



Fluid-mediated, brittle-ductile deformation at seismogenic depth: Part I- Fluid record and deformation history of fault-veins in a nuclear waste repository (Olkiluoto Island, Finland)

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12 Abstract. The dynamic evolution of fault zones at the seismogenic brittle-ductile transition zone (BDTZ) expresses the delicate interplay of numerous physical and chemical processes that occur at the time of strain localization. Deformation and flow of 13 14 aqueous fluids in these zones, in particular, are closely related and mutually dependent during cycles of repeating, transient 15 frictional and viscous deformation. Despite numerous studies documenting in detail seismogenic faults exhumed from the BDTZ, uncertainties remain as to the role of fluids in facilitating deformation in this zone, particularly with regard to the 16 mechanics of broadly coeval brittle and ductile deformation. We combine here structural analysis, fluid inclusion data and 17 18 mineral chemistry data from synkinematic and authigenic minerals to reconstruct the temporal variations in P, T and bulk 19 composition of the fluids that mediated deformation and steered strain localization in a strike-slip fault from the BDTZ. This is a fault formed within the Paleoproterozoic migmatitic basement of southwestern Finland, hosting in its core two laterally 20 21 continuous quartz veins formed by two texturally distinct quartz types - Qtz I and Qtz II, where Qtz I is demonstrably older 22 than Qtz II. Veins within the diffuse damage zone of the fault are infilled by Qtz I. Multi-scalar structural analysis indicates 23 recurrent cycles of mutually overprinting brittle and ductile deformation. Fluid inclusion microthermometry and mineral pair 24 geothermometry indicate that both quartz types precipitated from a fluid that was in a homogeneous state during the recurrent cycles of faulting, and whose bulk salinity was in the 0-5 wt% NaCleq range. The temperature of the fluid phase involved with 25 26 the various episodes of initial strain localization and later reactivation changed with time, from c. 240 °C in the damage zone to c. 350 °C in the core during Qtz I precipitation to < 200°C at the time of Qtz II crystallization. Fluid pressure estimates show an 27 28 oscillation in pore pressure comprised between 160 and 10 MPa during the fault activity stages. Our results suggest significant 29 variability in the overall physical conditions during the fault deformation history, possibly reflecting the interaction of several 30 batches of compositionally similar fluids ingressing the dilatant fault zone at different stages of its evolution, each with specific 31 T and P conditions. Initial, fluid-mediated embrittlement of the faulted rock volume generated a diffuse network of joint and/or hybrid/shear fractures in the damage zone, whereas progressive strain localization led to more localized deformation within the 32





33 fault core. Localization was guided by cyclically increasing fluid pressure and transient embrittlement of a system that was

- 34 otherwise at overall ductile conditions.
- 35 Our analysis implies that fluid overpressure at the brittle-ductile transition can play a key role in the initial embrittlment of the
- 36 metamorphic basement and strain localization mechanisms.
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38 1 Introduction

39 Physical and chemical properties of fault systems play a fundamental role in controlling the rheological behaviour of the Earth's 40 crust and in steering channelled fluid flow (e.g. Caine et al., 1996). Deformation and fluid flow are closely related and mutually 41 dependent via a number of feedbacks, such as the control that fluids exert upon the effectiveness of deformation processes and 42 the development of fault systems at all scales, and the control by rock heterogeneities and/or fracture system topology on the 43 net fault transmissivity (e.g. Crider and Peacock, 2004). The nucleation and development of permeable fault systems and the 44 mechanisms whereby individual faults may weaken and eventually fail are, therefore, complex functions of a number of 45 processes. In this perspective, the multiscalar interaction between fluid and mineral phases within fault rocks needs to be 46 studied with a system approach in order to single-out the roles and importance of all processes involved (Kaduri et al., 2017). 47 The most evident effect of fluid involvement, particularly in crustal volumes that have experienced large deformation-48 controlled fluid fluxes, is the precipitation of authigenic and hydrothermal minerals within faults (Oliver and Bons, 2001 Viola 49 et al., 2016) and their immediately adjacent host rock (Mancktelow and Pennacchioni, 2005; Garofalo, 2004). In the 50 seismogenic region of the crust, where fluids may even be the primary driver of the seismic cycle (e.g Miller, 2013), faults have 51 been shown to have the potential to function like a "fluid-activated valve", whereby they experience transient and cyclic fluid 52 pressure build-up before sudden fluid venting, pore pressure- and mechanical strength drop concomitant with seismic failure 53 (e.g. Sibson, 1989, 1992b, 1993; Cox, 1995; Viola et al., 2006; De Paola et al., 2007; Wehrens et al., 2016). Hydrothermal ore 54 deposits, where fault networks focus relatively large volumes of ore fluids and precipitate economic minerals (Cox et al., 2001), 55 are also pertinent examples of significant deformation-controlled fluid ingress. 56 The seismogenic depth down to 10-15 km (e.g. Kohlstedt et al., 1995) is thus a key region of the crust where to study the whole 57 range of fluid-rock interaction processes occurring within fault zones. Deformation at that depth might be accommodated under 58 overall brittle-ductile conditions along fault systems crossing or rooting into the brittle ductile transition zone (BDTZ). In detail, 59 the deformation style in the BDTZ is generally characterized by the cyclicity, also at the short time scale, between brittle and 60 ductile behaviour. This is induced and regulated by the complex and transient interplay of numerous parameters, among which the lithological composition and transient variation of temperature, pore pressure and strain rate within the deforming system. 61 Field studies have documented unequivocally that ductile and brittle deformation may even be simultaneously active during 62 63 deformation as a function of the transient and spatially heterogeneous evolution of the chemical and physical parameters steering deformation, leading to the broad coexistence of geological features expressing frictional and viscous deformation 64

65 mechanisms and to mutual crosscutting relationships thereof (e.g., Guermani and Pennacchioni, 1998; Kjøll et al., 2015;





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66 Pennacchioni et al., 2006 Wehrens et al., 2016).

Veins are particularly important in this context because they attest to the relative abundance of aqueous fluids in the deformation history (e.g. Cox et al., 2001). Portions of the seismogenic crust that experience large fluid fluxes host large and vertically extensive vein networks (Sibson et al., 1988), within which up to several millions of m^3 of hydrothermal minerals may deposit from the flowing fluid (e.g. Heinrich et al., 2000; Cox, 2005; Bons, 2001; Garofalo et al., 2002). In contrast, portions of the crust deforming in the absence of significant fluid flow would show scarce evidence of- or no veining, with only synkinematic H₂O-rich minerals within the fault rock attesting to hydrous conditions (cf. Mancktelow and Pennacchioni, 2004;

73 Menegon et al., 2017).

The physical-chemical conditions of fluid-rock interaction in the BDTZ have been extensively studied within exhumed faults
by applying a set of geochemical tools that include fluid inclusion data (Ault and Selverstone, 2008; Garofalo et al., 2014;

76 Siebenaller et al., 2016; Compton et al., 2017), isotopic compositions of fault fluids and mass transfer calculations between host

rock and fault rocks (Goddard and Evans, 1995; Garofalo, 2004; Mittempergher et al., 2014; Spruzeniece and Piazolo, 2015).

78 These data yield important constraints on the PT conditions of fluid-rock interaction within the studied faults, on the source

79 region of the fluids reaching and flowing within the BDTZ and on element mobility during syn-tectonic fluid flow. These

80 studies, however, provide only limited information on the role of fluids on the potentially complex mechanisms that trigger and

81 permit the aforementioned cycles of brittle-ductile deformation. Open questions remain, such as, for example, which Pressure-

82 Temperature-Composition (PTX) conditions are best for a fluid to trigger brittle-ductile deformation cycles in a fault system

83 within the BDTZ, and which fluid property is specifically most effective in controlling the cycles.

In this work, we combine the meso- and microstructural observations with geochemical analysis of fluids, petrographic documentation, microthermometric properties of fluid inclusion assemblages, electron probe microanalyses (EPMA) of fault minerals, Raman spectrometry of fluid inclusions, and electron probe cathodoluminescence imaging to study the effects of numerous cycles of fluid-rock interaction that have occurred in a vein-rich deformation zone from within the BDTZ of the seismogenic region of the Paleoproterozoic continental crust of southwestern Finland. The studied deformation zone belongs to an exhumed conjugate fault system that experienced a complex history of structural reactivation and fluid flow. Deformation zone BFZ300, the target of our study, crops out at c. 426 m below sea level within the deep nuclear waste repository that is

91 presently being built in the island of Olkiluoto (Fig. 1a).

92 We show that this dataset constrains the physical and chemical conditions that prevailed at the time of fault initiation and

93 subsequent cyclic strain localization. The adopted integrated approach provides detailed and new insights into the mechanisms

94 steering deformation within the BDTZ. We propose a mechanical conceptual model that accounts for the constraints derived

95 from our multidisciplinary approach.

96 2 Geological setting

97 The study area is located in southwestern Finland, on the island of Olkiluoto (Fig.1a) within the Paleoproterozoic Svecofennian 98 orogenic province, which is characterized by supracrustal high-grade metamorphic sequences and plutonic rocks. The most





99 abundant lithologies in the study area are variably migmatitic metasedimentary rocks interleaved with metric levels of 100 metavolcanic rocks, calc-alkaline synorogenic TTG-type granitoids, and late orogenic leucogranites (Fig. 1a). For a detailed 101 lithological characterization of the area, we refer the reader to Hudson and Cosgrove (2006) and Aaltonen et al. (2016).

102 Numerous studies carried out on Olkiluoto have highlighted the long geological evolution of the region, which is commonly

103 summarised by tectonic models for the Paleoproterozoic evolution of southern Finland proposing either an evolution during a

104 single and semi-continuous Svecofennian orogenic event (Gorbatschev and Bogdanova, 1993) or, instead, a sequence of up to

105 five distinct accretion events leading to the amalgamation of several microcontinents and island arcs at the margin of the

Archean craton between 1.92 and 1.79 Ga (e.g. Lahtinen et al., 2005). In this scenario, several subduction systems developed, and the collision of the involved microcontinents and island arc complexes resulted in conspicuous continental growth, forming

108 the major part of the Paleoproterozoic domain of the Fennoscandian Shield (1.89-1.87 Ga). According to Lahtinen et al. (2005),

109 this "Fennian accretionary event" ended with a phase of orogenic collapse associated with regional extension and remarkable

110 crustal thinning between c. 1.86 and 1.84 Ga. Renewed compression ensued during collision of the "Sarmatian Plate" with the

111 previously consolidated Svecofennian Shield, causing major crustal shortening, high temperature regional metamorphism

112 (Kukkonen and Lauri, 2009) and the emplacement of S-type granites (e.g. Ehlers et al., 1993). Tectonic activity ascribable to

this orogenic phase ceased with a new orogenic collapse at 1.79-1.77 Ga (Lahtinen et al., 2005).

Pervasive reworking of the Svecofennian domain took place in the Mesoproterozoic when the crust underwent significant stretching and was intruded by voluminous Rapakivi granites and diabase dykes resulting from the widespread melting of the lower crust at c. 1.65-1.50 Ga. This tectonic phase was probably due to the development of a rift along the present Baltic Sea (Korja et al., 2001). Crustal thinning caused also the formation of the "Satakunta Graben", a NW-SE trending graben located c. 50 km to the north of Olkiluoto, which was later filled by Mesoproterozoic sandstone (Jotnian sandstones, Fig. 1a). The latest stage of crustal evolution in southern Finland is expressed by the intrusion of 1.27-1.25 Ga, N-S striking olivine diabase dikes (Fig. 1a; e.g., Suominen, 1991).

121 As to the structural evolution of the study area, the bedrock was affected by complex, polyphase ductile deformation between 122 1.86 and 1.81 Ga. According to the evolutionary deformation scheme proposed by Aaltonen et al. (2010) the results of up to 123 five different phases, referred to as D_1-D_5 , are preserved in the local structural record, each characterised by structures with 124 distinctive mineral composition, metamorphic grade, geometry and kinematics. The most relevant phases to our study are D_2 to 125 D₄. During these ductile episodes, a regional and pervasive NE-SW striking and moderately SE-dipping foliation developed, 126 strain localized along mesoscopic shear zones parallel to subparallel to the foliation and extensive migmatization occurred 127 under amphibolite-facies metamorphic conditions. NNE-SSW and N-S striking mylonitic shear zones also formed under those conditions, whereas later ductile events developed under progressively lower-grade metamorphism until c. 1.7 Ga ago, when 128 129 brittle deformation became the dominant deformation style in response to progressive regional exhumation and cooling (Mattila 130 and Viola, 2014; Aaltonen et al., 2016). The penetrative, inherited ductile grain that by then characterised the crystalline 131 basement and that was suitably oriented with regard to the prevailing stress field was invariably reactivated. This is the case for





- 132 several NNE-SSW striking faults mapped underground in the Onkalo repository, which clearly overprint earlier D4 shear zones 133 and fully exploit the pre-existing ductile precursors. Other faults, however, do not show any clear relation to the older ductile 134 fabric and cut it discordantly. The subvertical, dextral deformation zone we discuss in this paper, BFZ300, is a good example of
- 135 this.

