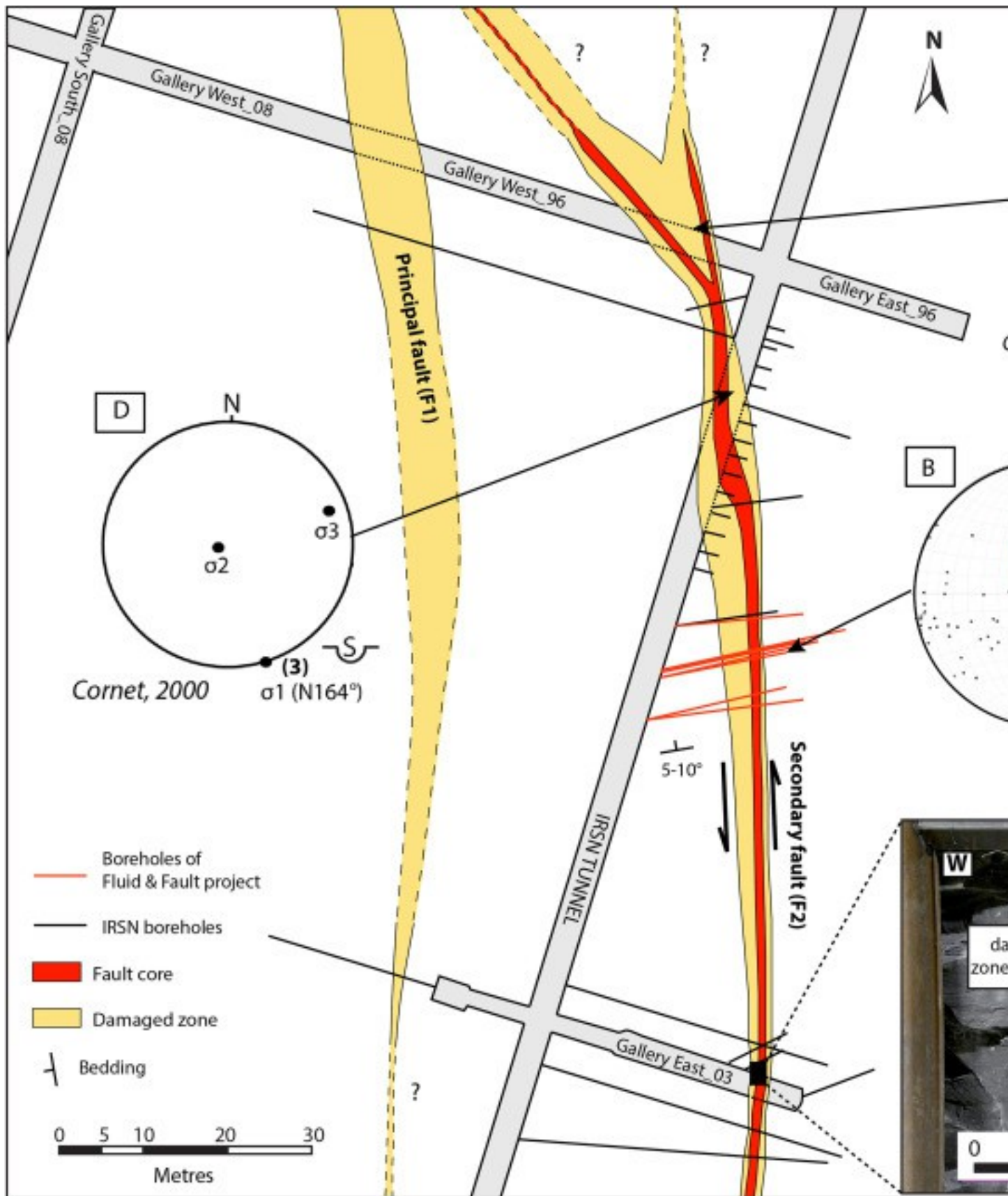


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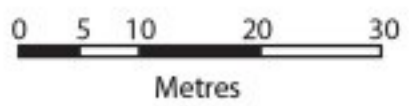
Fig. 1. a- Geological map, the studied fault is indicated in red. b- Simplified stratigraphic log (I1-2, Hettengian; I3-4, [Sinemurian](#); I5, Carixian; I6, Domerian; I7, Early [Toarcian](#); I8-9a, Late Toarcian-Early Aalenian; I9b, Late Aalenian; j1a, Early [Bajocian](#); j1b, Late Bajocian; j2a, Early [Bathonian](#); j2b, Late Bathonian; j3-5, Callovian-Early Oxfordian and j6-7 Late Oxfordian-Early Kimmeridgian). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

The URL is crossed by two main parallel fault zones (F1 and F2) separated by relatively undeformed rock ([Bretaudeau et al., 2014](#)). The location and the geometry of the fault zones were reconstructed using [boreholes](#) previously drilled by IRSN and by mapping the intersections between the faults and the URL galleries' walls ([Fig. 2](#)). Both faults (F1 and F2) have a similar dip and dip-direction spatially varying from N170°–N010°; 60°–90°W ([Fig. 2](#)), characterized by left-lateral strike slip movement with a reverse component ([Peyaud et al., 2006](#)). Some small (meter-to-decameter long) N130°–N150° faults connect F1 and F2 ([Constantin, 2002](#), [Bretaudeau et al., 2014](#)). Faults F1 and F2 are crossing a tectonic block bounded to the North by the N090° Cernon fault, to the South by the N090° St Jean d'Alcapies fault and the N040° Tournemire fault ([Fig. 1](#)) ([Constantin et al., 2004](#)). These three faults extend into the basement and are considered to be inherited structures from the [Hercynian orogeny](#) ([De Charpal et al., 1974](#), [Bonijoly and Delpont, 1982](#)).





- Boreholes of Fluid & Fault project
- IRSN boreholes
- Fault core
- Damaged zone
- Bedding



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Fig. 2. IRSN URL structural map (F1 and F2); A- Picture of the principal [fault plane](#) (N0°-N170°; 80°–90°W) at the boundary between the western compartment of the damage zone (FDZW) and the fault core (FC), in the Gallery East\_03. The black rectangle shows the block sample location ([Fig. 3](#)). B) Poles of fracture and fault planes were determined on the image of [borehole](#) walls and from core analyses of Fluid & Fault project boreholes drilled from the tunnel. C) Paleo-stress orientation determined by [slickensides](#) analyses in Gallery West\_96 ([Constantin, 2002](#)). D) Actual tensor orientation determined by leak off tests ([Cornet, 2000](#)).

Two main tectonic stress regimes have affected the Causses basin. An extension during the Upper [Jurassic](#) and the [Lower Cretaceous](#) ([Blès et al., 1989](#), [Martin and Bergerat, 1996](#), [Constantin et al., 2002](#)), characterized by  $\sigma_3$  oriented N–S was followed by a Mid-Cretaceous to [Eocene](#) Pyrenean compression during which  $\sigma_1$  varied from NE/SW to NW/SE, with two main pulses striking N020°–N030° and N160°–N170°. ([Constantin et al., 2002](#), [Constantin et al., 2004](#)). The Cernon and the St Jean d'Alcapies faults were reactivated as [normal faults](#) during the Mesozoic extension and as [reverse faults](#) during the Pyrenean compression. These faults display a cumulative vertical inverse offset of 100–400 m ([Constantin et al., 2002](#)). F1 and F2 faults presumably are associated to the compressive Pyrenean tectonic phase. Consequently, these faults may have activated as a right lateral strike-slip during the first compressive pulse, and a left-lateral ones during the second (or last) pulse. Left lateral [strain](#) features appear more developed in the fault zone, [Constantin \(2002\)](#) determined a  $\sigma_1$  oriented N134° from microstructural analyses conducted in the northern part of the fault zone ([Fig. 2C](#)). However, this principal stress direction has not been found outside the Tournemire URL, and could result from local stress rotation during the faulting process associated to the N160° tectonic episode ([Constantin, 2002](#)). The analysis of calcite twins on several veins in fault zones in the studied area (1 km<sup>2</sup> around the Tournemire URL, [Fig. 1](#)), [Constantin et al. \(2007\)](#) estimated that the paleo-deviatoric stress ( $\sigma_1$ – $\sigma_3$ ) at the onset of the Pyrenean [tectonic inversion](#) was  $\geq 40$ –50 MPa.

