

Crystal-plastic deformation in seismically active carbonate fault rocks

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Key points:

- Crystal-plastic deformation occurred in seismically deformed carbonate rocks
- Deformation and annealing produce a grain-boundary strengthening effect
- Repeated cyclic repetition of deformation and recrystallisation leads to formation of a nanogranular material

28 Abstract

29 The spatial separation of macroscopic rheological behaviours has led to independent
30 conceptual treatments of frictional failure, often referred to as brittle, and viscous deformation.
31 Detailed microstructural investigations of naturally deformed carbonate rocks indicate that both,
32 frictional failure, and viscous mechanisms might operate during seismic deformation of
33 carbonates. Here, we investigate the deformation mechanisms that were active in two carbonate
34 fault zones in Greece by performing detailed slip-system analyses on data from automated crystal-
35 orientation mapping transmission electron microscopy and electron backscatter diffraction. We
36 combine the slip system analyses with interpretations of nanostructures and predictions from
37 deformation mechanism maps for calcite. The nanometric grains at the principal slip surface
38 should deform by diffusion creep but the activation of the $(0001)\langle\bar{1}2\bar{1}0\rangle$ slip system is evidence
39 for a contribution of crystal plasticity. A similar crystallographic preferred orientation appears in
40 the cataclastic parts of the fault rocks despite exhibiting a larger grain size and a different fractal
41 dimension, compared to the principal slip surface. The cataclastic region exhibits microstructures
42 consistent with activation of the $(0001)\langle\bar{1}2\bar{1}0\rangle$ and $\{10\bar{1}4\}\langle\bar{2}021\rangle$ slip systems. Post-
43 deformational, static recrystallisation and annealing produces an equilibrium microstructure with
44 triple junctions and equant grain size. We propose that repeated introduction of plastic strain and
45 recrystallisation reduces the grain size and offers a mechanism to form a cohesive nanogranular
46 material. This formation mechanism leads to a grain-boundary strengthening effect resulting in
47 slip delocalisation which is observed over six orders of magnitude (μm – m) and is expressed by
48 multiple faults planes, suggesting cyclic repetition of deformation and annealing.

49 1 Introduction

50 Seismic slip and aseismic creep commonly occur in distinct portions of the lithosphere due
51 to the different dependencies of the underlying deformation mechanisms on conditions such as
52 pressure and temperature (Scholz, 1998). Frictional failure involves dilatant processes facilitated
53 by low confining pressures at shallow depths (Sammis *et al.*, 1987; Sammis and Ben-Zion, 2008),
54 whereas viscous deformation occurs by thermally activated processes promoted by higher
55 temperatures at greater depths (Sibson, 1982; Bürgmann and Dresen, 2008). However, the
56 temperature-increase through shear heating during seismic faulting (Rice, 2006) challenges this
57 strict separation by potentially activating temperature-dependent deformation mechanisms, such
58 as crystal plasticity and diffusion creep (Nielsen, 2017). Depending on the material, melting or
59 decomposition reactions can also occur at high temperatures, leading to severe microphysical
60 changes that severely alter the mechanical behaviour of faults (Di Toro *et al.*, 2011; Niemeijer *et*
61 *al.*, 2012). The main factor limiting the operation of crystal plasticity in the brittle regime is the
62 extremely short duration of the temperature-increase during and after fault slip. Thermal models
63 predict a temperature drop through thermal diffusion within one second after sliding ceases to a
64 value similar to the background temperature (Di Toro and Pennacchioni, 2004; Demurtas *et al.*,
65 2019). Therefore, a key objective of earthquake geology is to assess the extent to which thermally
66 activated processes impact fault structure and properties e.g., modifying the microstructure or
67 activation of deformation mechanisms, during the short interval of coseismic slip.

68 Deformed carbonates from principal slip zones of natural and experimental faults
69 commonly exhibit crystallographic preferred orientations (CPOs) (Smith *et al.*, 2013; Verberne *et*
70 *al.*, 2013; Delle Piane *et al.*, 2017; Kim *et al.*, 2018; Demurtas *et al.*, 2019; Pozzi *et al.*, 2019).
71 Most of the CPOs involve (0001) planes aligned subparallel to the shear plane, typically with an
72 antithetic inclination against the shear direction. In addition, the CPOs include alignment of the

73 $\langle\bar{1}2\bar{1}0\rangle$ axes subparallel to the shear direction. Similar CPOs are generated in high-temperature,
74 low-strain rate experiments, in which calcite is deformed by dislocation-mediated deformation
75 mechanisms (Pieri *et al.*, 2001). In general, the observations of CPOs in carbonate fault rocks
76 suggest that crystal plasticity contributes to accommodating applied strain during seismic
77 deformation. The contrast between frictional failure at the macroscale and the formation of CPOs
78 by dislocation-mediated processes at the microscale demonstrates the need to further constrain the
79 spatial and temporal evolution of deformation mechanisms during fault slip.