136 In detail, BFZ300 belongs to a set of subvertical, conjugate brittle-ductile to fully brittle strike-slip faults characterized by N-S-137 trending sinistral and NW-SE dextral faults. Both sets show a complex history of reactivation and contain evidence for cyclic 138 and transient switches between brittle and ductile deformation at all scales. Meso- and microstructural studies show that the 139 sinistral faults overprint and probably reactivate a dextral viscous precursor related to earlier, localized ductile deformation. 140 These faults locally contain pseudotachylyte injections, which attest to seismic behaviour during deformation (Menegon et al., 141 2018). In contrast, dextral faults cut across the foliation, do not exploit any ductile precursors and do not host pseudotachylytes. 142 The fault zone studied here belongs to this second group of faults. In the following, we describe its architecture, reconstruct its 143 deformation history and constrain the deformation mechanisms and faulting conditions that acted during its nucleation and 144 subsequent development. The architecture and deformation history of the remarkably different conjugate structure of 145 BFZ300, which is a sinistral brittle ductile deformation zone, whose seismic brittle failure was steered by the presence of a penetrative ductile precursor, will be described in a separate companion paper. 146

147 3 Applied methods

148 Field documentation and sampling were carried out at the underground BFZ300 exposures of Onkalo, which are necessarily 149 limited in extent but that, together with the logged diamond drill holes from the underground exploration, allow a well-150 constrained 3D reconstruction of the local geology. The studied fault section is located at a depth of 426 m b.s.l. (Fig. 1b) and is 151 about 8 m long. To characterize the fault architecture and constrain the spatial and temporal association of fault rocks and the 152 type of fluid involved in the deformation, several outcrop samples, each representative of a distinct structural domain, were 153 collected at the outcrop (TPH-2, TPH-3, TPH-4, TPH-5and TPH-6), in addition to samples PH 21 and PH22 from a diamond 154 drill core that intersects BFZ300 at the same depth in an area that is currently not excavated (Fig. 3). From these samples we 155 prepared 10 petrographic thin sections (samples: TPH120-2, TPH 120-4, TPH 120-6, PH-21 and PH-22) and 9 doubly-polished 156 sections for fluid inclusion analysis (thickness: ~150 µm, samples: TPH120-2, TPH 120-4, TPH 120-6, PH-21 and PH-22). Due 157 to the extensive reactivation of the fault zone and the consequent obliteration of the FI record, FI study was carried out only in 158 samples TPH 120-4, TPH 120-6, PH-21. Hand samples and drill cores localities are specified in Fig. 2. 159 Microstructural work was carried out on oriented petrographic thin sections cut orthogonally to the foliation and parallel to the

- 159 Microstructural work was carried out on oriented petrographic thin sections cut orthogonally to the foliation and parallel to the
- 160 striae that track the overall strike-slip kinematics of the deformation zone. Strie are defined by elongated trails of chlorite
- 161 grains, at the vein-host rock boundary.





162 **3.1 Fluid inclusions and mineral chemistry**

Fluid inclusion measurements were conducted on "fluid inclusion assemblages" - FIAs, i.e. on petrographically discriminated, 163 164 cogenetic groups of fluid inclusions located along trails or (less commonly) within clusters (Bodnar, 2003a; Goldstein, 2003). By definition, FIAs are groups of inclusions that have been trapped together at a specific stage of mineral formation, and, as 165 166 such, give the highest level of confidence when characterizing the properties of trapped fluids and discriminating possible 167 stages of post-entrapment re-equilibration (Bodnar, 2003b, and ref. therein). We identified appropriate FIAs that constrain the deformation history of BFZ300, but also applied the Roedder's identification criteria of FIAs according to the timing of 168 169 entrapment (i.e., primary, secondary, pseudosecondary) in order to link stages of fluid entrapment with stages of brittle and 170 ductile deformation of quartz. In this regard, we can consider FI trails as synkinematic features, where FIAs are entrapped 171 during stages of brittle deformation and fluid circulation, such that FIAs arranged along trails of the same orientation and with 172 similar petrographic features might be representative of the same deformational event.

In the selected samples, we studied 12 FIAs entrapped within two distinct generations of quartz infilling two different generations of veins (named Qtz I and Qtz II, see description below) and exhibiting the least petrographic evidence of postentrapment overprinting by later ductile and brittle deformation, which provided c. 400 microthermometric properties. Due to the well-documented tendency of fluid inclusions to modify their shape, volume, and composition after their initial entrapment even at low deviatoric stress conditions (e.g. Diamond et al., 2010; Kerrich, 1976; Tarantola et al., 2010; Wilkins and Barkas, 1978), working on FIAs that are as similar as possible to those preserving the pristine fluid conditions is essential when aiming at the study of the original physical and chemical properties of the fluid involved in the fault activity.

180 Microthermometric properties of fluid inclusions were determined at the Department of Biological, Geological and 181 Environmental Sciences of the University of Bologna using a Linkam THMSG 600 heating/freezing stage coupled with an 182 Olympus BX51 polarizing microscope. The microthermometry stage was calibrated by using synthetic fluid inclusion samples 183 at -56.6, 0.0, and 374 °C, which correspond to the melting of CO₂, ice melting, and final homogenization of H₂O inclusions, 184 respectively. Obtained accuracies were ± 0.3 °C for final ice melting temperature (Tmice) and ± 3 °C for final homogenization 185 temperature (Thtot). In order to produce an internally consistent dataset, all phase transitions were exclusively collected for 186 individual FIAs and measured by the same standard procedure. Samples were first rapidly cooled to c. -180 °C and then slowly 187 heated to detect the potential formation of a solid carbonic phase, eutectic phases, salt hydrates, ice, and clathrates. The Thtot 188 were later determined in the FIAs by heating the samples from room temperature and recording the mode of homogenization 189 (i.e., by bubble or liquid disappearance). All phase transitions were measured by using the cycling method described by 190 Goldstein and Reynolds, 1994, and care was taken in recording the minimum and maximum values for each assemblage. 191 Volume fractions of individual fluid inclusions determined as % of the ratio $\Phi = Vv/Vtot$ (cf. Diamond, 2003), were estimated 192 optically at room temperature using calibrated charts. Salinity, bulk densities and isochores were computed from the measured 193 Tmice values using the HokieFlincs Excel spreadsheet (Steele-MacInnis et al., 2012 and reference therein).





194 Micro-Raman spectrometric analysis of fluid inclusions was carried out at the Department of Mathematical, Physical and 195 Computer Sciences of the University of Parma (Italy) using a Jobin-Yvon Horiba LabRam spectrometer equipped with He-Ne 196 laser (emission line 632.8 nm) and motorized XY stage. The spectral resolution of the measurements was determined as nearly 197 2 cm^{-1} . The confocal hole was adjusted to obtain a spatial (lateral and depth) resolution of 1–2 µm. Most spectra were obtained 198 with a 50× objective (N.A. 0.75), although for shallow inclusions also a 100× objective (N.A. 0.90) was used. The calibration 199 was made using the 520.7 cm⁻¹ Raman line of silicon. A wide spectral range (100–3600 cm⁻¹) was scanned for each inclusion 200 for the presence of CO_2 , N_2 , CH_4 , and H_2S , but the final acquisitions were made mainly between 1100 and 1800 cm⁻¹ for the study of CO₂ spectra, and between 2500 and 3300 cm⁻¹ for CH₄ and H₂S. The acquisition time for each spectral window was 201 202 120-240 s, with two accumulations. The power on the sample surface is nearly 1 mW but the power on the analysed inclusions 203 has to be considered lower due to reflections and scattering. Analyses were carried out on the vapour bubbles of the fluid 204 inclusions. 205 After the calculation of representative fluid inclusion isochores for each FIA, the pressure corrections were assessed by using 206 the crystallization temperatures of two mineral pairs - namely chlorite-quartz and stannite-sphalerite - as independent input 207 parameters for Qtz I and Qtz II veins, respectively. Chlorite-quartz temperatures were calculated by using the method of 208 Bourdelle and Cathelineau (2015), which assumes quartz-chlorite equilibrium and uses ratios of chlorite end-member activities 209 to link the chlorite compositions with the corresponding formation temperatures through the quartz-chlorite equilibrium

constants. This method is based on the measurements of the concentrations of the major chlorite components (Si, Fe, Mg) and can only be applied to chlorites with $(K_2O + Na_2O + CaO) < 1wt\%$, indeed the case of our chlorites. To estimate the formation temperature of cogenetic sulphides associated with Qtz II we used the stannite-sphalerite formation temperature following the method proposed by Shimizu. and Shikazono (1985). This geothermometer uses the temperature dependency of iron and zinc

partitioning between stannite and sphalerite (Nekrasov et al., 1979) as a useful temperature indicator of the association Qtz II stannite and sphalerite.

216 **3.2 Electron Probe Microanalysis (EPMA)**

217 EPMA of fault minerals were carried out by using a JEOL-8200 wavelength-dispersive electron microprobe housed at the 218 Department of Earth Sciences of the University of Milan, Italy. The instrument fits 5 WDS spectrometers utilizing lithium 219 fluoride (LiFH), pentaerythritol (PETJ and PETH), and thallium acid pthalate (TAP) analysing crystals and an optical 220 microscope. Samples were probed with a beam size of ~1 μ m at 15 keV and 5 nA beam current. Synthetic and natural materials 221 were used as calibration standards at the beginning of each session. Analytical 1- σ errors are typically < 4% for major elements 222 and for the minor elements.

Panchromatic cathodoluminescence (CL) imaging was performed by using the CL CCD detector adjacent to the optical microscope of the JEOL-8200 on the sections used for microstructural work. The electron beam was focused on the sections with an accelerating voltage of 15 kV and 30 nA beam current. Black/white digital images were collected with a 40x





magnification by beam mapping with the CCD detector at a spatial resolution of 1 μ m (beam resolution), which resulted in imaged areas of 27.8 x 22.2 mm. The exposure time for image acquisition was 120 s.

228 4 Results

229 4.1 BFZ300 fault architecture

BFZ300 strikes NNW-SSE and dips very steeply to subvertically to the southwest (Fig. 2a). It cuts through high-grade veined migmatite, interlayered with gneiss and pegmatitic granite. At the studied underground outcrop with a length of 8 m, the fault is a strike-slip fault system formed by two main subparallel fault segments connected by a mesoscopic step-over zone. Subhorizontal striae and kinematic indicators such as chlorite slickensides and R and R' planes indicate invariably dextral kinematics. The most striking mesoscopic characteristic of BFZ300 is the presence in the fault core of a composite set of almost continuous quartz veins (between 1 and 20 cm in thickness) along the entire exposed strike length. A schematic representation

- 236 of the fault zone is shown in Figure 2.
- 237 The fault contains a 0.5-2 m thick damage zone separated by the host rock by two discrete bounding surfaces (Y planes
- 238 according to Tchalenko, 1970 Fig. 2a). The damage zone can be defined in the field on the basis of the presence of a fractured
- volume containing sets of conjugate dextral and sinistral hybrid fractures (Fig. 2b) intersecting to form a tight acute angle of c.
- 240 38° (Fig.2a). Laterally continuous, NNW-SSE striking quartz-filled Mode I fractures (joints) invariably bisect this angle 241 (Fig.2ab), helping to constrain the stress field orientation at the time of fracture formation, with the greatest compressive stress
- 241 (Fig.2ab), helping to constrain the stress field orientation at the time of fracture formation, with the greatest compressive stress
- axis σ_1 parallel to the Mode I fracture strike and oriented c. NNW-SSE. Joints are sharp and have a regular spacing of c. 10 cm. Ouartz infilling the joints and the hybrid fractures of the damage zone forms veins up to 1-1.5 cm in thickness and is referred to
- as Qtz I hereinafter. Its microstructural characteristics are described in detail below (Fig. 2b). Fractures and faults decorated by
- 245 Qtz I have a translucid look that reflect the generally fine grain size of Qtz I (< 1 cm, Fig. 3b.). Locally they are formed by en-
- 246 echelon tensional segments connected by shear planes not decorated by any quartz infill (Fig. 2c). Joints occur also as barren
- 247 fractures defining a penetrative sympathetic fracture cleavage (sensu Basson and Viola, 2004; stereonet of Fig. 2a). In general,
- 248 fracture density within the damage zone tends to increase towards the fault core.
- The fault core is bounded by two main discrete slip surfaces (Y, Figs.2ae and 3a). It contains, and is defined by, two distinct generations of quartz veins (Fig. 2d) that are interrupted and offset laterally by a metric step-over zone (Fig. 2e-g). The main quartz vein of the core is infilled by quartz exhibiting the same microstructural characteristics of Qtz I in the damage zone; we therefore refer to it as a Qtz I vein. It is accompanied by a younger, subparallel vein formed by a milky-white type of quartz with a significantly larger quartz grain size than Qtz I (>1 cm) that we refer to as Qtz II (Figs. 2deg and 3). Locally, pockets of cataclasite and breccia are also observed along and inbetween the two veins. Microstructural analysis reveals that they formed at the expense of the host gneiss (Fig. 3bd). The Qtz II vein shows a quite irregular, curved geometry (Figs. 2d and 3c) and a
- 256 variable thickness up to a maximum of c. 20 cm. The minimum Qtz II vein thickness coincides spatially with a lateral apparent
- 257 displacement of the vein. The BFZ300 core varies in thickness between 20 and 30 cm along most of the exposed fault length,





but becomes thicker (up to 50 cm) in the compressional step-over zone that connects the two fault segments that are offset laterally by c. 1 m. The step-over zone is defined by synthetic T fractures (Figs. 2eg) and contains a decimetric brecciated lens