In order to determine the burial depth of the Toarcian shales during the Pyrenean compression period, [Séranne \(2014\)](#) have reconstructed a [subsidence](#) curve based on data from [Barbarand et al., 2001](#), [Séranne et al., 2002](#), [Peyaud et al., 2005](#). This gives a range of maximal burial depths of 2.3–2.4 km roughly corresponding to burial stresses of 57–62 MPa at the onset of the Basin inversion, and at the potential activation time of the studied fault. Thus an estimated magnitude range of paleostress tensor's can be

inferred from the principal stresses applied on the studied fault considering a strike slip regime with a sub-horizontal  $\sigma_1 > 70$  MPa, a sub-vertical  $\sigma_2 \sim 60$  MPa and a sub-horizontal  $\sigma_3 \sim 30\text{--}40$  MPa.

One should note that the present day [stress tensor](#) orientation determined with a series of leak-off tests performed in a vertical 180 m deep borehole ([Cornet, 2000](#)) is identical to the paleostress tensor's orientation during the second Pyrenean pulse, revealing a “residual” strike slip regime on the studied fault ([Fig. 2D](#)). At the URL depth, stress magnitudes are characterized by  $\sigma_1 = 4 \pm 2$  MPa, horizontal and oriented  $N162^\circ \pm 15^\circ E$ ,  $\sigma_2 = 3.8 \pm 0.4$  MPa,  $7\text{--}8^\circ$  inclined from vertical in the  $N072^\circ$  direction and  $\sigma_3 = 2.1 \pm 1$  MPa,  $7\text{--}8^\circ$  inclined from horizontal in the  $N072^\circ$  direction ([Fig. 2C](#)). We may thus consider that this state of stresses has been at steady-state since the end of the uplift period at the Eocene times.

Previous studies ([Peyaud et al., 2006](#)) of carbon and oxygen [isotopic composition](#) of carbonates from the Tournemire URL highlighted two fluid sources: formation fluids contained in veins affecting the intact Toarcian formation (same composition than the shale cement) and meteoric fluids that influence the composition of veins in the main fault zones (and in a meter size alteration halo around the fault). In the fault zones, some mobility of iron, [zinc](#) and [REE](#) (rare earth element) was also reported. [Peyaud et al. \(2006\)](#) observed that pyrite [recrystallization](#) is associated with early calcite cements, which iron content may exceed 1%, indicating reducing conditions prevailed despite of some [meteoric water](#) input. Zoned calcite filling in some geodes indicates fluctuating [redox conditions](#), episodic connections with the overlying Aalenian [aquifer](#), and an evolution in time toward a more open hydrogeologic system ([Peyaud et al., 2006](#)).

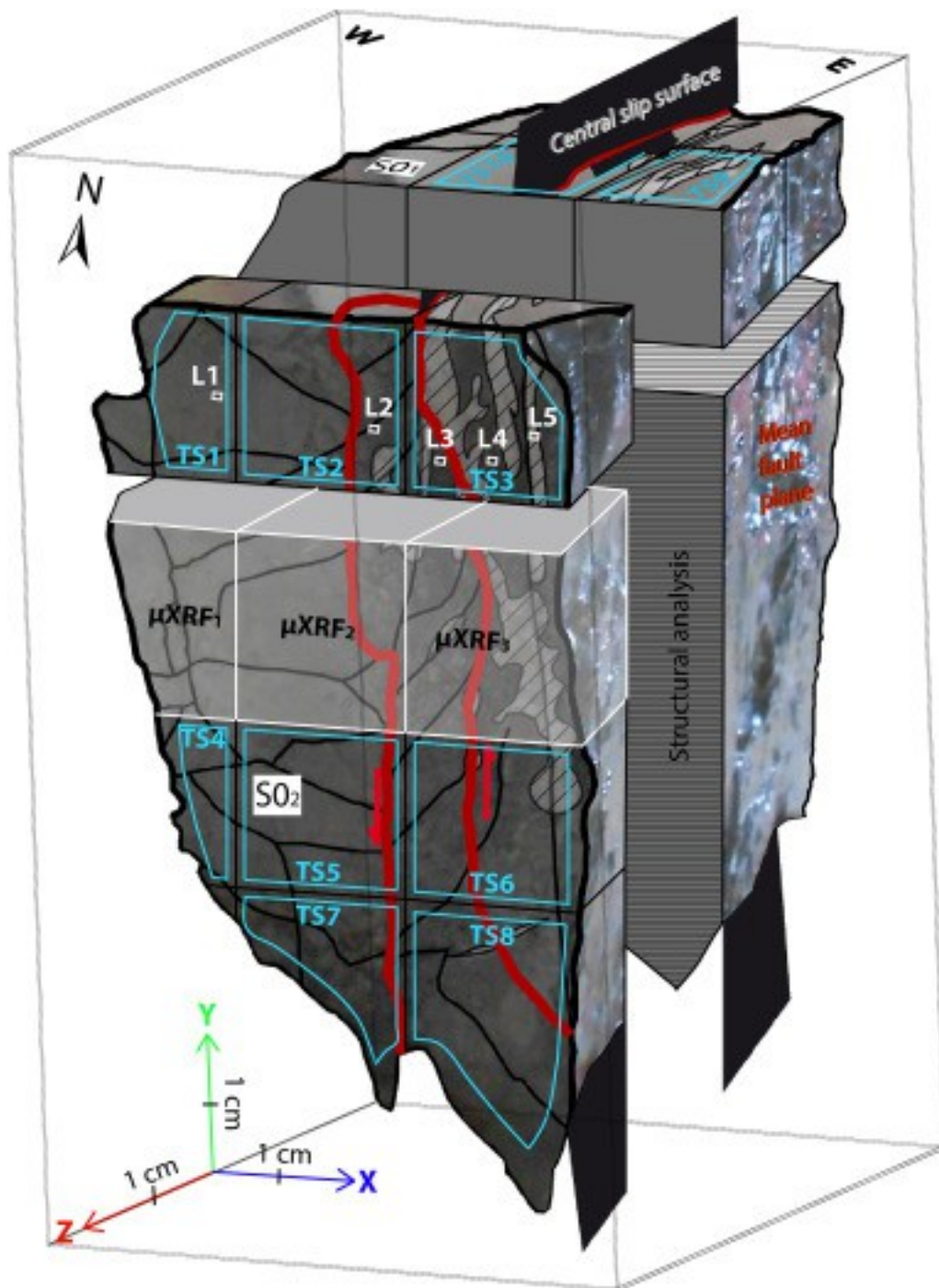
The fault zone studied in this paper (F2) is  $>100$  m long and the vertical offset is estimated to be 4–5 meters ([Dick et al., 2015](#)). The total thickness of the damage zone (footwall and hanging wall) varies spatially between 9 and 15 m depending on the location (estimations were conducted in borehole figured in red in [Fig. 2](#)). The damage zone thickness in the hanging wall (west fractured damage zone; FDZW) is about three times that in the footwall (east fractured damage zone; FDZE). While in the Gallery East\_03 ([Fig. 2](#)), the FDZW and the FDZE roughly have the same thickness. Different fracture and fault orientations characterize the FDZ ([Figs. 2A and B](#)); with main directions respectively trending  $N40^\circ\text{--}N90^\circ$  and dipping  $10^\circ\text{--}90^\circ$  NW or SW:  $N160^\circ\text{--}N020^\circ$  dipping  $20^\circ\text{--}90^\circ$  W, and  $N160^\circ\text{--}N170^\circ$  dipping  $75^\circ\text{--}90^\circ$  W ([Constantin, 2002](#)). Image of borehole (in red in [Fig. 2](#)) walls obtained by the [well logging](#) system OPTV (borehole optical televiewer probe) and cores were used for the [geological](#)

[structures](#) analysis. 10–20% of fractures are parallel or close to bedding. Most of the other fractures (>70%) display a dominant strike of N160°–N020° with large dip variations ([Fig. 2B](#)). Subvertical planes oriented N160°–N020° often expose slickenlines indicating strike-slip motion. The two first direction families display large calcite fillings and some geodes ([Peyaud et al., 2006](#)), while the third one is characterized by polished striated planes.

In the studied zone, the fault core (FC) is 0.1–2.4 m thick. It displays a structural heterogeneity characterized by centimeters thick [shear zones](#) filled with gouge material bounded by 0.1–0.2 m thick highly fractured zones as well as lenses of intact shale. The main orientation of the shear zones follows the N170°–N010°; 80°W average orientation of the fault ([Fig. 2A](#)). Millimeter to centimeter thick calcite veins are observed at the FDZW/FC boundary.