80 At the microscale, high-temperature grain-boundary sliding (GBS) has been suggested to
81 operate within the gouge volume near the principal slip surface (PSS) (De Paola *et al.*, 2015). In
82 the pursuit of predicting rheological behaviour during seismic fault slip, De Paola *et al.* (2015)
83 used deformation mechanism maps constructed from steady-state flow laws. For carbonates with
84 small grain sizes, these flow laws predict the operation of grain-size sensitive (GSS) deformation
85 mechanisms such as diffusion creep (Herwegh *et al.*, 2003) and dislocation-accommodated grain
86 boundary sliding (disGBS) (Walker *et al.*, 1990). In contrast, coarse-grained carbonates are
87 predicted to exhibit grain-size insensitive (GSI) behaviour inferred to result from dislocation glide
88 and dislocation cross-slip (Renner *et al.*, 2002; De Bresser, 2002). To reasonably use flow laws to
89 predict rheological behaviour, flow-law parameters, such as the stress exponent, n , the grain size
90 exponent, p , and the activation energy, Q must be known. Most of the parameters are derived from
91 laboratory experiments under well-constrained conditions and at steady state so that inferring these
92 parameters for the materials in any particular natural fault zone can be challenging. Strain rates
93 during experiments performed to constrain flow-law parameters are orders of magnitude lower
94 than those occurring during seismic slip on natural faults and therefore, predicting deformation
95 mechanisms during seismic deformation requires the flow laws to be extrapolated in stress/strain

96 rate. It is challenging to test the accuracy of such extrapolations based on mechanical data from
97 high-velocity deformation experiments, so microstructural analyses offer critical additional
98 information against which to test the accuracy of flow-law predictions.

99 The present study continues previous work on the nanostructural processes of the same
100 fault exposures. For more information and a detailed introduction to the geological background
101 the reader is kindly referred to Ohl et al. (2020). In the present study, we characterise the micro-
102 and nanostructures of natural carbonate fault rocks directly at a slip interface using multiscale
103 crystallographic orientation analyses to evaluate deformation mechanisms during seismic events.
104 The fault-rock microstructures reveal that crystal plasticity contributed during deformation and
105 that the microstructure was potentially modified by recrystallisation.