- 260 (Fig. 2e). T fractures are generally filled by Qtz I veins (Fig. 2f).
- 261 Chlorite is present as a secondary phase, with a modal abundance between 5 and 10 vol% in both Qtz I and Qtz II veins. In Qtz
- 262 I veins it occurs as euhedral/subhedral crystals that are up to 1-2 mm in size. Chlorite is present mostly as a disseminated,
- 263 interstitial phase, concentrated mainly in the internal part of the vein. In the Qtz II vein, however, it occurs as elongated crystals
- 264 (5-8 mm in length) arranged perpendicularly to the walls of the vein, which suggests orthogonal dilation at the time of opening
- 265 (Fig. 3b). The Qtz II vein contains also small (1-2 cm) aggregates of sulphides (sphalerite, pyrite, galena, and chalcopyrite)
- 266 mainly concentrated in the central part of the vein (Fig. 3b). Qtz II-sulphide mineral associations and their textures are 267 described in detail below.
- As observed in the field, the presence of Qtz I veins along the joints in the damage zone and the continuity of the fault core Qtz
- 269 I vein suggest Mode I fracturing during Qtz I emplacement (Fig.3ac). The semi-continuous parallelismQtz I and Qtz II veins in
- the fault core, combined with the location of the Qtz II vein along the walls of the Qtz I vein, suggest the partial reactivation of
- 271 the Qtz I vein during Qtz II emplacement. Dilation leading to Qtz II emplacement exploited and further reworked the Qtz I-host
- 272 rock contact, that seemingly had lower tensile strength than the pristine migmatite. The reconstructed time relationship between
- the two vein generations is also consistent with local evidence of the Qtz II vein partially crosscutting parts of the Qtz I vein
- 274 (Fig. 3a).

275 4.2 BFZ300 microstructural analysis

276 In the following we provide a description of the microstructural characteristics of BFZ300 by detailing our findings and 277 observations separately for the main structural domains of the fault zone.

- 278
- 279 4.2.1 Damage zone

280 Qtz I veins within the damage zone cut across the migmatitic host rock and form the infill of conjugate sets of hybrid fractures, 281 which, when studied at the microscale, appear as formed by dilatant segments joined by cataclastic shear fractures (Fig. 4a). 282 Shearing is well documented by the asymptotic bending into the shear surfaces of foliation planes formed by the alignment of 283 chlorite and muscovite, both partly altered to sericite and chlorite, respectively (Fig.4a). Qtz I infilling the tensional segments 284 has an average grain size between 200 µm and 3 mm and exhibits a rather heterogeneous texture, from purely blocky to mixed 285 elongated-blocky (Figs. 4bc). The largest crystals (800 µm to 1 mm) are elongated and stretch from the vein walls towards the 286 inner part of the vein (Figs. 4c and 5a), which is consistent with a syntaxial growth mechanism (Bons et al., 2012). At least two 287 episodes of vein growth/renewed dilation, as indicated by the presence of median lines (ML), are clearly visible within one of 288 the studied veins and confirm a syntaxial growth mechanism for the vein (Fig. 5). Medial lines are defined by the alignment of 289 chlorite, sericite, and carbonate aggregates (Fig. 5bc). Blocky euhedral quartz crystals are also found, with a grain size between





290 300 to 600 μ m. These crystals are juxtaposed to very fine grained quartz (<200 μ m) within sericite-rich cataclastic bands 291 (Fig.4b). These cataclasites contain also hydrothermally alterated host-rock fragments including pervasively altered K-feldspar-292 bearing lithic fragments and phyllosilicates.

With the exception of the blocky variety, Qtz I crystals exhibit various degrees of viscous deformation and recovery. They
contain widespread evidence of undulose extinction and extinction bands, indicating distributed internal plastic deformation.
Cathodoluminescence imaging of Qtz I shows the presence of a diffuse network of healed quartz microfractures (Fig. 4d),

296 which demonstrates healing subsequent to brittle deformation and fracturing.

297 Chlorite occurs as a disseminated phase within the Qtz I veins of the damage zone and in textural equilibrium with quartz. It has

a peculiar vermicular texture (Fig. 5b), crystal dimensions of about 100 μ m, and displays interference colours ranging from violet to Berlin blue. Vermicular chlorite forms small pockets mainly located in the central part of the veins and at the triple

300 junctions of blocky quartz crystals.

301 4.2.2 Fault core

302 In the BFZ300 fault core, the grain size of Qtz I is strongly variable within the vein, suggesting the presence of heterogeneous 303 and complex structural sub-domains of deformation. Qtz I has the smallest observed grain size (range: 30-800 µm, Fig. 6a) and 304 documents multiple and cyclic episodes of mutually overprinting brittle and ductile deformation leading to a complex 305 microstructural record. The earliest post-vein emplacement recognised deformation stage is reflected by the low-temperature, 306 intracrystalline deformation of the largest crystals (400-800 µm in size). Typical microstructures, such as undulose extinction, 307 wide extinction bands (WEBs, Derez et al., 2015), and bulging along grain boundaries are the most common microstructures 308 ascribable to this deformation stage (Fig. 6ab). A distinct, brittle deformation event is documented by narrow intracrystalline 309 fractures that crosscut the largest quartz crystals (Fig. 6bc), and which locally contain new grains of quartz ranging in size 310 between 20-100 μ m (Fig. 6d). More in detail, these new grains form parallel bands that are oriented at low angle (<30°) to the 311 vein walls and that can be up to 2 mm in length and 200 µm in thickness. Plastically deformed Qtz I crystals hosting these 312 intracrystalline bands of new grains are cut across by a distinct set of subparallel intercrystalline fractures, which are interpreted 313 as the expression of yet another deformation event that occurred under overall brittle conditions. These fractures are parallel to 314 the strike of BFZ300 and are in turn healed by new quartz grains (grain size: $50-150 \,\mu$ m), and can be up to 2.5 cm in length and up to 500 µm in width (Fig.6a). Their cathodoluminescence imaging shows that they are sealed and healed, yielding an 315 316 homogeneous dark cathodoluminescence signal.

Qtz II within the fault core is typically coarse grained (individual crystals: 300 µm-1 cm in size) and exhibits a regular blocky texture devoid of any shape or crystal preferred orientation (Fig. 7a). Locally, these large crystals display primary growth textures, such as primary FIAs oriented parallel to specific crystallographic planes. With the exception of undulose extinction, Qtz II does not show clear evidence of plastic deformation, although cathodoluminescence imaging of optically continuous Qtz

321 II has also shown that a dense network of healed quartz microfractures locally crosscuts Qtz II crystals (Fig. 7c). These are





relatively thin (hundreds of μ m thick) networks that are poorly visible to invisible by standard petrographic analysis. The only petrographic evidence for these healed microfractures within quartz are well defined trails of fluid inclusions crosscutting

- 324 primary growth bands (Fig. 7d).
- 325 Chlorite is the second most abundant phase within the fault core Qtz I and Qtz II veins and occurs with a variety of textures.
- 326 Aggregates of vermicular chlorite similar to that occurring in the damage zone are also present in the Qtz I from the core (Fig.
- 327 8e), although chlorite with flaky and radiate textures (Fig. 8f) is also present. The latter type is generally 100-300 μ m in size
- 328 and is in textural equilibrium with quartz and rare calcite. Radiate chlorite overgrowing fractured Qtz II (Fig. 7bc) suggests late
- 329 Qtz II precipitation. Associated with Qtz II, a sulphide assemblage made of pyrite, sphalerite, galena, and chalcopyrite (Fig.
- 330 7de, see also Fig. 3b) forms aggregates commonly located along quartz grain boundaries. In the studied sections, these
- 331 aggregates have dimensions between 10 and 600 µm. Chalcopyrite occurs as µm sized irregular inclusions within sphalerite
- forming the typical "chalcopyrite disease" texture (e.g., Barton and Bethke, 1987; Fig. 7e).
- 333 Multiply reworked breccias and cataclasites crosscut BFZ300. In the studied sections, a cataclastic band between 5 and 8 mm
- thick crosscuts both Qtz I and Qtz II veins (Fig. 8a), but is in turn crosscut by a different quartz-radiate chlorite vein displaying
- 335 evidence of syntaxial growth. This cataclasite contains poorly sorted and angular quartz clasts varying in size between 8 and 12
- 336 mm set in a finer (20-200 µm in size) white mica-quartz matrix. The largest quartz fragments show irregular, lobate grain
- 337 boundaries and are affected by undulose extinction. We interpret these textures as the product of dissolution and cataclastic
- 338 reworking of a previous generation, plastically deformed quartz.
- 339 Parallel sets of stylolitic seams strike N-S parallel to the strike of BFZ300 and mark the two sides of the cataclastic band (Fig.
- 340 9ac). They host anhedral sphalerite, stannite, galena, pyrite, and chalcopyrite (Fig. 9d), which are coeval with the formation of
- 341 Qtz II vein. We interpret the presence of these anhedral sulphide minerals along the stylolite as the product of passive
- 342 concentration by pression-solution processes. We use the stannite-sphalerite mineral pair as a geothermometer for Qtz II
- 343 emplacement.

344 4.3. Fluid inclusion data

345 The studied FIAs contain invariably a two-phase fluid (liquid-vapour) and are mainly arranged in secondary trails within Qtz I

346 crystals in the damage zone (Type S1) and also within Qtz I fault core, where they form dismembered (Type S2) trails and also

347 appear as individual clusters inside the crystals affected by viscous deformation (Type-S3). Within the Qtz II fault core, FIAs

- 348 are arranged as pseudosecondary (Type PS) and secondary (Type S4) trails. Representative examples of FI petrographic
- 349 features are shown for each BFZ300 structural domain in Fig. 9. Table 1 gives a schematic representation of the location of the
- 350 FI types presented above, in addition to their location in the fault architecture and their fluid properties.
- 351 Damage Zone: Within Qtz I grains (Fig. 9ab), secondary FIAs are found as trails (Fig. 9a) that parallel what we interpret as
- 352 healed, old intracrystalline microfractures. These microfractures are likely to be old joints and hybrid fractures whose
- 353 orientation mimics that of the mesosopic BFZ300 structural features. In these assemblages, FIs have a maximum size ranging





between 2 and 20 μ m, a regular equidimensional shape (i.e. negative crystal morphology), and relatively uniform φ of 5-15% (Fig. 0b)

355 (Fig. 9b).

356 Fault Core: Qtz I grains host secondary FIAs, which are transgranular trails (Fig. 9c) representing healed joints and hybrid 357 fractures. These trails are locally interrupted and dismembered by aggregates of new, fine-grained quartz grains (Fig. 9c), and 358 generate a texture that is indeed typical of Qtz I from the fault core (cf. Fig.6a). Fluid inclusions entrapped along these trails 359 (Type S2) vary in size between 1 and 10 μ m, have a ϕ of 10-20%, and show a negative crystal morphology (Fig. 9d). Fluid 360 inclusions are also found as isolated clusters inside intensely recrystallised quartz domains (Fig. 9c). FIAs inside these 361 recrystallized quartz domains were pervasively obliterated during later episodes of ductile deformation. The development of 362 WEBs, intercrystalline bands and bulging (cf. Fig. 6) resulted in the transposition of these assemblages is invariably observed and is documented, for instance, by the presence of short, secundary trails of regularly shaped inclusion oriented at a high angle 363 364 with respect to a longer, parent trail (Fig. 9c). Morphologically, these trails resemble the transposed trails documented in highgrade metamorphic rocks (Andersen et al., 1990; van den Kerkhof et al., 2014). Different types of fluid inclusion morphologies 365 366 are found within the intensely recrystallized quartz domains (Fig. 9f). Negative crystal morphology is observed in some areas of 367 the selected samples, but it is uncommon. More typical is instead the "dismembered" morphology (cf: Vityk and Bodnar, 1995; Tarantola et al., 2010), which is observed in the relatively large inclusions (> 20 μ m). This morphology is made of a central 368 369 (often empty) inclusion, showing several tails and re-entrants, surrounded by a three-dimensional clusters of small "satellite" inclusions. These clusters might be arranged with a quasi-planar geometry inside the host (i.e. in a trail-like fashion). Another 370 371 typical texture found in most assemblages is the "scalloped" morphology of small- to medium-sized inclusions (<10-15 μm), 372 which is defined by the presence of indentations, embayments, irregularities, and sharp tips of the inclusion walls (Fig. 9f). 373 Small inclusions ($<1 \mu m$) are also found at the edge of the straight, regular boundaries of new quartz grains; they are mostly 374 dark, i.e. they are vapour-rich or empty, and are equant in shape (Fig. 9e). Although small inclusions do not allow a 375 microtermometric study of the fluid-phase behaviour in this structural domain, they confirm the complex reactivation history of 376 BFZ300.