### 3. Methodology

In this paper, we describe a detailed study of [calcite](#) veins localized at the boundary between the damage zone and the fault core of F2 fault ([Fig. 2](#)). This interface is fragile, thus when drilling the boundary between the damage zone and the fault core is systematically damaged and only poor quality samples can be recovered. The gallery\_03 and gallery\_96 walls are the only places where this interface is exposed for favorable sampling conditions. The studied sample comes from the gallery\_03 where the fault's architecture is more simple compared to gallery\_96 where two fault cores are observed. Neither the less similar calcite structures are observed in the two galleries. We have sampled a 12 cm × 8 cm × 8 cm block ([Fig. 2](#), [Fig. 3](#)). The block was sealed in [epoxy resin](#) before being CT-scanned to avoid any fracturing induced by [shale](#) desiccation. A microstructural study was then conducted on polished vertical and horizontal sections. Then, the block was cut into a series of thirteen 22 × 22 × 22 mm cubes, which were analyzed by cathodo-luminescence (CL) on thin sections; three of the cubes were analyzed by micro [X-ray](#) Fluorescence (μXRF). The μXRF is an effective method to perform semi-quantitative chemical non-destructive micro-analyses and chemical mapping with 2D [high spatial resolution](#) (10–100 μm) ([Devès et al., 2012](#)).



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Fig. 3. Studied block sample (Location in [Fig. 2A](#)). Grey hatched block vertical and horizontal surfaces were subjected to a microstructural study presented in [Fig. 4](#). Contoured blue areas (TS1, TS2, TS3, TS4, TS5, TS6, TS7, TS8, TS9 and TS10) represent thin sections locations. White rectangles (L1, L2, L3, L4 and L5) represent the locations of the thin sections photographs presented in [Fig. 5](#). White cubes

( $\mu\text{XRF}_1$ ,  $\mu\text{XRF}_2$  and  $\mu\text{XRF}_3$ ) represent  $\mu\text{XRF}$  analyzed zones presented in [Fig. 8](#). S01 and S02 represent location of stratigraphic plane measurement presented in [Fig. 4](#). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

The  $\mu\text{XRF}$  is operated on an X-ray analytical microscope (XGT-7000) combining micro-X-ray fluorescence with X-ray [microscopy](#). The XGT-7000 consists of an excitation source, a parabolic capillary and an Energy Dispersive Sensor (EDS) detector. The excitation source of the XGT-7000 is an X “primary” ray (XR) produced by an X-ray tube equipped with an [anode](#) of [rhodium](#) (Rh) to which is applied a [voltage](#) of 30 kV.

The parabolic capillary provides a 10  $\mu\text{m}$  diameter very intense ( $10^8$  photons/s) incident XR beam which “excites” the atoms in the sample in order to generate an X fluorescence [emission spectrum](#). Under the influence of X-rays, electrons can be torn from the [energy level](#) on which they are located. The free place is then likely to be filled by electrons located at a higher energy level. These substitutions are accompanied by photon emissions (fluorescence radiation) collected by an EDS detector. This detector can quickly and simultaneously detect a wide energy band of the X fluorescence emitted by several elements (U–Na). Intensities of radiation-emitted fluorescence are proportional to the number of excited atoms and wavelengths (energy) being characteristic of these atoms.

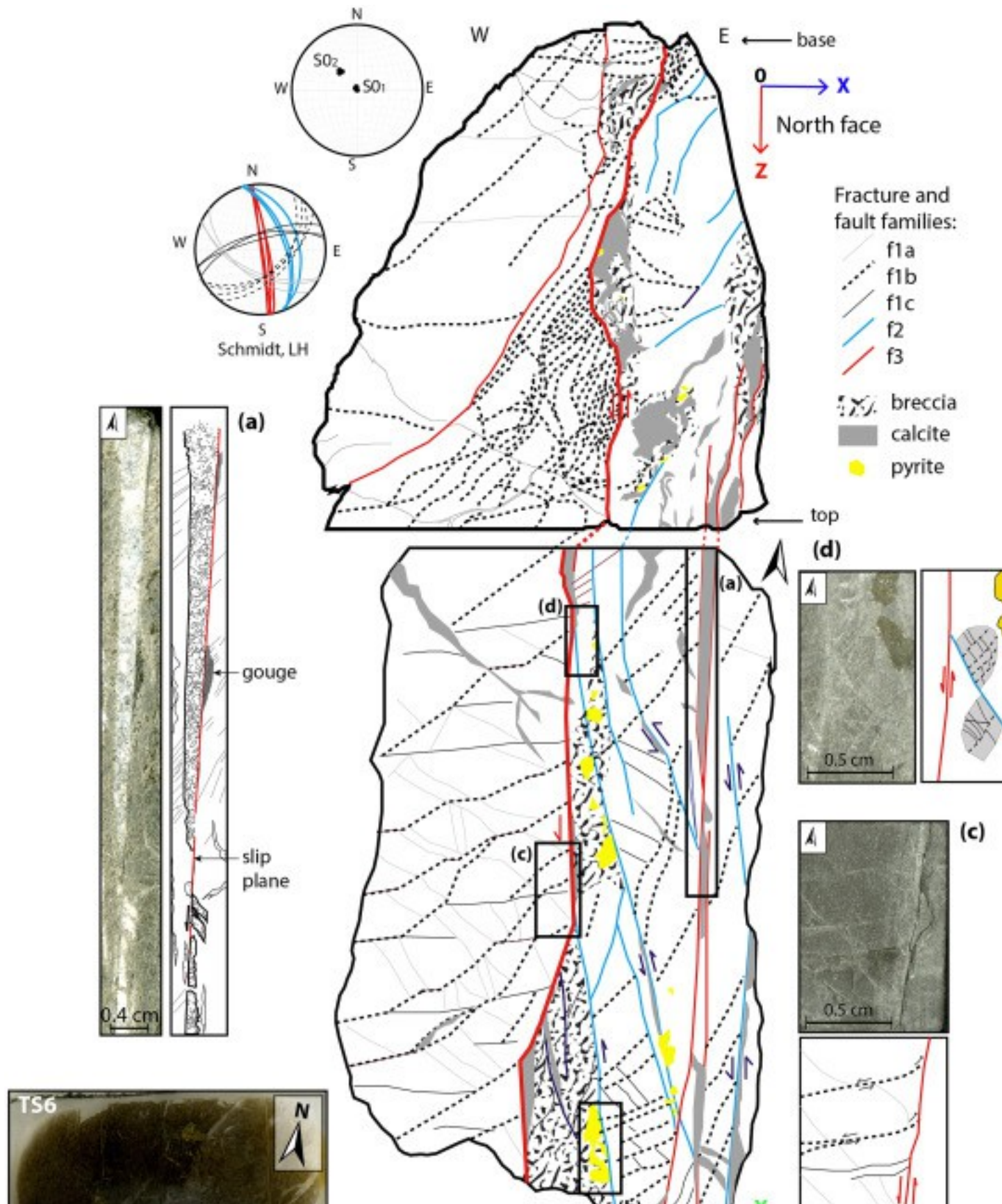
Eight polished thin sections (3 from the eastern compartment adjacent to the main [fault plane](#) displaying a higher intensity of deformation and 5 from the western fractured compartment, TS1 to TS8, [Fig. 3](#)) were examined by optical microscopy and cathodoluminescence. A Technosyn Cold Cathode [Luminescence](#) Model 8200 Mk II is attached to an Olympus™ BH2 microscope and to a Zeiss™ MRc5 digital camera in order to compare the results from optical microscopy and CL. The main purpose is to provide information on the distribution of the calcite and sulphides phase in the [microstructures](#).

## 4. Results

### 4.1. Shear induced dilation at the FDZ/FC transition zone inferred from microstructural analyses

The sampled block may be divided in two: (i) a western fractured compartment and (ii) an eastern compartment adjacent to the main slickenlined [fault surface](#) that bounds the block (eastern extremity of sample, [Fig. 3](#)). The eastern compartment displays a higher deformation intensity. The boundary between the two corresponds to a shear surface ([Fig. 3](#), [Fig. 4](#)). In the western compartment, an increase of the bedding dip is observed from horizontal at the block west boundary to 35°SE close to the central slip surface

(stereo in [Fig. 4](#) and  $S0_1$  and  $S0_2$  locations in [Fig. 3](#)). In the eastern compartment, the bedding is strongly deformed, and only preserved into centimeter-thick zones.