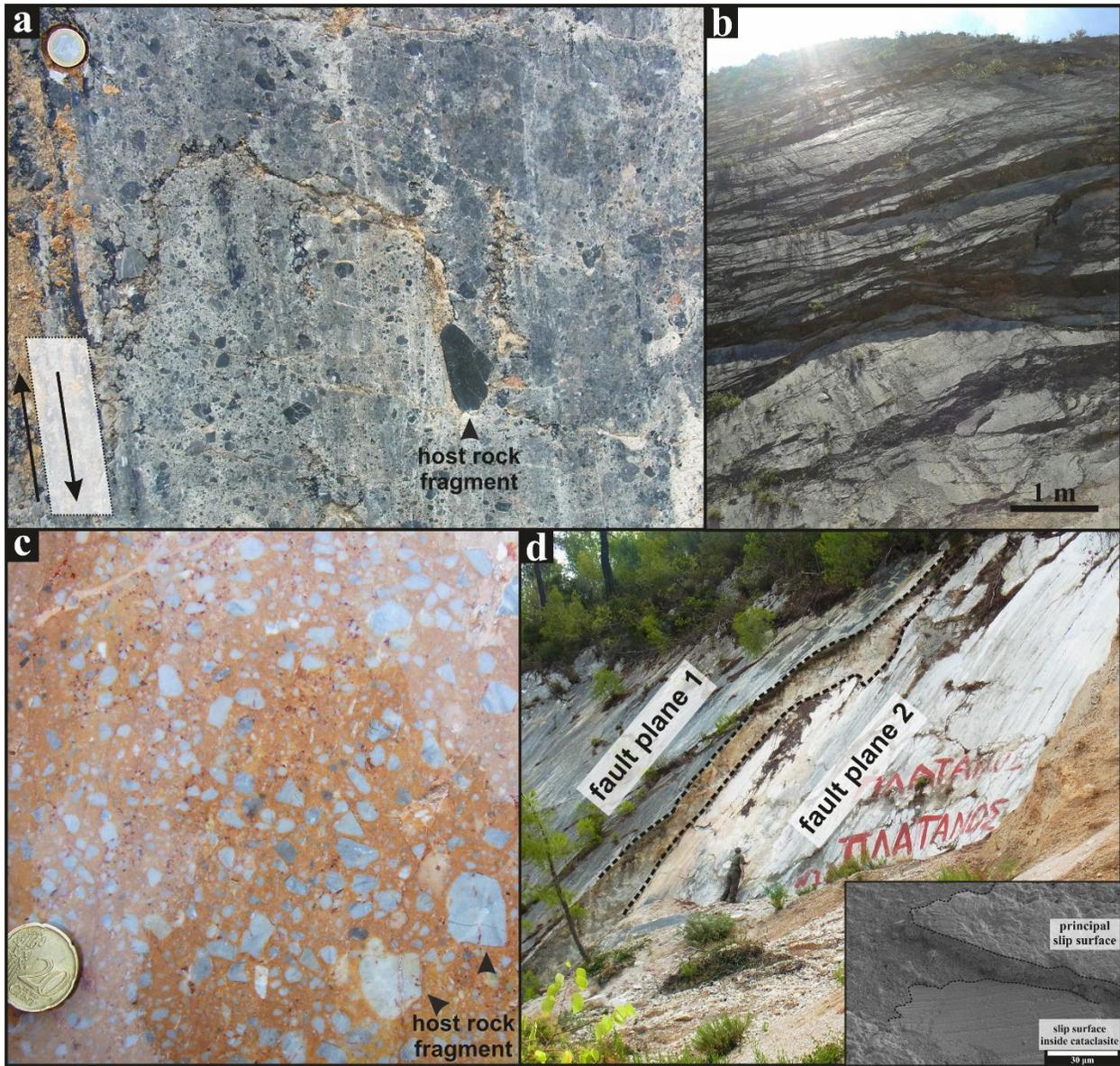
106 2 Geology and tectonic setting

107 The first investigated fault exposure (38°43'56.17"N, 23°0'27.41"E) is located close to
108 Arkitsa, along the northern coast of the Gulf of Evia, Greece. This fault exposure is part of the
109 Kamena Vourla fault system with a length of about 30–40 km (Ambraseys and Jackson, 1990). In
110 general, the ESE-WNW-striking, N-dipping fault planes separate Triassic to Middle/Late Jurassic
111 platform carbonates of the footwall from lower Pliocene-Pleistocene up to Quaternary hanging-
112 wall sediments (Kokkalas *et al.*, 2007). The footwall cataclasite is a greyish, matrix-supported
113 fault rock with host-rock clasts (Fig. 1a, S1a and S1b). Multiple fault planes are hosted inside the
114 damage zone, indicating fault-plane overstepping (Fig. 1b). Cumulative fault displacement is not
115 mentioned or documented in the geological literature, but present days outcrop situation shows the
116 fault rocks in contact with quaternary deposits. Records of historic seismicity document ~13 events
117 since 426 BC with the last major nearby event of M_s 6.9 in 1894 (Ambraseys and Jackson, 1990).

118 The second fault exposure (38° 2'14.40"N, 23° 0'22.33"E) is located close to Schinos,
119 Corinth area. Here, the fault exposure is part of a ~ 25-km long onshore fault line with an E-W
120 strike, dipping towards N. The host rocks are Upper Triassic limestones and dolomites (Kaplanis
121 *et al.*, 2013) and a reddish cataclasite with light-grey host-rock clasts forms the footwall fault rock
122 (Fig. 1c, S1c and S1d). In the field, the fault-plane exposure shows at least one stepover (Fig. 1d).
123 Cumulative fault displacement is not mentioned or documented in the geological literature, but
124 present days outcrop situation shows the fault rocks in contact with quaternary colluvium deposits.
125 The last seismic event in the region was recorded in February 1981, when three major events
126 occurred with a maximum magnitude of M_s 6.7 (Collier *et al.*, 1998).