Qtz II contains both pseudosecondary (Type-PS) and secondary (Type-S4) assemblages (Fig. 9g-j). The first type is arranged in trails, that cut at low angle the hosting quartz but not the neighbouring phases (e.g., chlorite, Fig. 9g). In these assemblages, FIs are relatively large (2-45 μ m). They show elongated shape and their ϕ varies between 15 and 30 % (Fig. 9h). Type-S4 FIAs (Fig. 9i) have two-phase inclusions whose size (5-35 μ m) is similar to that of PS trails, but show a ϕ between 30 and 40 % (Fig. 9j).

Rare primary FIAs are also present along growth planes of Qtz II and are best observed predominantly in the least deformed Qtz II crystals, where they have a relatively large size (20-50 µm). Elsewhere they present irregular and "dismembered" textures, which suggest intense post-entrapment re-equilibration.





In summary, our microtextural study shows that the FIAs to be selected for the microthermometric study are only those hosted within Qtz I and Qtz II crystals with the minimum degree of recrystallization and whose inclusions have textures corresponding

- 387 to the least intense post-entrapment re-equilibration (Bodnar, 2003b, and ref. therein; Tarantola et al., 2010). These are the
- pseudosecondary and secondary FIAs in which dendritic or transposed inclusions are absent, and in which the host quartz
 exhibits only undulose extinction (S1, S2, S4 and PS).
- 390 The majority of the secondary FIAs hosted within Qtz I from the damage zone show a range of Tmice between -5.9 and 0 °C,
- 391 which corresponds to a salinity of 0-9 wt% NaCleq (Fig. 10a). In these FIAs, final homogenization occurs by disappearance of
- 392 the vapour bubble and mainly between 150-330°C (Fig.10e). The secondary FIAs hosted within Qtz I of the fault core (Type-
- 393 S2) show a range of Tmice between -3.9 and 0 °C, which corresponds to salinities between 0 and 6 wt% NaCleq (Fig.10b), and
- 394 final homogenization by bubble disappearance is between 150 and 420°C (Fig.10f).
- 395 The Type PS FIAs show a range of Tmice between -11 and 0 °C, which corresponds to a salinity range of 0-15.2 wt% NaCleq
- 396 (Fig.10c), and Thtot by bubble disappearance is comprised between 150 and 440°C (Fig.10g). The Type S4 FIAs have a range
- of Tmice between -7.3 and 0 °C, which corresponds to a 0-10.9 wt% NaCleq range of salinity (Fig.10d), while final homogenization by bubble disappearance is comprised between 150 and 430°C (Fig.10h). Final homogenization occurs always
- 399 by bubble disappearance.

400 Raman spectra from the vapour phase of fluid inclusions hosted by the Qtz I and II show peaks at the characteristic 401 wavenumbers of CH₄ (2917 cm⁻¹), and CO₂ (1388 cm⁻¹). These peaks were determined as weak in all spectra, and CO₂ detection 402 was only sporadic in one of the sample of the fault core (TPH-120 4A). Such spectroscopic determinations are consistent with 403 the lack of microthermometric evidence for CO₂ or CH₄ occurrence in the FIAs, i.e., with the failure to detect melting of a 404 carbonic phase or clathrate hydrates during the freezing experiments (cf. Rosso and Bodnar, 1995; Dubessy et al., 2001). The 405 impossibility to detect CO₂- and CH₄-bearing fluids via microthermometric determinations indicates a gas pressure in the 406 analysed inclusions that is systematically lower than that required to observe clathrate dissociation (e.g., 1.4 MPa in CO₂-H₂O 407 fluids, Rosso and Bodnar, 1995), i.e. it shows low gas concentrations. Considering the broad salinity range of 0.1-14 wt% NaCl_{co} for the BFZ300 fluids (which corresponds to NaCl concentrations of 1.7 $10^{-3} - 2.4$ M), we cannot estimate a maximum 408 409 CH₄ concentration.

410 **4.4 Chlorite and sulphide geothermometry**

411 Chlorite composition has been determined for several generations of chlorites in association with Qtz I and II, namely

- 412 vermicular chlorite associated with Qtz I from the damage zone, vermicular and radiate chlorite associated with Qtz I from the
- 413 fault core, and radiate chlorite associated with Qtz II from the fault core (Table 2). The EPMA data show that, in general, the
- 414 BFZ300 chlorites are Fe-rich (XFe = Fe/(Fe+Mg) between c. 0.65 and 0.90), have concentrations of (Na₂O+K₂O+CaO) <1
- 415 wt%, and result mainly from a solid solution of the sudoite and daphnite end-members, i.e., of Mg₂Al₃(Si₃Al)O₁₀)(OH)₈ -
- 416 (Fe,Mg)₅Al(Si,Al)₄O₁₀(OH)₈. The dataset shows in particular that the compositions of the distinct chlorite generations vary





417 systematically with vein generation, as shown by the vermicular chlorite associated with Qtz I veins (Fig. 8e) with a XFe 418 between 0.65 and 0.67 and the radiate chlorite associated with Qtz I from the fault core and Qtz II (Figs. 8f and 7b) with a XFe

- 419 between 0.76 and 0.86.
- Temperature-composition relationships for the quartz-chlorite pair portrayed in the T-R²⁺-Si diagram of Bourdelle and 420 421 Cathelineau (2015) show that, in the hypothesis of quartz-chlorite equilibrium, the precipitation of vermicular chlorite within 422 the Qtz I of the damage zone took place in the 175-250 °C range (green diamonds of Fig. 11a). This range is distinct from that 423 of the vermicular and radiate chlorite from Qtz I of the fault core, which is probably >350 $^{\circ}$ C because the measured R²⁺–Si 424 compositional parameters ($R^{2+} = Mg + Fe$) plot at the edge of, or slightly outside, the calibrated region of the Bourdelle and 425 Cathelineau plot (red diamonds of Fig. 11a). We stress that the high-T chlorite plots parallel to the 350 °C isotherm, suggesting 426 that it precipitated most probably at the same, or at a similar, temperature. Radiate chlorite associated with Qtz II from the fault 427 core is instead compatible with an equilibrium precipitation at 160-200 °C (light-blue diamonds in Fig.11a).
- The collected EPMA data show that the sulphides associated with Qtz II have compositions that approach those of pure phases (Table 3). Pyrite has trace element concentrations (Cu, As, Pb, Ni, Zn) that are in general below the EPMA detection limit, while galena, sphalerite, and chalcopyrite show only some significant trace contents of Fe and Zn (e.g., Fe: 0.22-1.00 wt% in galena; Zn: 0.11-3.95 wt% in chalcopyrite). Pyrite and sphalerite from the Qtz II veins (Fig. 7e) have trace element concentrations that are, again, mostly below detection limits.
- 433 The stylolites bordering the cataclasite bands described above and formed at the contact between the Qtz I and Qtz II vein 434 contain pyrite, galena, and the sphalerite-stannite pair (Fig. 8acd), with the latter showing the largest compositional variation. 435 This pair represents a mineral geothermometer because the partitioning of Zn and Fe between sphalerite and stannite was 436 demonstrated to be temperature dependent but pressure independent (Nekrasov et al., 1979; Shimizu, M. and Shikazono, 1985). 437 In the fourteen analysed pairs, stannite shows a range of Zn concentrations varying between 0.48 wt% and 3.25 wt%, while 438 those of Fe, Cu and Sn vary within narrow ranges (Fe: 12.74±0.56 wt%; Cu: 28.30 ±0.33 wt%; Sn: 27.65 ±0.71 wt%). 439 Sphalerite in the pair has concentrations of Fe and Zn of 7.63±0.87 wt% and 56.68 ±1.17 wt%, respectively. These ranges allow 440 the calculation of the partition coefficient (K_D) of the reaction: Cu_2FeSnS_4 (in stannite) + ZnS (in sphalerite) = Cu_2ZnSnS_4 (in 441 stannite) + FeS (in sphalerite). We have used the $\log K_D$ -T relationship of Shimizu and Shikazono (1985) to calculate the formation temperature of the pair, which is portrayed in the (X_{Cu2FeSnS4}/X_{Cu2ZnSnS4})-(X_{FeS}/X_{ZnS}) plot of Shimizu and Shikazono 442 443 (Fig. 11b). The resulting 220-305 °C interval lies at the low end of, or slightly outside, the 250-350 °C interval of the 444 geothermometer. Therefore, while the 250-305 °C interval can be taken as an estimation of the formation T of sphalerite-445 stannite in the stylolite, the 220-250 °C interval should be taken with caution.

446 5 Discussion

447 Our work constrains the structural architecture and the environmental conditions at which BFZ300 deformation took place.
 448 Field and petrographic observations support the idea of transiently elevated fluid pressures, cyclic frictional-viscous





deformation and progressive, yet discrete strain localization (Figs. 2 and 3). Analytical data suggest that these deformation cycles took place at the BDTZ. In the following, we discuss these constraints by systematically considering our different analytical results.

452 **5.1. Fluid inclusion data and mineral-pair geothermometry**

- 453 Microthermometric and Raman spectrometry data show that the fluid entrapped within the studied FIAs at the time of formation
- 454 of the damage zone and fault core during precipitation of Qtz I and Qtz II veins can be represented by a H₂O-NaCl model fluid.

455 The fluid was in a homogeneous state at the time of entrapment, as testified by the consistent final homogenization by bubble

456 disappearance. It also had a low bulk salinity, as shown by the distribution of >80% of the ice melting (Tmice) measurements

457 skewed towards values of -3 °C or higher, which corresponds to bulk salinities of 5 wt% NaCl_{eq} or less (Fig. 10a-d).

458 The documented Thtot varies between c. 150 and 430 °C without a single mode or a skew (Fig. 11e-h). Such distribution shows

- that no common range of entrapment temperatures can be identified in the entire dataset, and that the properties of individual,
- 460 well-identified FIAs do not correspond to those of well-preserved assemblages. Indeed, the ranges of Thtot in individual FIAs
- 461 are typically of the order of 150-200 °C (Fig. 10e-h), i.e. a value that is much higher than the ~10 °C range expected for
- 462 homogeneous FIAs entrapped isochorically and isoplethically (Fall et al., 2009; Vityk and Bodnar, 1998) and that demonstrates

463 post-entrapment re-equilibration (cf. Bodnar, 2003b; Sterner and Bodnar, 1989).

A major implication of fluid inclusion re-equilibration in our study is that the calculated fluid properties do not rigorously reflect those of the pristine fluid originally entrapped within BFZ300, but rather that of a fluid that modified its properties during the fault activity. This is comparable to the results of other fluid inclusions studies from faults (Boullier, 1999; Garofalo

467 et al., 2014; Roedder, 1984).

468 In spite of these difficulties, a possible approach to interpret our FI dataset is the comparison with the experimental work on

- synthetic fluid inclusions subjected to a range of post-entrapment re-equilibration conditions (Bakker, 2017; Bakker and Jansen,
- 470 1990, 1991, 1994; Vityk and Bodnar, 1995, 1998; Vityk et al., 1994). Such comparison is in our case difficult because most
- 471 experimental work was carried out at high TP conditions (500-900 °C; 90-300 MPa) and also because only few experiments 472 were carried out under deviatoric stress conditions that approach those of natural rocks (Diamond et al., 2010; Tarantola et al.,
- 473 2010). Despite these limitations, however, some key experimental results provide fundamental constraints on our dataset. First,
- 474 both hydrostatic and uniaxial compression experiments showed that in each re-equilibrated FIA a number of inclusions survive
- 475 virtually intact the modified post-entrapment PT conditions, showing that only severe deformation brings to total re-
- 476 equilibration and complete obliteration of pristine inclusions (i.e., $\Delta \sigma > 100$ MPa in uniaxial compression experiments; >400
- 477 MPa change of confining P in hydrostatic experiments). Second, under conditions leading to only low to moderate re-
- 478 equilibration, the bulk chemical composition of the fluid inclusions does not change significantly from that of the pristine

479 inclusions.