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Fig. 4. Structural analyses of vertical (0zx) and horizontal (0xy) surfaces perpendicular to the principal [fault plane](#) direction (location in [Fig. 3](#)). Grey lines, dashed dark and black lines represent family 1a, 1b and 1c fractures (f1a, f1b and f1c), blue lines and red lines represent family 2 and 3 fractures (f2 and f3), respectively. a- Details of a slip plane zone. b- Details of a brecciated zones cemented with [calcite](#). c- Zoom of anticlockwise rotation and a left-lateral strike slip of family 1 faults close to left-lateral strike family 3 faults. d- Zoom of left-lateral strike slip of family 2 faults indication. e- and f- Details of sigmoidal veins of f1b fault observed in TS6 and TS8 thin sections, respectively (location in [Fig. 3](#)). g- (Pol) is the observation with a polarizing microscope and (CL) are the cathodo-luminescence observations (location in [Fig. 4e](#)). S01 and S02 represent stratigraphic plane measurement (location in [Fig. 3](#)). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

The block is affected by five geometric faults families (f1a, f1b, f1c, f2 and f3):

- 

Families f1a, f1b and f1c (thin grey continuous lines, dashed dark lines and black continuous lines in [Fig. 4](#)) correspond to faults N110°–N130°, dipping 55°–65°SW; N030°–N060°, 45°–60°SE and N060°–N080°, 60°–80°N. Towards the East, the faults are cut by f2 and f3 faults, both strike and dip were rotated anticlockwise, respectively to N30° and >60°–80° dip. f1a, f1b and f1c faults are mostly observed in the western part of the block where they entirely cut the block ([Fig. 4](#)). They are less observed in the eastern part of the block. Sigmoidal veins indicating a dextral movement are observed on f1b fault ([Fig. 4e–f](#)), and an anticlockwise rotation and a left-lateral strike slip reactivation ([Fig. 4c](#)) are observed on f1b and f1c faults close to f3 faults. The apparent offsets are small (a few mm at most).

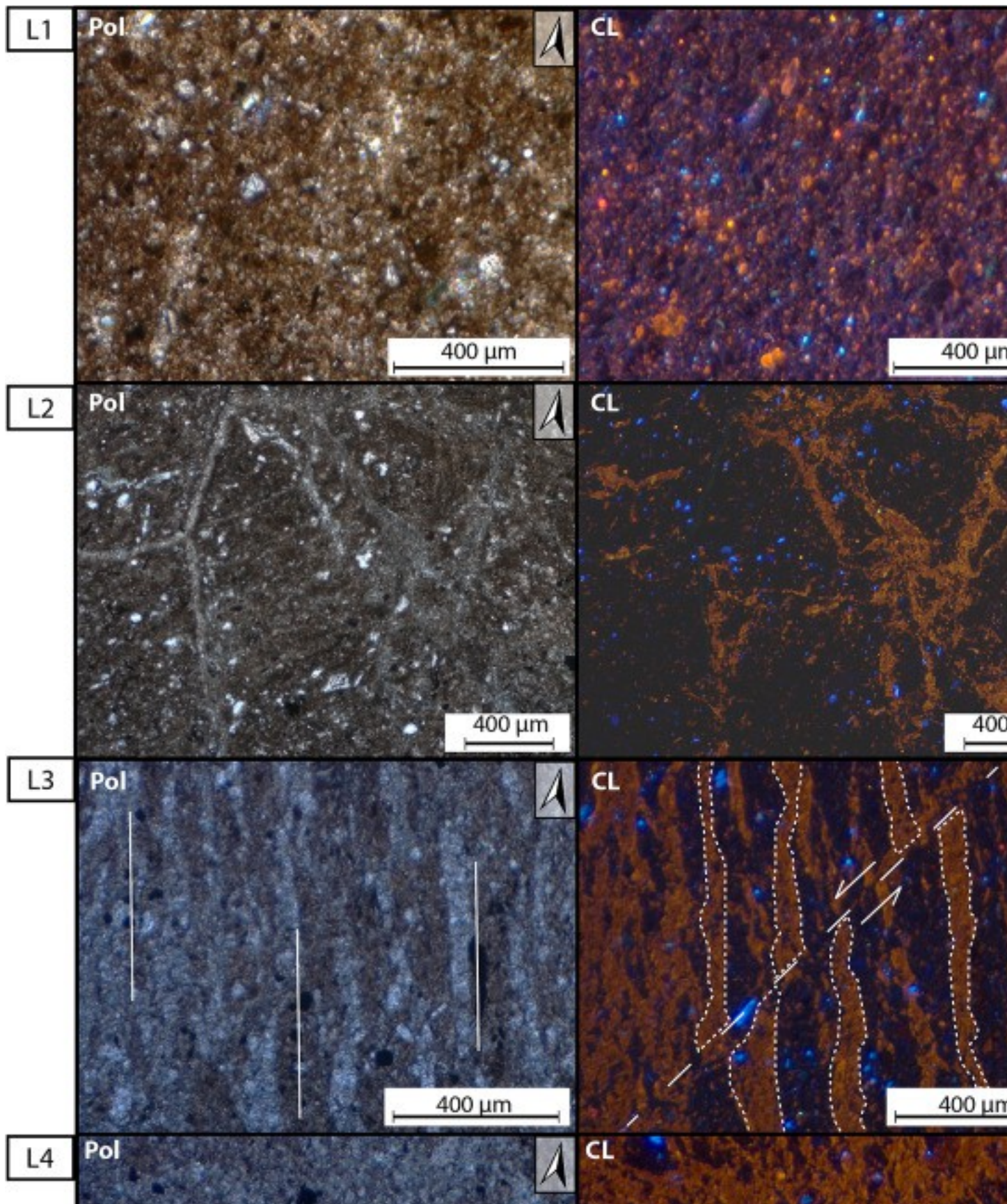
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f2 (blue lines in [Fig. 4](#)) corresponds to left-lateral [strike slip faults](#) N150°–N160° dipping 60°–80° to the E. These faults are mainly located in the eastern part of the block ([Fig. 4d](#)). They do not cut the entire block and branch on to the f3 fault surfaces. They may be considered as Riedel R1-shear planes associated to f3 shear planes during the second compression episode, oriented NW–SE.

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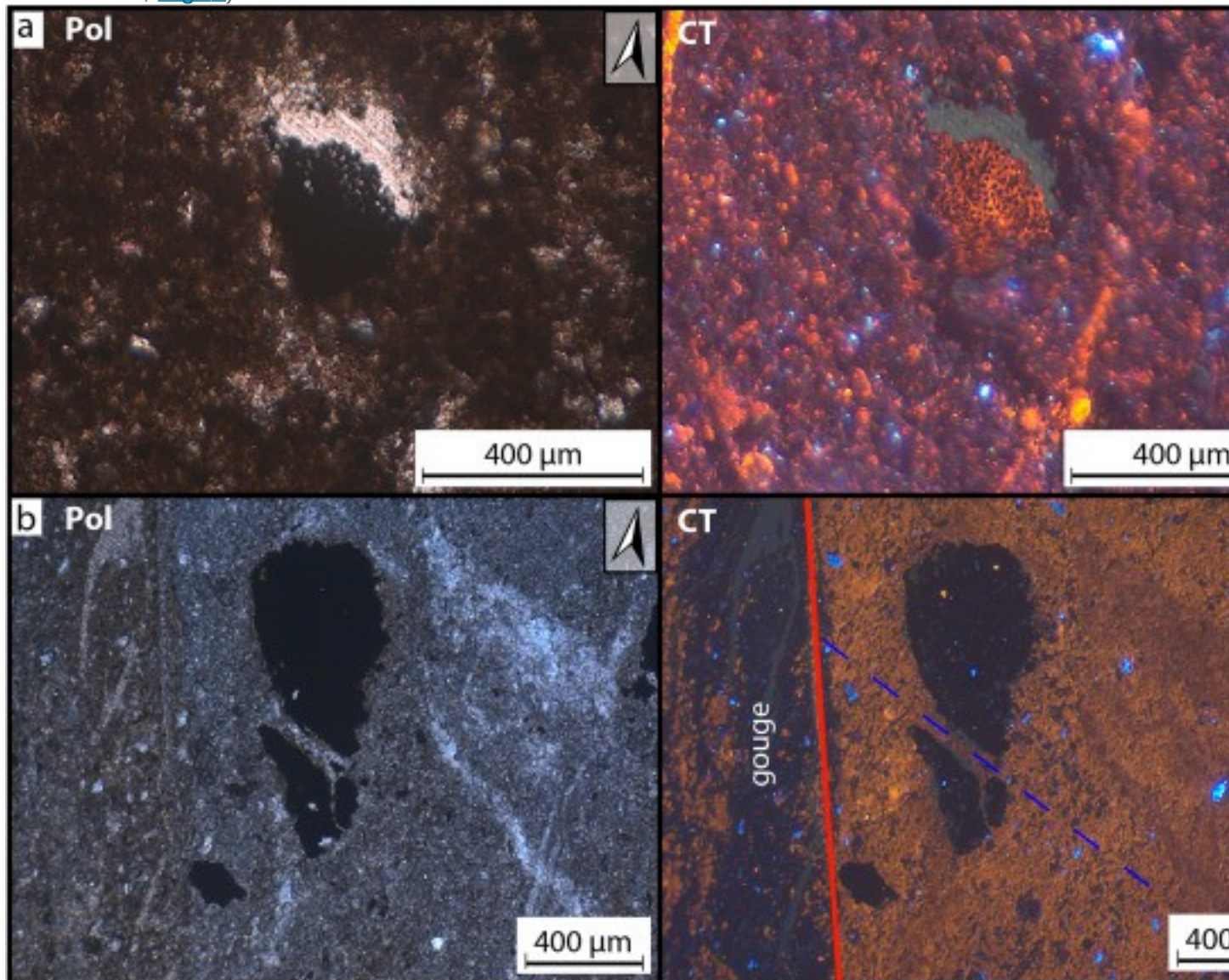
f3 (red lines in [Fig. 4](#)) corresponds to strike slip faults N170°–N180° dipping 80°–90° to the E or W ([Fig. 4c](#)). A left lateral movement is observed with a small vertical component characterized by slickensides dipping 10°–20°N. This family includes the main slickenlined fault surface, the central fault surface bounding the two compartments, and several (4–5) faults located within the eastern compartment of the block. The minimum offset estimated from the displacement of small [calcite](#) shear veins along each plane is several centimeters, thus the total slip on the fault surface may in fact exceed block's length (>5 cm, [Fig. 4](#) and a).