127 Subduction-related back-arc volcanism, combined with extensional tectonics caused by
128 rollback of the Hellenic subduction zone (Thomson *et al.*, 1998), results in a high geothermal
129 gradient across the Aegean region (Papachristou *et al.*, 2014; Lambrakis *et al.*, 2014). The
130 geothermal gradient measured from geothermal exploration boreholes in the Sperchios basin,
131 approx. 50 km west of Arkitsa, is 35 °C/100 m (Metaxas *et al.*, 2010). Similar measurements at
132 Kamena Vourla indicate 46 °C at 200 m depth (Mendrinis *et al.*, 2010). Clay-mineral assemblages
133 in the Arkitsa fault formed from 100–150 °C (Papoulis *et al.*, 2013). However, the clays are found
134 inside the hanging-wall breccia and may not reflect the processes and temperatures on the fault
135 plane. Also, in the Sousaki-Loutraki region close to Schinos, geothermal exploration drilling
136 revealed high temperatures at shallow depth. In this region, (Mendrinis *et al.*, 2010) measured 63
137 °C at 500–1100 m depth, which is in agreement with (Lambrakis *et al.*, 2014) obtaining ≥ 75 °C
138 at 600–900 m depth. Because the above-mentioned temperature indications stem from geothermal
139 explorations, it is not clear whether they represent temperatures of host rock or fluid temperatures.

140 However, thermal models of the Aegean region predict temperatures from 200 °C (Limberger et
 141 al., 2014) to 360 °C (Larède, 2018) at 5 km depth.



142
 143 *Figure 1: Overview of geological features. **a:** View onto Arkitsa fault plane. Dark, large host-rock clasts are*
 144 *incorporated into the light-grey footwall cataclasite. Arrow indicates slip direction. One-Euro coin for scale. **b:***
 145 *Multiple slip planes hosted inside the damage zone of the Arkitsa fault exposure exhibit overstepping. **c:***
 146 *View onto Schinos fault plane. Light-grey host rock clasts incorporated into red hanging-wall cataclasite. **d:** Field view of*
 147 *Schinos fault plane exposure. Two distinct and overstepping fault planes are visible, hosted inside the damage zone.*
 148 *Person for scale. **Inset:** Secondary electron image showing development of secondary slip surface inside the Schinos*
 149 *footwall cataclasite. The secondary slip surface is situated about 10 μ m below the principal slip surface.*

150 3 Methods

151 3.1 Crystal orientation acquisition

152 Thin sections were prepared from drill cores by cutting parallel to the slip direction and
153 normal to the slip surface. Electron backscatter diffraction (EBSD) data were acquired using a
154 Philips XL30 scanning electron microscope (SEM) equipped with an Oxford Instruments Nordlys
155 2 CCD camera. Maps were acquired with an accelerating voltage of 30 kV, probe current of 9.5
156 nA, and step size of 0.5 μm for the Arkitsa sample and 20 kV accelerating voltage, 9.5 nA probe
157 current, 0.7 μm step size for the Schinos sample.

158 Crystal-orientation data were also acquired in a transmission electron microscope (TEM)
159 using the automatic crystal orientation mapping technique (ACOM-TEM, (Rauch and Véron,
160 2014)). TEM foils were prepared with a FEI Helios G3 focussed ion-beam scanning electron
161 microscope (FIB-SEM). ACOM-TEM data were acquired using the NanoMEGAS
162 ASTAR/SPINSTAR system on a FEI Tecnai G²-20 twin. Beam conditions during ACOM-TEM
163 were 200 kV and spot size 11, giving a nominal 1 nm probe diameter, resulting in a step size of 2
164 nm. During acquisition, the primary electron beam was set to precession movement, with an
165 opening angle of 0.5°. In a separate step, the acquired electron diffraction patterns were matched
166 with a pre-calculated bank file containing the simulated crystal orientations in kinematic
167 conditions, resulting in a unique crystal-orientation solution.

168 3.2 Data treatment

169 Orientation data from EBSD and ACOM-TEM were processed using the MTEX 4.5.2
170 toolbox (Hielscher and Schaeber, 2008; Bachmann *et al.*, 2011). The reference frame was set to
171 x-axis to the east, y-axis to the south and z-axis out of plane. Grain boundaries were defined as
172 misorientation angles $>10^\circ$ and subgrain boundaries were defined as misorientation angles in the

173 range 1–10° for EBSD and 2–10° for ACOM-TEM. Unindexed pixels or single pixels matched as
174 a different phase were removed and unindexed pixels were filled with the average orientation of
175 their grain neighbours. Grains <5 pixels were removed from EBSD datasets. Grains and subgrains
176 <20 pixels were removed from the ACOM-TEM dataset. A Kuwahara filter with a kernel size of
177 5x5 was applied to the ACOM dataset to reduce orientation noise. All crystal orientation plots
178 were visualized before denoising to guard against the introduction of artefacts. Contoured pole
179 figures are based on one-point-per-grain orientation data. The optimum half-width for contoured
180 EBSD pole figures was estimated using the De la Vallée Poussin kernel approach. Because this
181 estimation was inconsistent with the low estimated optimum half-width for the ACOM-TEM data,
182 we chose 15° to match the EBSD pole figures. Misorientation inverse pole figures (MIPF) were
183 plotted for subgrain-boundary misorientation angles of 1–10° for EBSD and 2–10° for ACOM-
184 TEM.