480 All of this implies that natural quartz samples with microstructures typical of moderate T deformation, such as deformation 481 lamellae, deformation bands, undulose extinction and bulging, and hosting FIAs with moderately re-equilibrated textures, 482 should still contain a number of inclusions whose properties resemble those of the pristine fluid. Thus, it is possible that an 483 aliquot of the 1-5 wt% NaCleq FIAs from Qtz I and II crystals from both the damage zone and fault core is still representative 484 of the pristine sampled fluid. These inclusions would be those that survived or were relatively less affected by deformation 485 events postdating their entrapment. Inclusions falling outside the most typical 1-5 wt% NaCleq salinity range would instead 486 correspond to those that experienced significant H₂O loss and consequent salinity increase during the successive stages of fault 487 deformation (cf. Bakker and Jansen, 1990; Diamond et al., 2010). The large documented range of Thtot lacking a specific mode 488 observed in individual FIAs is the product of fluid density changes caused by fluid inclusion re-equilibration during post-489 entrapment deformation. This would have happened repeatedly and cyclically within the host quartz during all ductile and 490 brittle stages of deformation of the multi-stage deformation history of BFZ300. The cumulative effect of these cyclic events on 491 fluid inclusions causes the impossibility to determine the exact T_f and P_f at entrapment; however, the ranges of PT values that 492 approach the true PT conditions can be assessed with some degree of confidence. Therefore, we suggest that the 200-350 °C 493 Thtot interval (Fig. 10 e-h) of the 1-5 wt% NaCleq fluid might reflect the characteristic range of the vast majority of damage 494 zone and fault core fluid. This value overlaps with the 175-350 °C range estimated with the quartz-chlorite geothermometer 495 (Fig. 11a) and the 220-305 °C range estimated with the sphalerite-stannite geothermometer (Fig. 11b).

496 Fully aware of these limitations, we have combined the microthermometric data of the studied FIAs with the independent 497 quartz-chlorite and sphalerite-stannite geothermometers to constrain the most probable Pf during faulting events. With this 498 approach, we use the formation temperatures of the mineral pairs as independent geothermometers and consider the intersection 499 between these values and the FIA isochores to derive the ranges of trapping pressure (cf. Roedder and Bodnar, 1980). In Fig. 12, we present the P_f ranges that are calculated using the entire salinity range of the studied FIAs (cf. Fig. 10); however, we 500 501 highlight the most probable P_f ranges that are consistent with what we consider best preserved salinity range (0-5 wt% NaCleq). 502 Accordingly, for the damage zone we estimate a P_f interval of 20-90 MPa (Fig.12a) by intersecting the range of T obtained 503 from the chlorite-quartz pair in the damage zone Qtz I (T c.170-240 °C, Fig. 11a) with the range of isochores from the same 504 quartz. As to fluid pressure estimations in the fault core, we combine the 350 °C obtained from the chlorite-quartz pair from the 505 fault core Qtz I (T>350 °C are outside the calibrated range of the geothermometer) with the ranges of isochores from the same 506 quartz, from which we obtain P_f ranging between c. 140 and 120 MPa (Fig. 12b). Similarly, the intersection between the 507 equilibrium T of the sphalerite-stannite pair in the Qtz II fault core (250-305 °C) and the range of isochores of the Type PS FIAs of Qtz II (Fig.9) defines Pf values ranging between 10 and 140 MPa (Fig.12c). Estimations from Type-S4 FIAs (Fig.9) 508 509 constrain a range between 40 and 160MPa (Fig. 12d). We propose that these values are sufficiently accurate to constrain multiple stages of fault slip, each one triggered by a fluid pulse having a distinct pressure. Hence, fault activity started at 200 °C 510 and at P_f varying between 20 and 90 MPa and continued through higher temperatures (305-350 °C) and P_f (120-160 MPa). 511





512 **5.2. Structural evolution and fluid flow**

513 Based on the integration of field, microstructural, thermometric and fluid inclusions constraints (Table 1), we propose a 514 conceptual model for the structural evolution of BFZ300 (Fig. 13). The fault's finite strain results from several slip episodes 515 mediated by multiple events of fluid ingress and fluid-rock interaction. A first constraint provided by our study is that the bulk 516 chemical composition of the fluids that flowed within the fault did not change significantly during the documented fault 517 activity, as the best preserved 0-5 wt% NaCleq salinity range points to a compositionally homogeneous fluid. This suggests a compositionally homogeneous source region of the fluid, or, alternatively, that the studied section of the fault did not interact 518 519 with fluids of substantially different composition. 520 The embrittlement of the Olkiluoto metamorphic basement (time t_1 of Fig. 13ab) represents the initial stage of the deformational 521 history of BFZ300 when conditions for brittle dilation and fracturing of the Paleoproterozoic basement were first met in a 522 transien fashion. We propose that brittle failure under still ductile environmental conditions was caused by transiently elevated 523 $P_{f_{r}}$ as demonstrated by field evidence of hydrofracturing (Figs. 2 and 3) and the pore pressure estimations (Fig. 12 and Table 1). 524 Hydrofracturing of the host basement is also expressed by the emplacement of Otz I veins along the diffuse network of joints 525 and conjugate hybrid/shear fractures of the damage zone (Figs. 13a and 2g). These brittle features are quite evenly distributed 526 within the damage zone suggesting initial volumetrically diffuse strain distribution. Their formation caused the overall 527 mechanical weakening of the actively fracturing host rock volume, which in turn facilitated later strain localization. Brittle 528 structures formed during this stage are discordant to the ENE-WSW striking metamorphic foliation (Fig. 1b), which they cut at 529 high angle (Fig. 13a). Conditions for tensional and hybrid failure require low differential stress, i.e. σ_1 - σ_3 ~ 4T, where T is the 530 tensional strength of the rock. Opening of fractures caused a stress drop, sudden increase of permeability, fluid venting and 531 inhibited further build-up of P_f. Dilatant fractures were partially infilled by Qtz I, precipitated from a first pulse of the low-532 salinity fluid. Crystallization of Qtz I and formation of veins within these fractures caused hardening of the system. The 533 progressive recovery of shear stresses concomitant with the progressive sealing of dilatant fractures altered the overall 534 background stress conditions such that failure, after causing initial pure dilation, was accommodated by hybrid extensional 535 failure and, eventually, by shear fracturing (Fig. 13b), thus forming laterally continuous and interconnected shear fractures associated to breccia pockets and cataclasites (Figs. 2e and 3bd). Conjugate shear fractures connected the previously formed 536 537 extensional fractures through a fracture coalescence mechanism (e.g. Griffith, 1921; Fig. 13a). At the micro-scale this is 538 demonstrated by the elongated blocky texture of Qtz I crystals from the damage zone (Fig. 11), where crystals grew at high 539 angle to the vein boundaries (thus suggesting initial near-orthogonal dilation) and are physically connected by cataclastic shear bands to form a fault-fracture mesh (e.g.Sibson, 1996; Figures 4ab). Cataclastic bands formed at the expense of the migmatitic 540 541 host rock are enriched in authigenic, synkinematic sericite, likely due to the interaction between K-feldspar and fluids 542 circulating in the dilatant fault zone (Fig. 4b). Shear fractures thus deformed the migmatitic host rock to connect dilatant and 543 mostly Qtz I-filled tension gashes during a continuum of deformation. The conjugate shear fractures ascribable to this stage





544 invariably define tight acute angles (Fig. 2b), which we take as further evidence of overall low differential stress conditions at 545 the time of failure (Fig. 13b).

546 In synthesis, Qtz I veins from the damage zone are interpreted as the expression of the earliest stage of fault nucleation, before

547 strain localization affected a progressively narrower rock volume to eventually form the main fault core. Indeed, Qtz I in the

(548) damage zone preserves mostly brittle microstructures and lacks a pervasive ductile overprint, which is instead prevalent within

(549) the fault core. As a consequence, we interpret the chemical properties of the fluid derived from these veins as the closest to the

550 initial conditions of the first fluid involved in BFZ300 nucleation. Fluid inclusion and geothermometric estimations from the

sinkinematic chlorite crystals associated with the damage zone Qtz I (Figs.5a and 11a), suggest chlorite precipitation at a T of c. 200° C and P_f between c. 90 and 20 MPa at the time of fault nucleation. Based on geometric, kinematic and deformation style

characteristics, we assign this deformation episode to Stage 1 by Mattila and Viola (2014) (their Fig. 18), i.e. to a discrete brittle

episode that they consider the expression of the earliest onset of brittle conditions in southwestern Finland c. 1.75 Ga ago, under

555 overall NW-SE to NNW-SSE transpressive conditions.

556 Further deformation of the BFZ300 (time t₂ of Fig. 13c) occurred by progressive inward strain localization and narrowing of the 557 actively deforming volume of the deformation zone (from a wide damage zone to a narrow fault core). The early BFZ300 core, 558 consisting of the main Qtz I vein is interpreted as having formed at this stage, within an overall dextral strike-slip kinematic 559 framework. Emplacement of the Qtz I vein in the core represents the last pulse of this brittle deformational episode (Fig. 13b). 560 Major fluid venting was likely associated with it, such that the system, once brittle failure in the core had occurred by 561 hydrofracturing, moved back to a more diffuse deformation style typical of the still prevailing ductile conditions. Microscopic evidence of ductile deformation by dynamic recrystallization (Fig. 6ab, Table 1) overprinting the early brittle structures of Qtz I 562 563 in the fault core supports slow strain rate conditions during deformation. However, this viscous background deformation was punctuated by renewed and cyclically transient embrittlement as documented by healed fractures shown by trails of secondary 564 565 fluid inclusions cutting across both the ductile fabrics and the earlier brittle deformational features (Fig.6c, d and e). Repeated 566 pulses of high P_f (120-140 MPa) likely triggered these brittle-ductile oscillations. Repeated fluid ingresses and related deformation would, in addition, also have caused some of the post-entrapment equilibration of the FI, as discussed above. 567

568 The cycles of brittle and viscous deformation may be explained as follows. Cyclic brittle failure would have repeatedly lowered 569 $P_{f_{t}}$ which lowered the background stress and strain rate and favoured ductile deformation by dynamic recrystallization at T > 570 300 °C between the slip events (e.g. Passchier and Trow, 2005). The fault regained cohesive strength after each brittle failure 571 episode through vein formation and sealing/healing of the fracture networks. Porosity destruction by mineral crystallization and 572 fracture sealing, as clearly shown by CL imaging (Fig 4d), induced a progressive reduction of permeability and mechanical 573 healing of the fault, which promoted an increase of P_f and ultimately triggered a new brittle failure. Therefore, pore pressure 574 build-up promoted episodic brittle fracturing followed by cementation and plastic deformation/recovery. The compelling evidence for this deformation occurring at $T \approx 350^{\circ}$ C indicate that the described processes identify the BDT of the quartz-575

576 feldspathic crust (Kohlstedt et al., 1995).





577 Mattila and Viola (2014) described a second brittle stage (referred to as Stage 2, their Fig. 18) during which a c. N-S to NNE-578 SSW-oriented episode of transpressional deformation affected southwestern Finland. Geometric and temporal relationships 579 between structures of Stages 1 and 2 (see also Viola et al., 2009) were used to infer a clockwise rotation of the horizontal 580 compression direction from NW-SE (Stage 1) to NNE-SSW (Stage 2). Consistent with the kinematic framework of Stage 2, we 581 propose here that during progressive regional exhumation and cooling to entirely brittle conditions, the BFZ300 deformation 582 continued through a further, distinct deformation phase (t_3 of Fig. 13e). This stage accommodated the selective reactivation of 583 the BFZ300 core, with renewed dilation due to the rotated σ_1 during Stage 2 acting subparallel to the strike of the Qtz I vein in 584 the BFZ300 core. Localised dilation in a still fluid-rich system allowed the emplacement of the Qtz II vein (Fig. 13e). Our estimations indicate that P_f and T conditions at that time were between 140 and 10 MPa and T $\approx 305^{\circ}$ C, respectively. The 585 586 BFZ300 core was reactivated under overall hybrid conditions (Fig. 13f), as suggested by the irregular thickess and curved 587 geometry of the Qtz II vein therein, and by the synkinematic chlorite crystals that are stretched orthogonally to the vein 588 boundaries (Fig. 3c). The Qtz II vein invariably localized along at the contact between Qtz I and the host rock (Figs. 2, 3 and 589 13e) suggesting selective reactivation along the pre-existing principal slip zones (Riedel shears and boundary shears, 590 Tchalenko, 1970), which represented the weakest part of the fault (strength profile Fig. 13h). Evidence for mesoscale hybrid 591 fracturing and our P_f estimates (Fig. 11) suggest that P_f was lower than that of the earlier deformation stages.

BFZ300 underwent one or more events of brittle fracturing and induration (Fig.13g), as suggested by CL imaging of Qtz II
crystals (Fig.7c). Fluid pressure estimations for this structural phase are between 160 and 40 MPa.

The stylolitic seams having a strike parallel to the BFZ300 fault zone suggest a direction of maximum compression (σ_1) oriented c. E-W, i.e. subparallel to the inferred Sveconorwegian main shortening direction (e.g., Viola et al., 2011). The sphalerite-stannite mineral pairs arranged along these structures are supposed to be concentrated through a pression-solution mechanism during this deformational stage.