Compared with the western block compartment, the eastern one displays a higher intensity of deformation characterized by brecciated zones composed of [shale](#) material elements cemented with large amounts of calcite intercalated with deformed shale ([Fig. 4b](#)) where the bedding appears rotated anticlockwise. Thin discontinuous black bands (no calcite, [Fig. 6b](#)) show, strong deformation looking like gouge material and calcite shear veins are observed along several f3 slip planes ([Fig. 4](#), [Fig. 6b](#)). Along the central slip plane, at the branching with the f2 faults, centimeter-thick [breccia](#) made of shale and fractured [pyrite](#) elements cemented with calcite are observed ([Fig. 4b](#)), these cemented breccia thus formed during left-lateral strike-slip motion. In these local highly dilatant zones, the breccia displays poorly matrix-supported to cement-supported textures.



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Fig. 5. Thin sections of different fault zone materials. For each thin section, (Pol) is the observation with a polarizing microscope and (CL) is the cathodo-luminescence observation. L1, L2 analyzed zones are localized in the FDZW (in TS1 and TS2 thin sections, [Fig. 3](#)) and L3, L4 and L5 are localized in the DZ/FC transition zone (in TS3 thin section, [Fig. 3](#)).



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Fig. 6. a- Luminescent [calcite](#) (C1) fills the [pores](#) of framboidal [pyrite](#). b- Analyze from TS3, thin section, in the DZ/FC transition zone, ([Fig. 3](#)). Presenting a partly recrystallized pyrite cut by N110°–N120° cracks (dashed blue line) with no apparent

shear displacement. The dark zone (lack of calcite and grain reduction) along a family 3 fault (red line) represent a gouge band. (Pol) is observation with a polarizing microscope and (CL) is cathodo-luminescence observations. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

To summarize, the western compartment bearing fractures between intact rock zones, appears characteristic of the fault damage zone deformation texture, while the eastern compartment represents the edge of the damage zone/fault core (DZ/FC) transition zone with brittle/plastic structures and highly fractured residual blocks. Here, this zone corresponds to a 5 cm thick zone where bedding is rotated parallel to the fault surface. Riedel shear are observed, associated with high concentrations of calcite, centimeter scale displacements and dilatant textures such as opening along pre-existing bedding and breccia.

#### 4.2. Episodes of calcite cementation

The views L1 and L2 respectively from thin sections TS1 and TS2 ([Fig. 3](#)) show two typical [microstructures](#) of the FDZW ([Fig. 5](#)). L1 is representative of relatively intact or slightly-deformed rock, composed of carbonates in the form of highly cathodoluminescent and bright orange (in CL) calcite grains, bioclasts, [quartz](#), framboidal pyrite and a dominant fraction of [clay minerals](#). The mineral distribution is relatively homogeneous without any apparent deformation. L2 shows carbonates (mainly high CL calcite cement) concentrating into N010°–N170° and N080°–N100° [micro-cracks](#) and fractures while the neighboring clay matrix has fewer calcite grains when compared to L1 ([Fig. 5](#)).

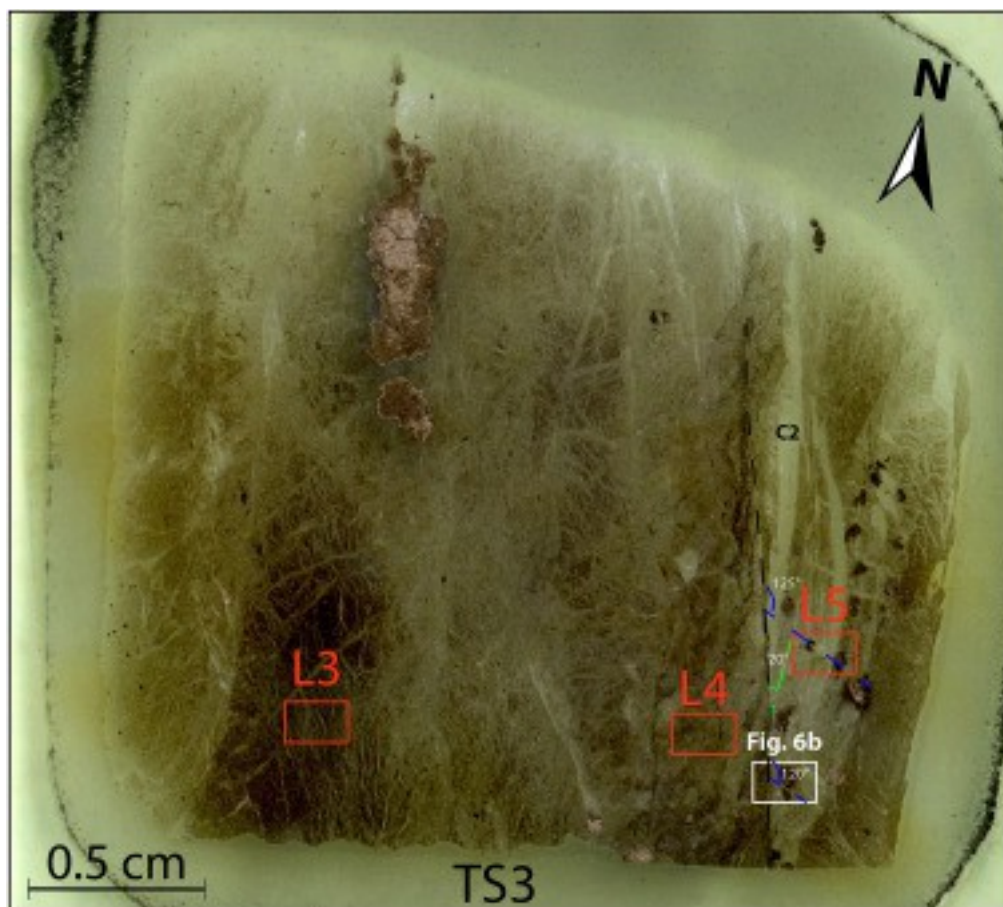
L3, L4 and L5 are views ([Fig. 5](#)) from the thin section TS3 cut perpendicularly to the central slip surface in the DZ/FC transition zone ([Fig. 3](#)). L3 is located in the eastern wall of the central slip surface, a zone where stripes of calcite irregularly alternate with stripes of clay material. Stripes are roughly oriented N160°–N170° and are hundreds of [micrometer](#) long. They are sub-parallel to the central fault plane and are cross cut by a N040° left lateral shear fault (associated with f1b, dashed white line in L3, [Fig. 5](#)), which could in this context be tentatively interpreted as a R2 shear during the first compression episode, oriented NE–SW. The framboidal pyrite which is located in the [bedding plane](#) in the intact rock (L1 sample) here is located in the stripes. This observation suggests that the calcite stripes formed from opening of the bedding and were rotated parallel to the main fault surface (figured by the white continuous lines in L3, [Fig. 5](#)) ([Fig. 6](#)).

The L4 thin section is located in a brecciated DZ/FC transition zone ([Fig. 3](#)). The stripe pattern in L4, appear to be more complex than in L3 as calcite veins more or less surround a partly recrystallized pyrite framboid bearing larger [crystals](#) of irregular shape. Luminescent calcite fills the [pores](#) in some framboidal pyrites, which also appear altered compared to L1-3 thin sections ([Fig. 6a](#)).