185 **3.3 Grain size analysis**

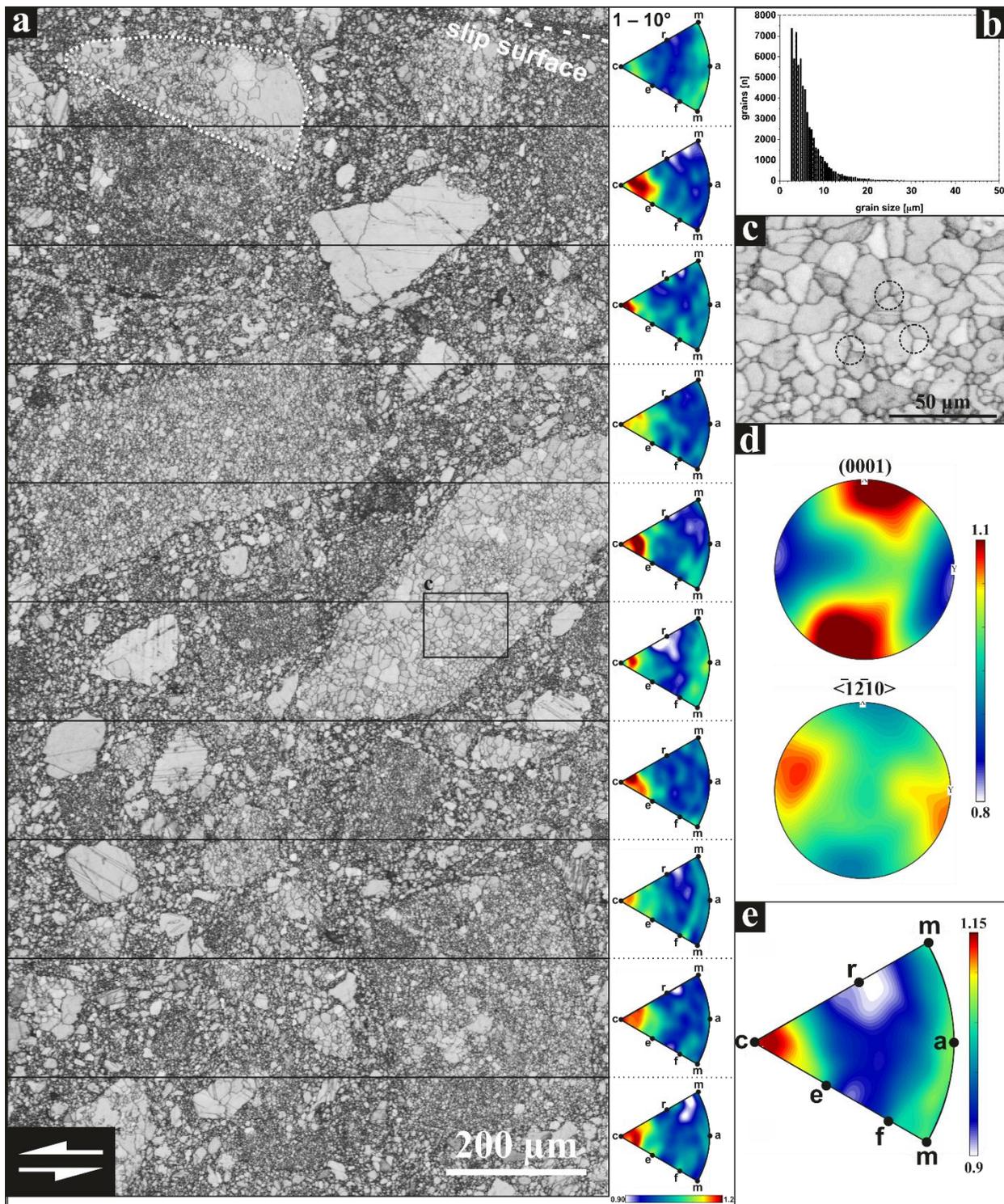
186 A grain-size distribution was determined from the EBSD and ACOM-TEM data. The
187 ACOM-TEM grain-size distribution was based on a grain-boundary trace map by combining a
188 reliability map and an indexed crystal-orientation map. In order to ensure comparability of EBSD
189 and ACOM-TEM data fractal dimension analysis, the calculated grain-size frequencies from the
190 ACOM-TEM data were scaled with the difference in area resolution, due to differences in step
191 size, by a factor of 62500. In this procedure, a 500x500 nm pixel (EBSD) was divided by a 2x2
192 nm pixel (ACOM-TEM) which equates to the factor of 62500. To obtain a grain-size distribution,
193 we chose the dataset binning to be continuous (i.e., equal to the mapping step size), to reduce
194 undersampling of small grains. Each dataset was individually fitted with a linear equation where
195 the negative slope of the linear fit in log-log space equals the fractal dimension D . The grain-size