598 Skyttä and Torvela (2018) proposed that BFZ300 is a brittle structure localized onto a zone of incomplete structural 599 transposition inherited from the earlier ductile history of the Olkiluoto basement. However, in our mesoscale and 600 microstructural analysis we did not find evidence of any ductile precursor, and we note that BFZ300 cuts the ductile structural 601 grain at high angle, which excludes any reactivation of precursor ductile fabrics.

602 5.3. Implications for seismic deformation at the base of the BDTZ

This study demonstrates the role of overpressured fluids on strain localisation during the incipient stages of fault nucleation and subsequent reactivation(s) at the BDTZ. The maximum estimated PT conditions derived in this study (peak conditions of 160

605 MPa and 350 °C) are indeed realistic for the base of the seismogenic zone in the continental lithosphere (e.g., Scholz, 1990, and

606 references therein) where the brittle-ductile transition for quartz occurs.

- 607 Mechanical models of long-term deformation (Rolandone and Jaupart, 2002) propose that deformation at the brittle-ductile
- 608 transition can be reasonably described as being mostly accommodated by intermittent and concomitant coseismic slip and





609 ductile flow. Major hydrofracturing, as that documented in this study by the Qtz I and II veins, is possibly related to seismic 610 failure. Faults accommodating hydrofracturing are indeed commonly interpreted as seismogenic (e.g. Sibson, 1992a; Cox, 1995)

611 particularly at depth, where the reactivation of misoriented faults is only possible for fluid pressures exceeding σ_3 (e.g. Sibson,

612 1985).

613 Our study confirms this view because BFZ300 contains not only brittle fault rocks overprinting and overprinted by veins, but 614 also clearcut evidence of mutually overprinting brittle and ductile deformation (Fig.6). In the light of the field observations 615 discussed and of the constraints derived, we suggest therefore that BFZ300 behaved in a seismic way at least during the 616 emplacement of the principal Qtz I and Qtz II veins. Hydrofracture veins are largely interpreted in the literature as the evidence 617 of earthquake in fluid-rich faults (Cox, 1995).

In this perspective, two possible scenarios can be considered to explain the genetic relationships between BFZ300 and a possible seismic behaviour of the crust during the Svecofennian orogeny. In a first scenario, the quartz veins of the fault core would represent the result of coseismic rupture during the mainshocks of a fully developed seismic cycle. Pore pressure fluctuations caused the repeated transient embrittlement of the rock mass, wich was otherwise under overall ductile conditions. The documented brittle-ductile cycles are thus the expression of coseismic fracturing and aseismic creep between the individual shocks, as shown by viscous deformation overprinting the brittle features, guided by the residual differential stress.

624 A second possibility is that faulting occurred in the absence of a well-defined sequence of main- and aftershocks. As in the case 625 of man-induced earthquakes triggered by high-pressure fluids during injection of fluids (e.g. Healy et al., 1968), where 626 deformation is typically accommodated by diffuse swarms of low magnitude seismicity rather than well-defined mainshock-627 aftershock sequences (Cox, 2016), we propose that BFZ300 might have localised strain by diffuse veining with crack and seal 628 textures (Cox, 2016). Breccias and cataclasites (Fig.s 3 and 8) mutually overprinting with veins show that failure and veining 629 were indeed broadly coeval (e.g. Cox, 1995; Cox, 2016). Healing in fluid-rich environments can occur over short periods of 630 time (days-months) when compared with recurrence time of large earthquakes (10-100 years) (Olsen et al., 1998; Tenthorey 631 and Cox, 2006). Therefore, the documented repeated switches between brittle and ductile deformations would then be steered 632 again by transient episodes of fluid overpressuring but in this case would express the accommodation of swarms of minor

633 background earthquakes within overall ductile conditions.

Microstructures of fault-rocks exhumed from the brittle-ductile transition in other geological settings, are mostly in agreement with our hypotheses of seismic deformation. Transient and short term high-stress deformation followed by phases of stress

636 relaxation, which is prevalently characterized by recovery and recrystallization processes, has been documented by several

- 637 authors in deformed quartz (Trepmann and Stöckhert, 2003; Trepmann et al., 2007; Bestmann et al., 2012; Trepmann and
- 638 Stöckhert, 2013; Trepmann et al., 2017).

639 To conclude, BFZ300 represents an interesting case of likely seismic deformation within a fluid rich system at the base of the

seismogenic crust. The absence of later, thoroughgoing and high-strain, potentially obliterating deformation episodes allows the

641 documentation of a complex structural evolution, from the earliest localisation to the mature structural stage.





642 6 Conclusions

- 643 Our analysis shows that a multi-disciplinary, multi-technique study of faulting initiation and evolution has indeed the potential
- to provide useful insights into the complex and cyclic processes of fluid-fault interaction and effects thereof at the base of the
- 645 seismogenic crust. It further constrains, moreover, the importance of cyclic seismicity and fluids in the fragmentation of
- 646 Precambrian cratons when deformed at the brittle-ductile transition zone, something that is not yet that well understood for the
- 647 Fennoscandian Shield. Our study, moreover, provides potentially important inputs to many modern geological applications,
- 648 including site characterization of deep geological disposal facilities for spent nuclear fuel. Results from the detailed geological
- 649 characterization of faults at the Olkiluoto site can thus be used toward the continuous updating of the geological site description
- and yield further constraints on the mechanics of faulting at those conditions and at that time.
- 651

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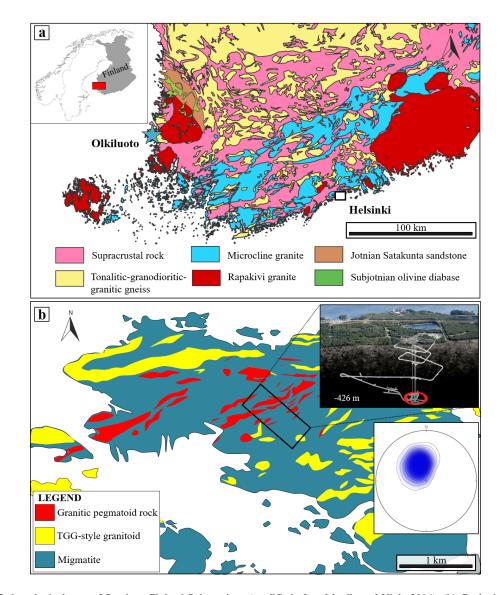
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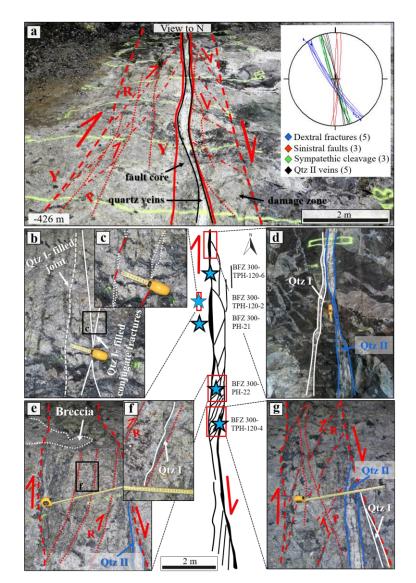




- 870 Figure 1. (a) Simplified geological map of Southern Finland Subprovince (modified after: Mattila and Viola 2014). (b) Geological sketch of 871 the Olkiluoto site. The upper inset on the right is a panoramic photo with an overlay drawing of the underground infrastructure (photo:
- courtesy of Posiva Oy, Finland). The red circle shows the depth location of BFZ300. The lower inset on the right shows the poles to foliation
 planes measured from the Olkiluoto drill cores (Mattila and Viola, 2014). This is mainly low-angle and SE-dipping.





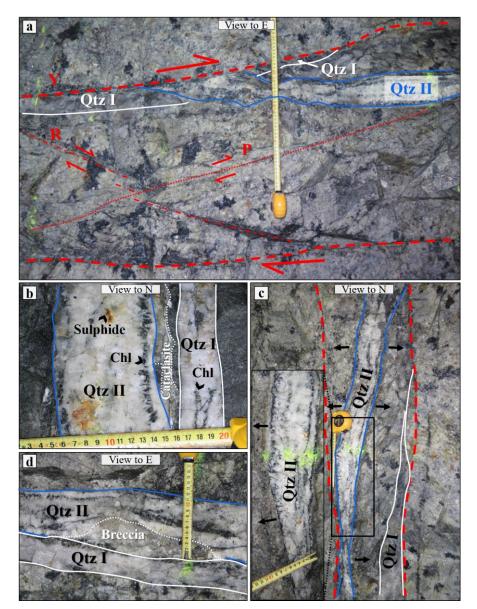


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875 Figure 2. BFZ300 fault geometry and architecture (centre of figure) and examples of representative outcrop features. The red rectangles locate 876 the areas along the fault segments where detailed outcrop photos were taken. Stars locate hand and drill core samples. Stars with black layout 877 identifysamples used for the microthermometric study. Note that the fault is made of two segments offset laterally at a astep over zone with 878 overall dextral kinematics. Fault core quartz veins are shown by thicker black lines in the schematic model. (a) View and interpretation of the 879 structural elements of the fault, whose core hosts two generations of quartz-chlorite veins. Inset shows lower-hemisphere, equiangular 880 projection of conjugate fault segments, cleavage and quartz-chlorite veins. (b) Damage zone made of mm-thick, en-echelon veins connected by 881 conjugate shear segments. (c) Detail of (b) showing fractures filled by the first quartz generation (Qtz I). (d) Two distinct generations of 882 quartz-chlorite veins recognised in the fault core (Qtz I and Qtz II). (e) Detail of the compressional step-over zone characterized by multiple 883 and parallel T fractures, filled by Qtz I. A brecciated body is crosscut by the Y planes. (f) Detail of a Tensional fracture infilled by Qtz I. (g) 884 Compressional structures (P shears) from the step-over zone and relationships between Qtz I and Qtz II within the fault. The Riedel geometry 885 suggests that the Qtz II vein formed due to the reactivation of the principal slip zones..





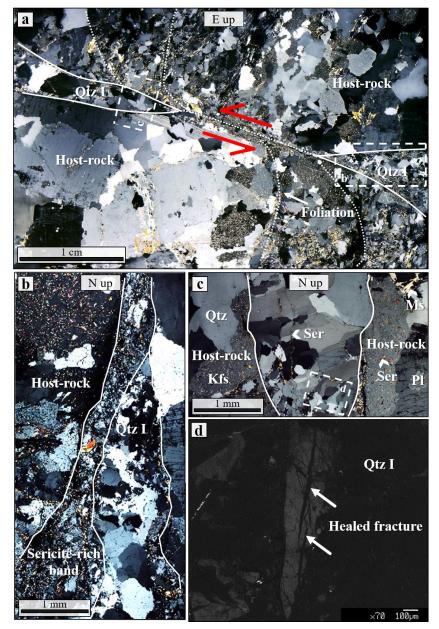


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887 Figure 3. Relationships between Riedel shears and quartz-chlorite veins. (a) Sharp shear boundaries along fault veins and Riedel shears. The 888 geometry of the R and P shears suggests a dextral kinematics. White lines highlight the positions of the two types of quartz veins. Note the Qtz 889 II vein cutting the Qtz I vein. (b) Juxtaposed Qtz I and Qtz II veins. Qtz I veins are thinner and made of a translucid, small grained quartz. In 890 contrast, Qtz II veins, which contain pockets of sulphide aggregates, are thicker and made of larger and euhedral quartz. Chlorite occurs as 891 minor phase in both types of veins, but only in Qtz II veins it forms long and prismatic aggregates growing perpendicular to the fracture walls. 892 In Qtz I veins, chlorite is small grained and forms thin levels within the quartz. Notice the presence of a cataclastic band between the two 893 veins. (c) Spatial continuity of the chlorite aggregates within the Qtz II veins, which grow always orthogonal to the vein boundaries. Inset 894 shows the detail of the prismatic aggregates forming long and parallel ribbons. This open space filling texture suggests hybrid conditions of 895 reactivation of the older Qtz I veins. (d) Small quartz breccia formed between the two generations of quartz veins.





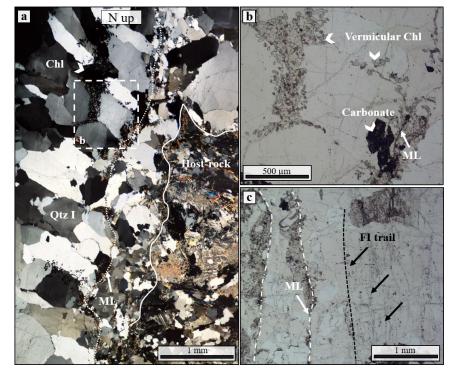


896

Figure 4. Microtextural characteristics of Qtz I from the damage zone of BFZ300 (sample: TPH-120-2). (a) Composition of photomicrographs
of a Qtz I vein interconnecting with a sinistral shear band (crossed nicols). Faulting kinematics is suggested by drag folds in the host rock. (b)
Tip of Qtz I vein hosted by a sericite-rich cataclastic band of the host rock. (c) Detail of panel a showing open-space filling texture in the Qtz I
vein. Notice the sericite microfractures crosscutting Qtz I. (d) Panchromatic cathodoluminescence image of Qtz I showing healed
microfractures crosscutting the crystal.