The thin section L5 is located close to the f3 [shear zone](#) ([Fig. 3](#)). There are few shale fragments and quartz grains remaining. Two phases of calcite veins appear, intense CL calcite (C1) being cross-cut by a low luminescent (C2) one (L5, [Fig. 5](#)).

C1 [luminescence](#) is close to the calcite luminescence observed in the intact shale matrix in thin sections L1 and L2, which could support the hypothesis that C1 originates from the intact shale formation. Likewise, the sigmoid veins indicating a dextral movement, thus presumably formed early in the strike-slip fault history, are composed of the C1.

C2 calcite crosscuts the C1 phase along two main directions. One corresponds to centimeter-thick cracks connecting to the f3 shear planes with an angle of about  $20^\circ$  (dashed green line in L5, [Fig. 5](#), [Fig. 7](#)) and then evolving into subparallel secondary planes. The other one corresponds to thin ( $<0.1$  mm) 0.2–0.5 cm cracks making an angle of  $120^\circ$ – $130^\circ$  (dashed blue line in L5, [Fig. 5](#), [Fig. 7](#)) with the f3 shear planes (dashed black line in [Fig. 7](#)). These  $N110^\circ$ – $N120^\circ$  cracks (dashed blue line in L5, [Fig. 5](#), [Fig. 7](#)) cut the recrystallized framboidal pyrite with no apparent shear displacement, while the  $N180^\circ$ – $N020^\circ$  (green line in L5, [Fig. 5](#), [Fig. 7](#)) veins induce a stretching of the pyrite crystals ([Fig. 6b](#)).

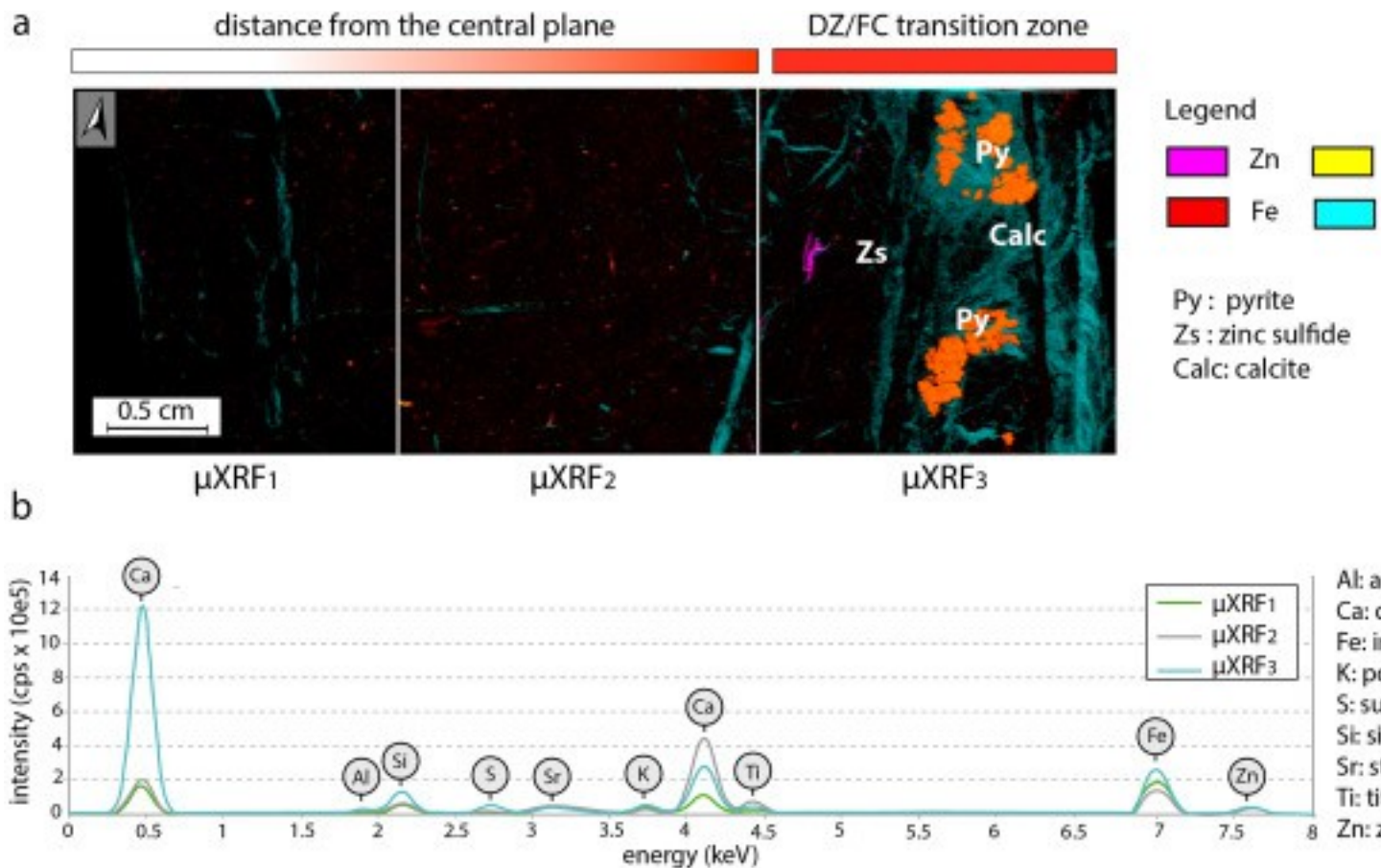


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Fig. 7. Picture of the TS3 thin section with L3, L4, L5 (presented in [Fig. 5](#)) and [Fig. 6b](#).

#### 4.3. Micro-analyses

The main identified chemical elements are Ca, Fe, Zn, S, Si, Al, K, Sr ([Fig. 8](#)). The  $\mu$ XRF chemical mapping of three horizontal thin sections along a profile perpendicular to the [fault slip](#) surface ([Fig. 3](#), [Fig. 8a](#)) shows a difference in the distribution of Ca and Fe in the DZ/FC transition zone (eastern compartment of the sample:  $\mu$ XRF<sub>3</sub>, [Fig. 3](#)) compared to DZ (western compartment  $\mu$ XRF<sub>1</sub> and  $\mu$ XRF<sub>2</sub>). [Fig. 8b](#) shows an increase in the Ca (Ca intensity peak  $\times 6$ ) and Fe (Fe intensity peak  $\times 2$ ) concentrations and the appearance of [zinc sulphides](#) in the DZ/FC transition zone.



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Fig. 8. a-  $\mu\text{XRF}$  studied zone (1 px = 100 microns, incident: beam: spot 100 microns, 30 kV, [anode](#) Rh, counting time: 800 s  $\times$  10),  $\mu\text{XRF}_1$  and  $\mu\text{XRF}_2$  are localized in the FDZW and  $\mu\text{XRF}_3$  is localized in the DZ/FC transition zone (see [Fig. 3](#) for locations). b- Mean spectra for each of the  $\mu\text{XRF}_1$ ,  $\mu\text{XRF}_2$ , and  $\mu\text{XRF}_3$  zones showing the chemical variations between the FDZW and the DZ/FC transition zone.

In the damage zone ( $\mu\text{XRF}_1$  and  $\mu\text{XRF}_2$  zones in [Fig. 8a](#)), Ca concentrates in f1 faults. Close to the main slip plane ( $\mu\text{XRF}_3$  zone upper right in [Fig. 8a](#)), f1 faults are sheared by f3 and f2 faults. In the DZ/FC transition zone, there is a high concentration of diffuse Ca zones affected by veins associated to the f3 shear zones. These shear zones are made of micro-to-millimeter bands of clay minerals and vein shapes are in good accordance with left lateral slip along the shear zones. In details in  $\mu\text{XRF}_3$  zone (bottom east in [Fig. 8a](#)), calcite veins appear thicker at the [contact](#) with the shear zone, and then evolve into thin calcite veins parallel to the shear zone. The Fe content is diffuse in the damage zone where it corresponds to the small infra-millimeter framboidal pyrite grains, and concentrated in large 0.3–0.5 cm patches of mainly recrystallized pyrite in the

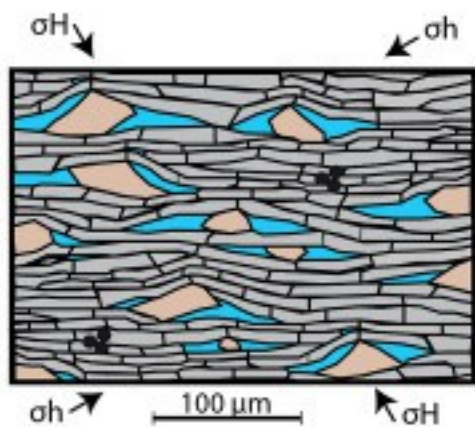


DZ/FC transition zone ([Fig. 8a](#)). In  $\mu\text{XRF}_3$  zone (lower part), the large patch of pyrite displays a sigmoid shape, showing that pyrite is involved in the shear deformation associated to the f3 slip planes.