196 bin width for the fractal-dimension plot was set to 1 μm to adequately subdivide for the large
197 number of small grains.

198 4 Results

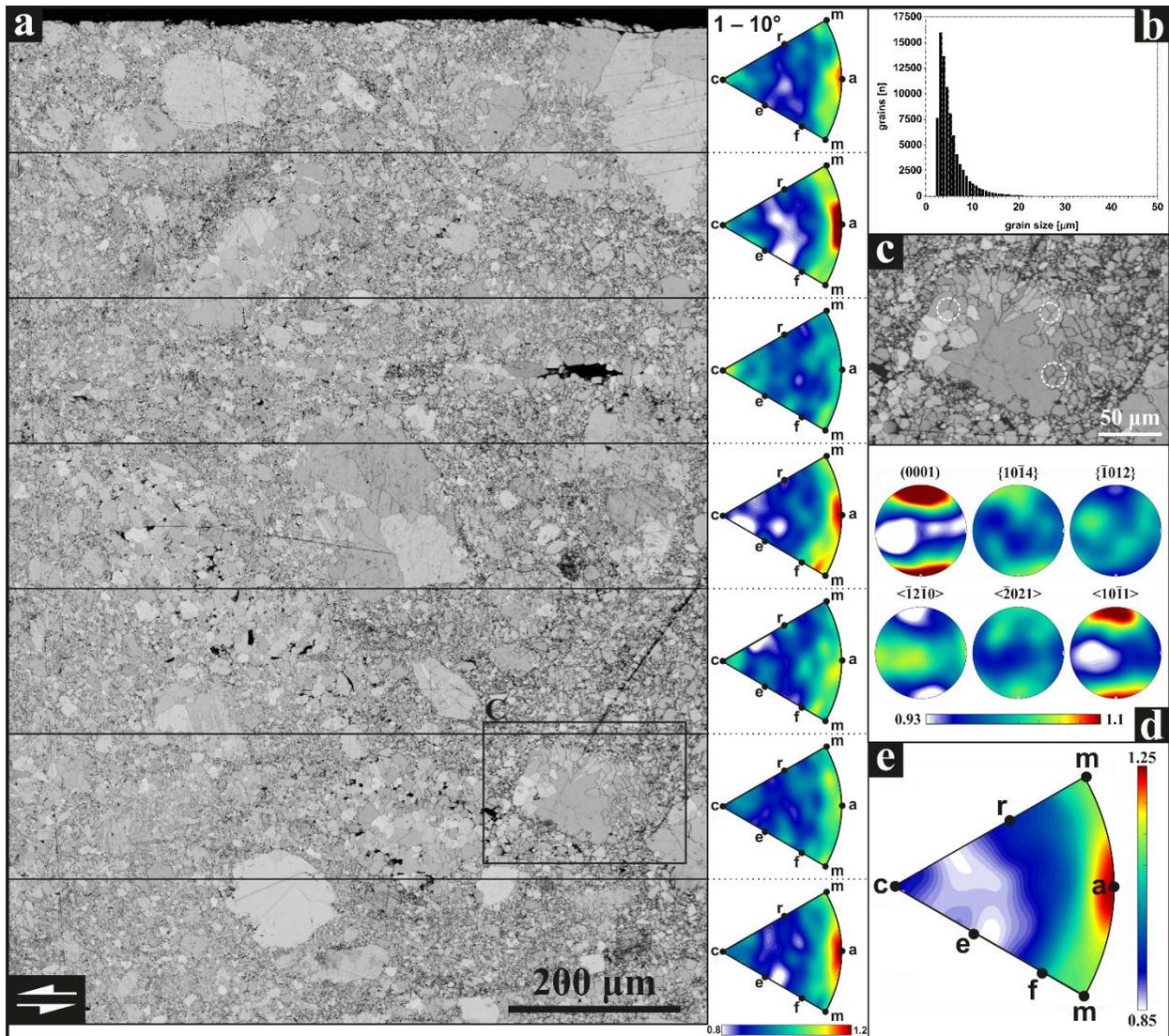
199 4.1 Microscale crystal-orientation data