904

Figure 5. Microtextural characteristics of Qtz I from the damage zone of BFZ300 (sample PH-21). (a) Stitched microphotographs of a Qtz I vein showing elongated-blocky texture with crystals growing obliquely with respect to the vein boundaries, which suggests growth under oblique dilation. Mode I opening is highlighted by the presence of a median line (ML) that is decorated by syn-kinematic chlorite and aggregates of REE-bearing carbonate. (b) Detailed view of the chlorite and carbonate found along the ML. (c) Photomicrograph (plane polarized light) showing a set of fluid inclusion trails parallel to the median lines and crosscutting Qtz I, which suggests crack-seal opening and sealing.





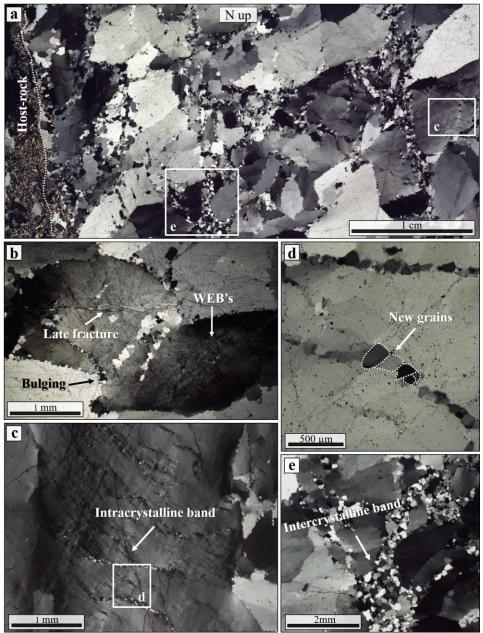
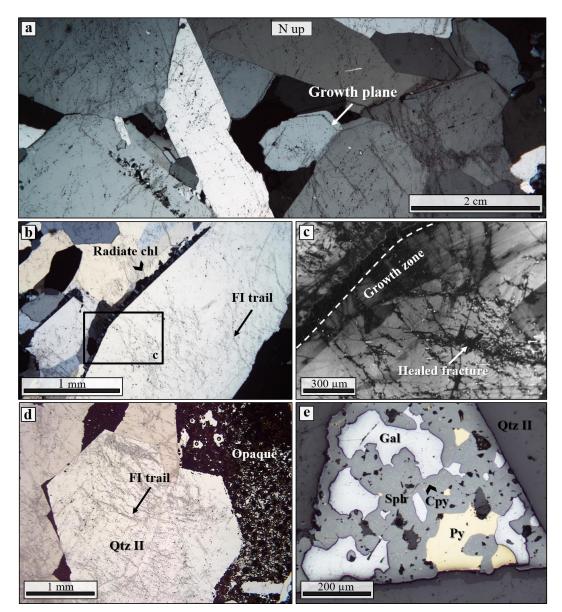


Figure 6. Microtextural characteristics of Qtz I from the BFZ300 core (sample TPH-120-4). (a) Stitched photomicrographs showing the typical 913 heterogeneous grain size of Qtz I (30-800 µm). (b) Evidence of plastic deformation of Qtz I from the fault core given by bulging of the largest 914 crystals, wide extinction bands and undulose extinction. Note the late brittle fractures crosscutting all the previously formed plastic features. 915 (c) Intracrystalline deformation bands within a large crystal. (d) Detail of (c) showing the typical grain size of the band (50-250 µm). 916 Intracrystalline deformation bands are oriented at $<30^{\circ}$ with respect to the BFZ300 vein walls and can be up to 2 mm in length. (e) 917 Intercrystalline deformation band showing a thickening at the triple junction of larger grains. These intercrystalline bands are parallel to the 918 strike of BFZ300.





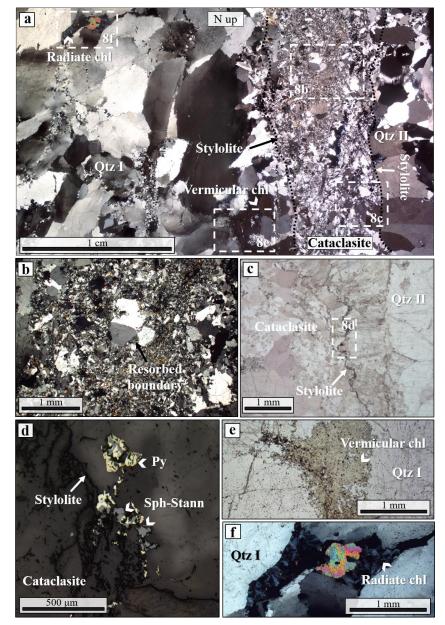


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920 Figure 7. Microstructural characteristics of Qtz II from BFZ300 (samples TPH-120-6, PH-22). (a) Stitched photomicrographs of Qtz II vein 921 from the fault core. Notice the coarse quartz crystals and their elongated-blocky texture. Primary growth textures are sometimes visible and are 922 marked by solid inclusions and decrepitated FIAs. (b) Radiate chlorite crystals along a prismatic Qtz II crystal boundary. Note that Qtz II is 923 crosscut by numerous trails of FIs. (c) Panchromatic cathodoluminescence image of the same large Qtz II crystal from panel b, showing radiate 924 chlorite along the crystal boundary and a primary growth zone cut by a set of healed fractures. (d) Euhedral quartz crystals set within opaque 925 phases and crosscut by a network of thin microfractures. (e) Reflected light photomicrograph showing the opaque mineral assemblage 926 typically associated with Qtz II, i.e. subhedral to anhedral sphalerite, pyrite, and galena. Chalcopyrite is a minor phase and occurs as small 927 round inclusions within sphalerite (chalcopyrite "disease") or as large subhedral/anhedral masses together with galena.





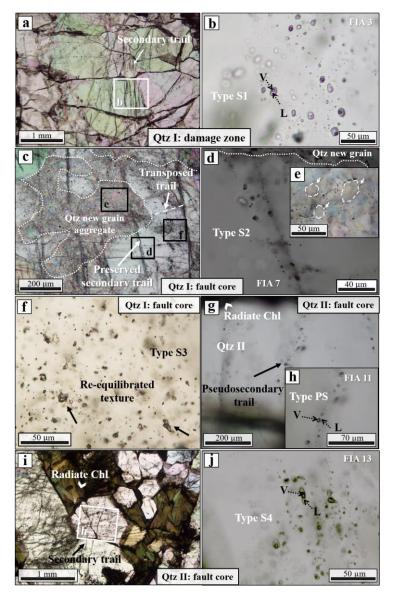


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929 Figure 8. Microtextural characteristics of the cataclasite juxtaposing Qtz I and Qtz II veins (sample TPH-120-4). (a) Stitched 930 photomicrographs covering the contact between the two quartz veins and the intervening, 5 mm-thick cataclastic band. (b) Cataclastic band 931 made of large Qtz I fragments (8-12 mm) embedded within a finer matrix (20-200 µm in size) made of sericite and quartz. The largest crystals 932 show lobate boundaries, suggesting dissolution and local resorption along the clast-matrix interface. (c) Detail view of the cataclastic band, 933 which is defined by two stylolite seams that strike parallel to the BFZ300. (d) Reflected-light photomicrograph showing anhedral to subhedral 934 pyrite, chalcopyrite, stannite, and sphalerite arranged along the stylolite as residual product of pressure solution. (e) Vermicular chlorite 935 associated with Qtz I close to the cataclastic band. (f) Radiate chlorite associated with Qtz I.







937

938 Figure 9. Characteristics textures of FIAs hosted within the BFZ300 quartz (samples PH-21, TPH-120-4, TPH-120-6). (a) Secondary trails 939 crosscutting large Qtz I crystals of the damage zone. (b) Detail of (a) showing the phase ratios of one of the studied secondary assemblages 940 (FIA3), most representative of Type S1 FIA. (c) Long secondary transgranular trails crosscutting Qtz I of the fault core, dismembered by 941 intercrystalline fractures, infilled by quartz new grains. Qtz I fault core also hosts set of short sub-trails developed at high angle with respect to 942 the long trails. (d) Detail of Type S2 FIA entrapped along a preserved secondary fracture trail. (e) Small inclusions (<1µm) arranged along the 943 boundaries of new polygonal quartz. (f) Example of Type S3 FIA arranged as isolated clusters inside ductile deformed fault core Qtz I. These 944 trails formed during a brittle deformation stage that pre-dates ductile re-crystallization. (g) Pseudosecondary FIA associated with Qtz II-945 chlorite (FIA11) and (h) its phase ratio details. (i) Small scale view of secondary FIAs crosscutting Qtz II. (j) Detail of secondary trails 946 crosscutting euhedral Qtz II (FIA 13). In all photographs north points up.





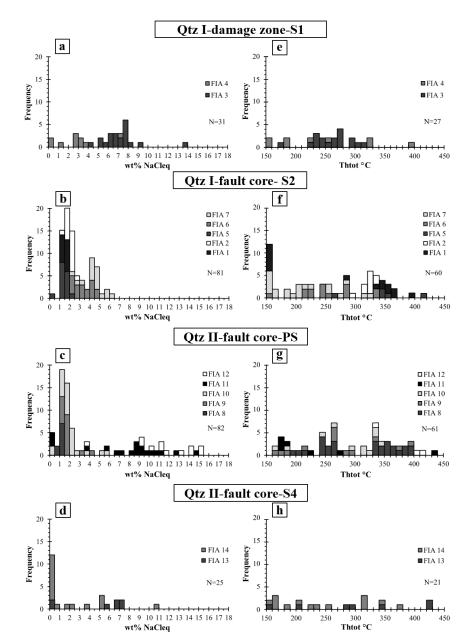
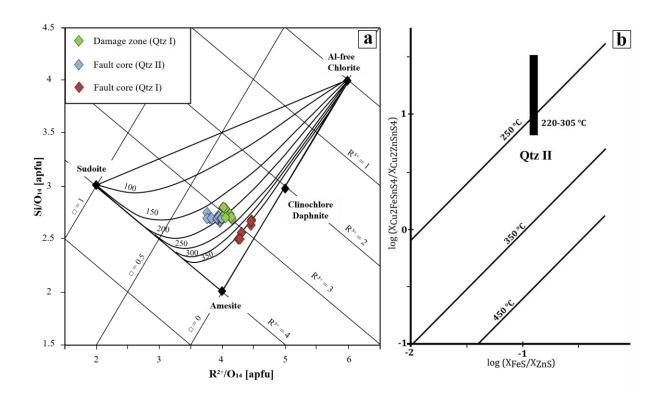


Figure 10. Microthermometric data of the studied FIAs. Panels a-d show the bulk salinities of individual FIAs calculated from the Tmice data while panels e-h refer to the temperatures of final homogenization of the same assemblages. Notice that the data report the properties of individual FIAs according to their occurrence within Qtz I of the damage zone, Qtz I from the fault core, and Qtz II from the fault core. Notice that pseudosecondary (PS) and secondary (S) FIAs identify progressive later stages of fluid entrapment, and can be used to constrain the fluid properties in the fault zone. Notice also that the measured ranges of Thtot spread across T intervals that are too large to represent entrapment at equilibrium (e.g., FIA7 of Qtz I from fault core: 150-330 °C), which suggests post-entrapment re-equilibration of the inclusions. Fluid bulk composition is expressed as salinity, which is conventionally reported as weight percent of NaCl equivalents (wt%NaCleq, Roedder,1984).