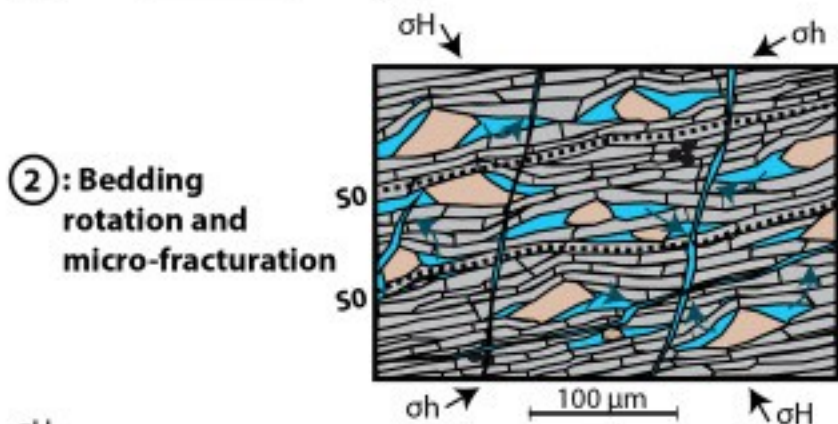
## 5. Interpretation/discussion

### 5.1. Relationships between vein growth, fluid composition and deformation within the fault zone

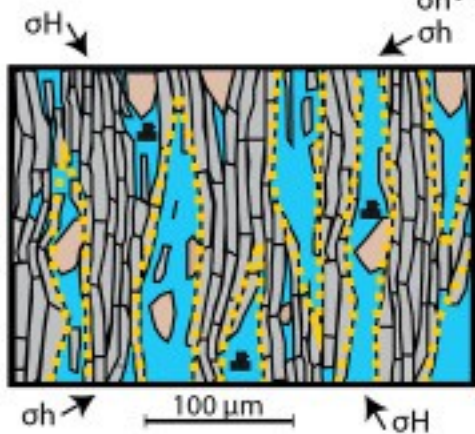
Our observations clearly show ([Fig. 9](#)): (i) a concentration of [calcite](#) in the most deformed part of the DZ/FC transition zone, (ii) two successive calcite precipitation phases associated to different processes of fault deformations and (iii) that slip planes are composed of clay gouge, most calcite veins being located off the slip planes ([Fig. 6b](#)). The [Fig. 10](#) highlights two deformation sequences observed in the sample for the two calcite phases; the “Crack + Seal” and the “Crack + Slip + Seal” deformation sequences ([Petit et al., 1999](#)).



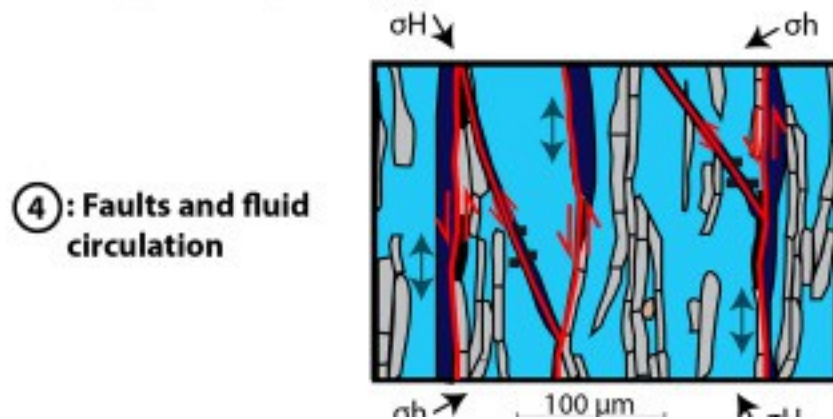
①: Initial state



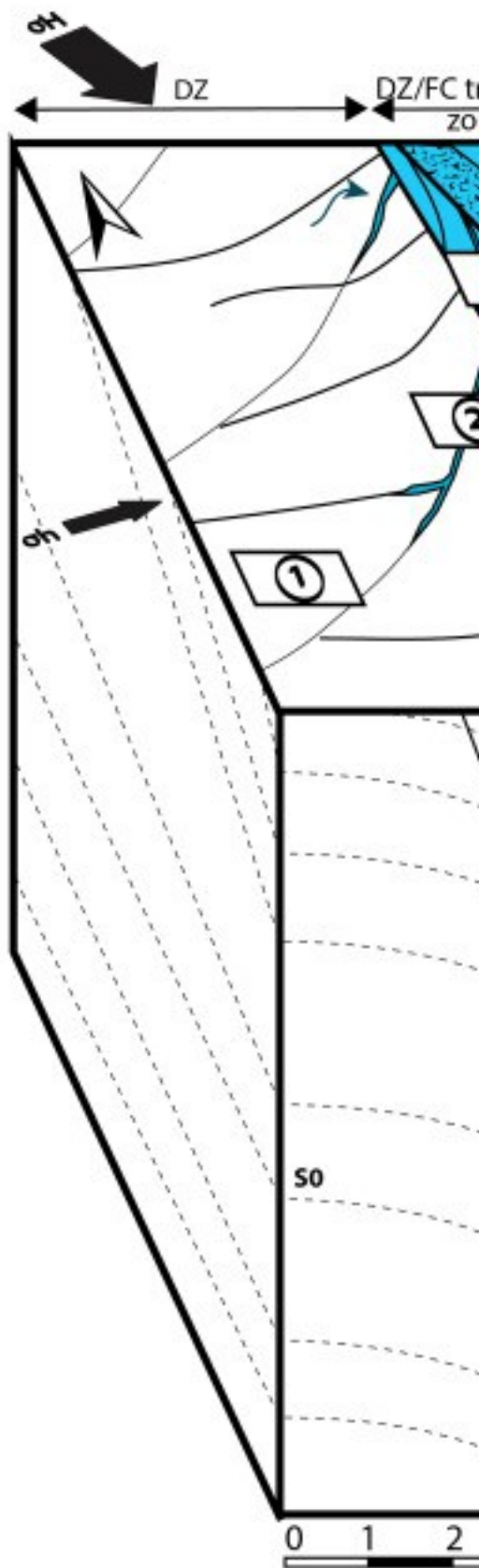
②: Bedding rotation and micro-fracturation