200 Figure 2a presents the EBSD map of the Arkitsa footwall cataclasite. The map exhibits
201 small matrix-forming grains and larger host-rock clasts, where the clasts show an internal fine-
202 grained foam microstructure. The fine-grained matrix and the foam microstructure display straight
203 grain boundaries that meet in 120° triple junctions (Fig. 2c and a). Grain boundaries are typically
204 not aligned over distances greater than one grain diameter. Monocrystalline calcite clasts
205 occasionally host twin lamellae. An elongated host-rock grain at the top left of Figure 3a exhibits
206 a gradual increase of small grains from monocrystalline to polycrystalline calcite. The median
207 grain size is $5.0 \mu\text{m}$ (Fig. 2b). MIPFs for each subset in Figure 2a reveal concentrations of
208 misorientation axes approximately centred on $[0001]$. The pole figures of (0001) and $\langle\bar{1}2\bar{1}0\rangle$ (Fig.
209 2d) display a weak CPO with multiples of uniform distribution (MUD) in the range 0.8–1.2. The
210 (0001) planes are parallel to the slip plane and the $\langle\bar{1}2\bar{1}0\rangle$ axes are parallel to the slip direction
211 (noting the orientation of the trace of the slip surface at the top right of Fig. 2a). The subgrain-
212 boundary MIPF for the overall map data exhibits a cluster of misorientation axes parallel to $[0001]$
213 (Fig. 2e), like the individual subsets in Fig 2a.



214
 215 *Figure 2: Electron-backscatter diffraction results of the Arkitsa fault-exposure cataclasite. a:* Band-contrast map and
 216 *MIPF for each subsection. Fault surface with hanging-wall in top-right corner. b:* Grain-size distribution. $n_{total} =$
 217 78143 . *c:* Detailed view of host-rock clast microstructure. Black circles mark triple junctions and 120° angles. *d:* Pole
 218 *plots of (0001) planes and $\langle 1\bar{2}10 \rangle$ axes in the same reference frame as the map in a. e:* MIPF of the full map area.
 219 *Labels indicate crystal directions or plane normals. Contours are multiples of uniform distribution.*

220 Figure 3a presents the EBSD results from the Schinos footwall cataclasite. The band-
221 contrast map reveals a microstructure with large calcite host-rock grains incorporated into the
222 cataclasite matrix. Like Figure 2a, several host-rock grains exhibit an increase of small grains from
223 monocrystalline to polycrystalline (Fig. 3c). Whilst many grain boundaries are curved, several in
224 both, the matrix and host-rock grains are straight and meet in 120° triple junctions (Figure 3c,
225 white circles). The outer margins of the host-rock grains display a rim with grain boundaries,
226 creating an incipient core-mantle structure (Fig. 3c). The median grain-size is $4.4 \mu\text{m}$. (Fig. 3b).
227 The pole figures in Figure 3d display a weak CPO with multiples of uniform distribution (MUD)
228 in the range 0.8–1.2. The (0001) planes are parallel to the slip plane and the $\langle\bar{1}2\bar{1}0\rangle$ axes are
229 parallel to the slip direction. Furthermore, $\{10\bar{1}4\}$ poles exhibit a weak cluster approximately
230 parallel to the slip-plane normal and the $\langle\bar{2}021\rangle$ axes exhibit three maxima sub-perpendicular to
231 the slip plane. In addition, $\{\bar{1}012\}$ planes exhibit one maximum and a girdle, whereas $\langle 10\bar{1}1\rangle$
232 directions are oriented perpendicular to the slip plane. MIPFs for subgrain-boundary
233 misorientation axes in each vertical section in Figure 3a exhibit a pronounced maximum centred
234 on the $\langle a\rangle$ direction. Secondary maxima are centred on $\langle m\rangle$, $\langle c\rangle$, or $\langle a\rangle$, or a combination of
235 all three directions. The overall MIPF in Figure 3e exhibits subgrain misorientation axes
236 predominantly around $\langle a\rangle$, consistent with most misorientation axes in the vertical sections from
237 Figure 3a.