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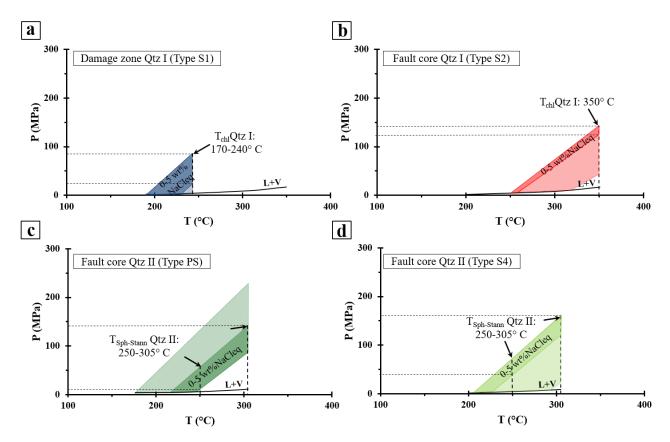


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Figure 11. Mineral-pair geothermometry applied to the assemblages of the QtzI- and Qtz II veins. (a) Chlorite-quartz formation temperature estimated using the method of Bourdelle and Cathelineau (2015). Green, red, and light blue symbols indicate the distinct textural types of chlorite in Qtz I and II, respectively. The maximum temperature is from the Qtz I-chlorite pair from the fault core. The other groups of chlorites in the 150-250 °C range indicate a second stage of quartz-chlorite precipitation in the fault core and damage zone, in line with microthermometric constraints. (b) Temperature of sphalerite-stannite formation estimated with the method of Shimizu & Shikazono (1985) that uses Fe and Zn partitioning between stannite and sphalerite. The region of the plot that was calibrated with this geothermometer lies between the 250 and 450 °C isotherms. Hence, compositions corresponding to T<250 °C should be interpreted with caution.</p>



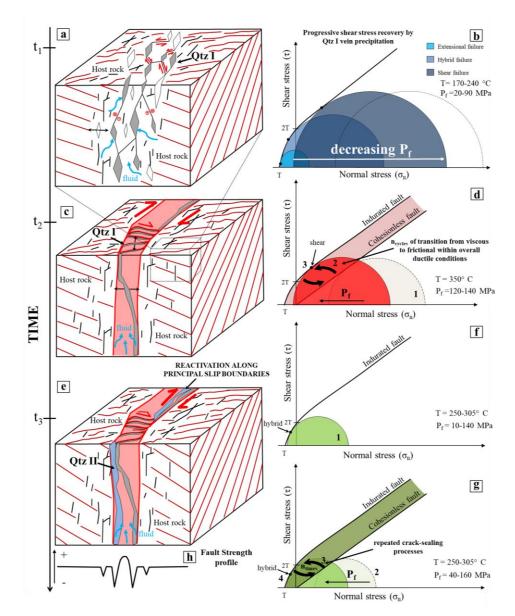




967 Figure 12. Estimated fluid pressure for the various typologies of FI discriminated petrographically and on the basis of the identified structural 968 domains. Fluid pressures are related to (a) damage zone Qtz I, (b) fault core Qtz I, (c-d) fault core Qtz II. The light-coloured areas are defined 969 by the uppermost and lowermost sets of fluid inclusion isochores, while the dark-coloured areas are related to the pressure range calculated for 970 isochores computed for the most probable composition of the pristine fluid (salinity between 0 and 5 wt%NaCleq, see text for more details). 971 The pressure-temperature areas are also defined by the mineral pairs thermometry (dotted vertical lines), and by the liquid-vapour equilibrium 972 curves for the H2O-NaCl model fluid. The 240 °C vertical line represents the equilibrium temperature between chlorite and damage zone Qtz I. 973 The 350 °C vertical line is the equilibroum temperature between chlorite and fault core Qtz I. The 250-305 °C vertical lines mark the lowest 974 and highest sphalerite-stannite equilibrium temperature with Qtz II in the BFZ300 fault core. The thick lower curve marks the bundle of liquid-975 vapour curves for a 1-5 wt% NaCl fault fluid.





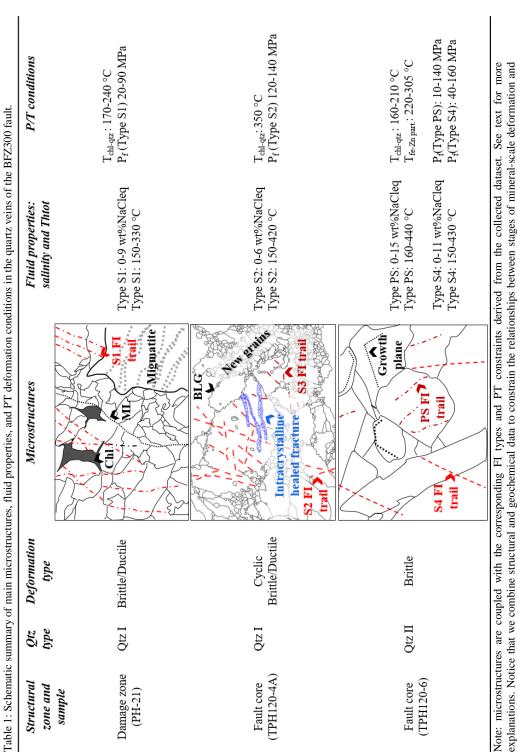


978 Figure 13. Conceptual model of the temporal and mechanical evolution of the BFZ300 fault zone (see text for more details). Red lines: traces 979 of metamorphic foliation. Black lines: fractures related to the BFZ300 structural development. (a) Initial embrittlement of the migmatitic 980 basement occurred by fracture coalescence under (b) initial lower differential stress conditions and high fluid pressure and followed by a 981 transient increase of differential stress. A first generation of quartz veins (Qtz I) precipitated inside the diffuse network of joints and 982 hydrid/shear fractures which formed during this first deformation stage. (c) Progressive strain localization and fluid channeling within the fault 983 core occurred by (d) episodically renewed fluid-pressure build-up driven by cycles of brittle and ductile deformation. (e-g) Progressive 984 exhumation and cooling of the fault system occurred concomitant with several brittle reactivation episodes of the fault zone under hybrid 985 conditions and fluid pressure lower than during the previous deformational stages. Lastly, a second generation of quartz veins (Qtz II) was 986 emplaced, mainly along the principal slip boundaries of the fault core, following the Qtz I vein as shown by (h) the strength profile across the 987 fault architecture, that suggests lower tensile strength values (and hence higher reactivation potential) along the Qtz I vein / host rock walls.





988



fluid circulation, which in turn defines the relative chronology of stages of fluid flow during faulting.

ML: median line; Blg: bulging.





Sample 4A	4A 4A	·	4A 4A		4A	PH21	PH21	PH21	2	2	2	9	9	9	9
Structural zone	FC FC		Æ	FC	FC	DZ	DZ	DZ	DZ	DZ	DZ	FC	FC	FC	FC
Quartz type Q	Qtz I Qtz I		-	Qtz I	Qtz II	Qtz II	Qtz II	Qtz II							
Textural type V(Verm Veri			Rad	Rad	Verm	Verm	Verm	Verm	Verm	Verm	Rad	Rad	Rad	Rad
Na ₂ O 0				0.08	0.03	0.05	0.02	0.04	0.03	0.01	0.05	0.04	0.06	0.01	0.01
TiO ₂ 0				0.03	0.01	0.09	0.04	0.01	0.01	0.01	0.03	0.03	0.03	0.04	0.13
MnO 0				0.56	0.48	0.24	0.24	0.30	0.48	0.37	0.43	0.64	0.57	0.71	0.60
K2O 0	0.06 0.02	2 0.04	4 0.07	0.06	0.04	0.01	0.01	0.03	0.10	0.05	0.07	0.03	0.02	0.05	0.01
MgO 13	13.66 13.7			5.13	6.75	13.95	14.06	13.29	12.85	12.57	12.59	4.85	4.87	8.73	8.05
SiO ₂ 25	_			22.89	23.91	27.24	27.02	27.49	27.43	27.88	27.79	25.63	25.64	26.5	26.13
Cr ₂ O ₃ 0.	0.00 0.01			0.00	0.06	0.04	0.03	0.06	0	0.06	0.01	0	0.02	0.01	0
FeO 27	27.86 27.7	_		38.49	36.75	24.68	25.21	26.07	25.97	26.06	25.77	34.26	33.84	30.08	30.47
CaO 0	0.03 0.04			0.06	0.03	0.01	0.02	0	0.05	0.05	0.03	0.01	0.04	0.04	0.02
Al ₂ O ₃ 22	22.04 22.13			23.35	22.98	24.13	24.75	24.91	24.02	23.48	23.21	24.23	24.64	24.49	25.02
CI	0.00 0.00			0.02	0.04	0.01	0	0	0.01	0	0	0.03	0.02	0.02	0.01
Total 85	89.78 90.45	0,	0,	90.67	91.09	90.69	91.42	92.23	91.12	90.81	90.08	89.82	89.94	90.78	90.48
No. ions in formula Based on 28 (O,OH)	(HO														
Na 0	0.03	3 0	0.03	0.03	0.01	0.02	0.01	0.01	0.01	0.01	0.02	0.02	0.03	0	0.01
Ti	0 0			0	0	0.01	0.01	0	0	0.01	0	0.01	0.0	0.01	0.02
Mn 0	0.10 0.11		-	0.10	0.09	0.04	0.04	0.05	0.08	0.06	0.07	0.12	0.10	0.12	0.11
K	0.01 0			0.02	0.01	0	0	0.01	0.02	0.01	0.02	0.01	0	0.01	0
Mg 4		5 4.25		1.68	2.17	4.18	4.19	3.93	3.86	3.79	3.82	1.55	1.55	2.69	2.49
Si				5.02	5.15	5.48	5.40	5.46	5.53	5.64	5.66	5.49	5.47	5.48	5.43
Cr.	0 0			0	0.01	0.01	0	0.01	0	0.01	0	0	0	0	0
Fe ²⁺ 4.		9 4.83	3 6.66	7.06	6.62	4.15	4.21	4.33	4.38	4.40	4.39	6.14	6.04	5.20	5.29
Ca				0.01	0.01	0	0	0	0.01	0.01	0.01	0	0.01	0.01	0
				6.04	5.84	5.72	5.83	5.83	5.71	5.59	5.57	6.12	6.20	5.97	6.13
CI	0 0		-	0.01	0.01	0	0	0	0	0	0	0.01	0.01	0.01	0
Fe 4.				7.06	6.62	4.15	4.21	4.33	4.38	4.40	4.39	6.14	6.04	5.20	5.29
Al Tetr 2	2.68 2.63	3 2.64	4 2.86	2.98	2.85	2.52	2.60	2.54	2.47	2.37	2.34	2.51	2.53	2.52	2.57
Al Oct 2	2.73 2.76			3.06	2.99	3.20	3.22	3.29	3.24	3.23	3.23	3.61	3.67	3.45	3.56
Based on 28 (O,OH)															
R ²⁺ 9	9.11 9.04	4 9.08	8 8.80	8.74	8.79	8.33	8.40	8.26	8.24	8.19	8.21	7.69	7.59	7.90	7.79
Si 5	5.32 5.37			5.02	5.15	5.48	5.40	5.46	5.53	5.64	5.66	5.49	5.47	5.48	5.43
Based on 14 (O,OH)															
R ²⁺ 4.	4.55 4.52	2 4.54	4 4.40	4.37	4.40	4.17	4.20	4.13	4.12	4.10	4.10	3.84	3.79	3.95	3.89
Si 2	2.66 2.68			2.51	2.58	2.74	2.70	2.73	2.77	2.82	2.83	2.75	2.74	2.74	2.71





1001 Table 3: Representative EPMA of sulphides associated with Qtz II

	Structural	Qtz											
Analysis	zone	type	Mineral	S	Fe	Cu	As	Pb	Ni	Zn	Ti	Sn	Total
TPH120-6-14	Core	II	pyrite	55.02	47.50	0.01	0.00	0.00	0.02	0.00	0.00		102.55
TPH120-6-17	Core	II	pyrite	54.08	47.19	0.00	0.01	0.00	0.00	0.00	0.00		101.28
TPH120-6-18	Core	II	sphalerite	34.46	6.46	0.09	0.01	0.00	0.03	59.62	0.02		100.69
TPH120-6-19	Core	II	sphalerite	34.48	6.24	0.08	0.06	0.00	0.04	59.61	0.02		100.53
TPH120-4A-34	Core	Π	pyrite	54.49	47.40	0.05	0.00	0.00	0.00	0.00	0.00		101.94
TPH120-4A-35	Core	II	pyrite	54.13	47.26	0.02	0.04	0.00	0.00	0.01	0.55		102.01
TPH120-4A-38	Core	II	galena	13.40	0.00	0.00	0.00	86.63	0.00	0.32	0.01		100.36
TPH120-4A-59	Core	II	galena	13.50	0.06	0.00	0.01	87.04	0.00	0.10	0.01		100.72
TPH120-4A-40	Core*	II	sphalerite	35.06	9.46	0.05	0.00	0.00	0.00	56.74	0.01		101.32
TPH120-4A-43	Core*	II	sphalerite	34.69	9.04	0.01	0.03	0.00	0.00	57.51	0.01		101.28
TPH120-4A-41	Core	II	chalcopyrite	35.40	30.53	33.51	0.00	0.00	0.00	1.32	0.00		100.76
TPH120-4A-42	Core	II	chalcopyrite	35.78	30.78	33.59	0.03	0.00	0.01	1.22	0.01		101.42
TPH120-4A-19	Core **	Π	stannite	29.79	12.53	28.41	0.07	0.08	0.00	0.92	0.000	27.86	99.66
TPH120-4A-22	Core **	Π	sphalerite	33.82	8.15	0.06	0.00	0.03	0.02	57.27	0.006	0.00	99.36

1002 1003 1004 Note: * - located within cataclastic band and close to stylolite. ** - located along stylolite

Sphalerite and stannite compositions from locations indicated by ** have been used to calculate the temperatures of sphalerite-stannite equilibrium

following the geothermometer of Shimizu and Shikazono (1985). See text for more explanations.