③: Segregation and destructureation

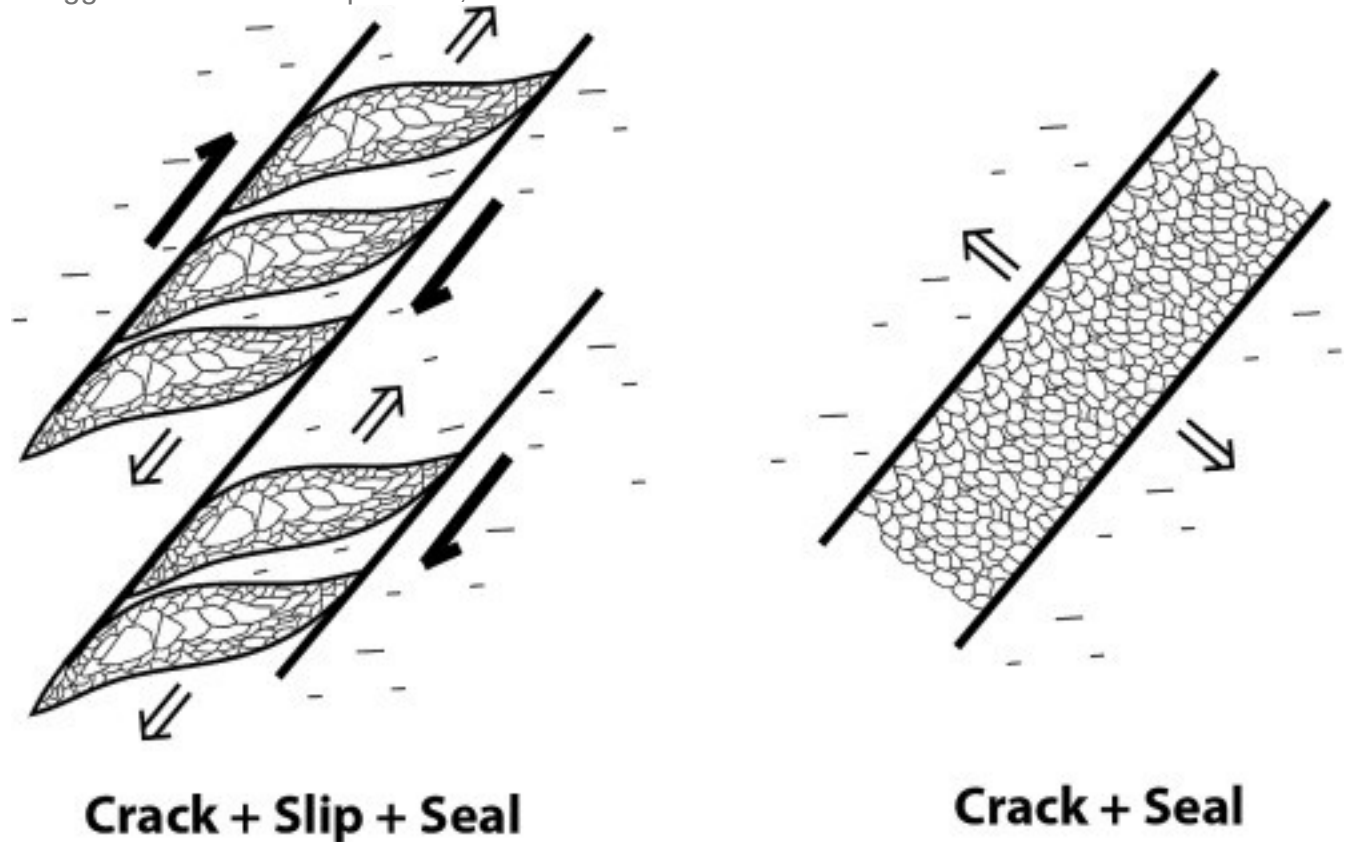


④: Faults and fluid circulation



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Fig. 9. Synthesis of the coupled paleofluids-strain observations at the Tournemire fault main slip surface. Family 1 fractures have not shown and the fluid proportion is exaggerated in 3 and 4 pictures, for a better visualization.



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Fig. 10. Representation of “crack, then seal” and “crack + slip + seal” deformation sequences.

The first calcite (C1) phase presumably precipitated from a fluid in equilibrium with the formation. It is associated with f1-to-3 fractures in the damage zone, opening of [bedding planes](#) and breccification within the fault DZ/FC transition zone. [Pyrite recrystallization](#) occurs more or less synchronously. The second calcite (C2) phase is associated to late dilation associated with slip on f3 [shear zones](#). It is characterized by sigmoid veins branching from f3 shear zones and small mode 1 cracks. Calcite from this phase displays lower orange [luminescence](#) than earlier calcite cements, presumably indicating a lower Mn content (e.g. [Cazenave et al., 2003](#)). Some larger [crystal](#) of partly recrystallized framboidal pyrite associated with the first

calcite [cementation](#) phase have been fractured and these fractures are filled with low luminescence calcite of the second phase. The second phase is thus associated to late shearing and brittle fracturing within a cemented part of the fault core. There is no evidence of pyrite [crystallization](#) during this phase.

[Peyaud et al. \(2006\)](#) show that the later calcite cements are formed under less reducing conditions than the earlier cements. The fault zone studied here displays comparable evolution with an earlier episode of high luminescence calcite precipitation and pyrite recrystallization and a later episode of low luminescence calcite precipitation without pyrite recrystallization. We infer the precipitation of these two phases correspond to a similar evolution from more reducing to less reducing conditions and indicate hydrogeologic opening of the system. The later cements represent a comparatively small volume and only occur in association with left lateral slip surfaces. The earlier cements formed during left-lateral slip as well as during an earlier right-lateral episode and possibly also during a syn-sedimentary extensional episode. The hydrogeologic opening of the system occurs relatively late in the faulting history and we wonder whether this opening is the consequence of a better connectivity of the [fracture network](#) in the fault zone or the effect of [exhumation](#), erosion and karst development in the overlying formations.

Faults from f1 may be explained by both the state of stresses during the early Basin [tectonic inversion](#) and the intact [shale anisotropy](#) related to bedding (transverse isotropy). Indeed, according to laboratory tests ([Ambrose et al., 2014](#)), if we consider an early [upper-cretaceous](#) pulse characterized by N020°–N030° trending horizontal  $\sigma_1$  ([Constantin et al., 2004](#)), magnitude over 70 MPa affecting consolidated shales parallel to bedding and a confining stress of about 60 MPa may generate shear rupture surfaces in the shale matrix creating angles of about 20°–30° with the bedding. It could explain the [reverse faults](#) N110°–N130° dipping 55°–65°SW and N060°–N080° dipping 60°–80°N. Another explanation of f1 faulting could relate to a Riedel shear zone system, in accordance with an early dextral movement on a pre-existing mean [fault plane](#) (during the first NE–SW compression episode).

f2 and f3 faults cross cut f1b and f1c fractures which display an anticlockwise rotation (close to the f3 planes) in good accordance with a left-lateral strike slip reactivation. This reactivation highlights a second compression episode characterized by a N160°–N170° trending horizontal  $\sigma_1$ , in good accordance with the principal stress rotation during the second Pyrenean pulse ([Constantin et al., 2002](#), [Constantin et al., 2004](#)). The C1 phase is associated with dextral movement of the fault probably during the first episode of the Pyrenean compression (NE–SW) and continued at least during the beginning of the left-

lateral phase, notably associated with [breccia](#) formation at the intersections of slip planes. The C2 phase is exclusively associated with the left-lateral reactivation of the fault during the second episode of the Pyrenean compression (SE–NW). The differences between both calcite [microstructures](#) may mainly result from the degree of pre-rupture cementation of the fault zone that may alter the fault compliance, the C2 phase affecting already cemented and less compliant fault rocks than the C1 phase.

## 5.2. Permeability variations during faulting

The distribution of veins in shear zones is controlled by enhanced [dilatancy](#) along releasing segments of the shear zones, which may [channel fluid flow](#) and eventually contribute to resealing the fault by calcite precipitation. Different ways of potential permeability increase related to dilation induced by fault movements affecting shales are observed in the studied block. Most dilation occurs at a distance from the main slipping planes. Patches of brecciated shale with high fracture [porosity](#) formed around the intersections between the main shear planes and secondary faults (e.g. R1) or inherited structures such as re-oriented bedding or small faults formed in a different stress regime. Interestingly, these high dilatant zones take place in relatively low [strain](#) areas, in the transition zone between the damage zone and the fault core. In this case study, the zone containing potential high permeability conduits for flow is a few centimeters-thick. During the reactivation phase the same dilatant effects may occur much more localized close to a few active shear planes with clay gouge. This is in accordance with permeability tests conducted during laboratory [shear tests](#) on a sample pre-cut by a fracture that showed series of fractures and a complex pattern of deformation occurring off the slipping fracture surface to a distance of about 1 mm ([Cuss et al., 2011](#)). In addition, [Bolton et al. \(1999\)](#) showed that after being deformed at a high over-consolidation ratio, shear zones in [fine-grained sediments](#) can dilate and thereby act as efficient fluid-flow conduits. In the Tournemire shales, both over-consolidation under a high burial stress and calcite sealing may have produced the conditions for brittle dilatant failure in shear and a potentially strong permeability-vs-stress dependency.

## 6. Conclusions

The micro-structural and chemical analyses of a block sample from a fault zone intersected in the Tournemire URL, indicate two [calcification](#) phases took place. The fault zone recorded two episodes of compressive deformation during the Pyrenean [orogeny](#) along NE–SW and SE–NW directions that result in the formation of both right lateral and left lateral shear structures, the latter being more developed in the

block studied. We show that the first [calcite](#) phase has fossilized fluids [channels](#) conditioned by the brittle dilatant behavior of the anisotropic intact shales under moderate confining stress. Then, sealing of the initial fault zone structures occurred, conditioning much more localized and comparatively less dilatant [strain](#) during a late episode of left-lateral slip ([Fig. 9](#)). Our observations indicate that dilation occurs in a wider zone during fault initiation (all the studied block is affected) or at least when strain was not localized on discrete slipping surface, than when the fault is reactivated along a main shear plane at the boundary of the fault core. In any case, most dilation occurs off the main shear planes, and is primarily associated to interaction between structures, either pre-existing like bedding or early fractures, or coevolving such as Reidel shears.

Although dilation, as indicated by vein volume, appears more developed during the early stages of deformation, the coeval [recrystallization](#) of [pyrite](#) suggests the prevalence of reducing conditions. The later opening of the hydrogeological system to more oxidizing conditions is probably the result of a better hydraulic connectivity of the system during later stages of fault activity.

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