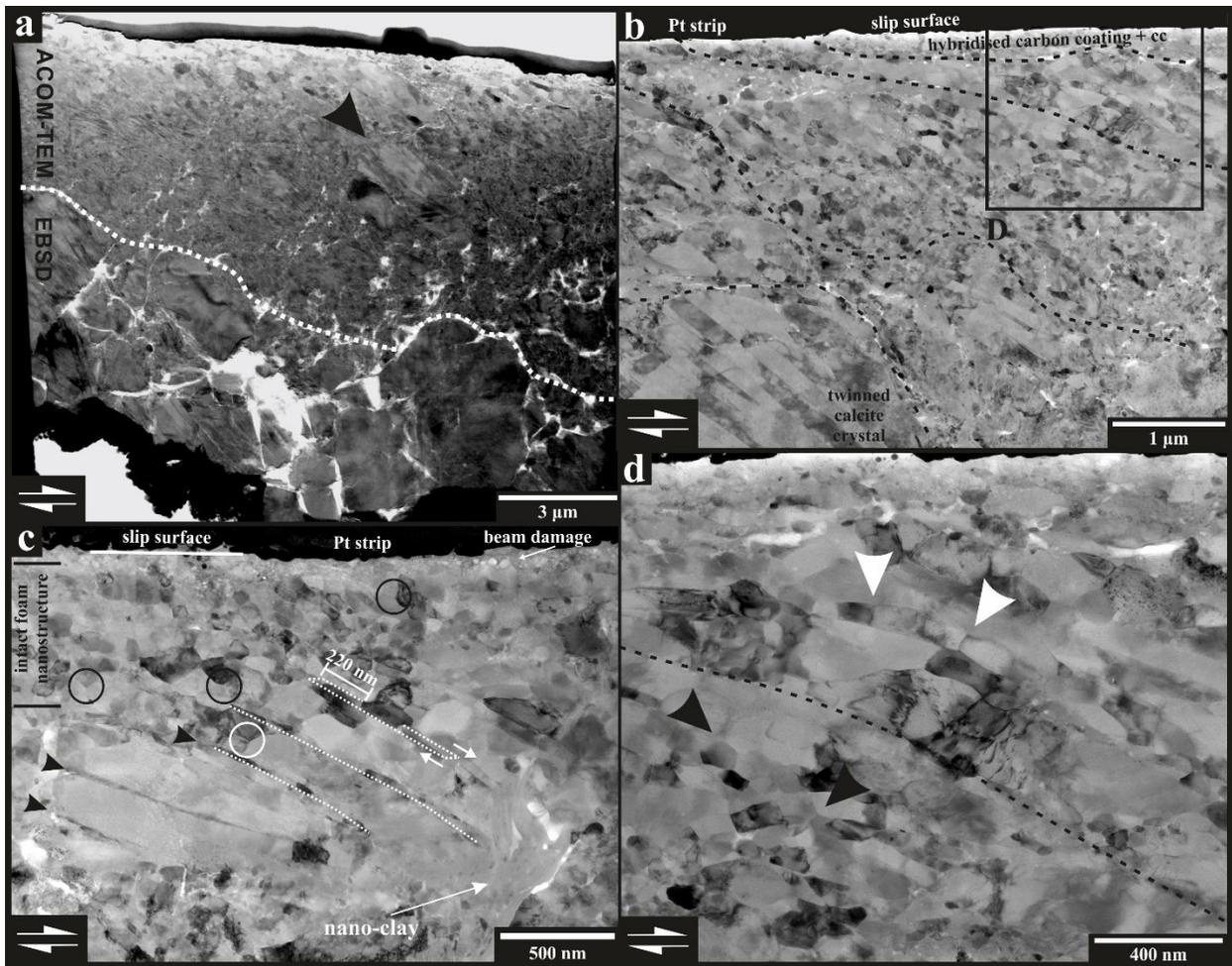
238
 239 *Figure 3: Electron-backscatter diffraction results of the Schinos fault-exposure cataclasite. **a:** Band-contrast map and*
 240 *MIPF for each subsection. Fault surface at the top (black). **b:** Grain-size distribution. $n_{total} = 90803$. **c:** Clast in matrix*
 241 *displaying a mantle of grains around a host-rock clast with internal triple junctions (white dashed circle). **d:***
 242 *Combined pole plots of relevant slip systems from **a**. **e:** Misorientation inverse pole figure from the full dataset in **a**.*

243 4.2 Nanostructures

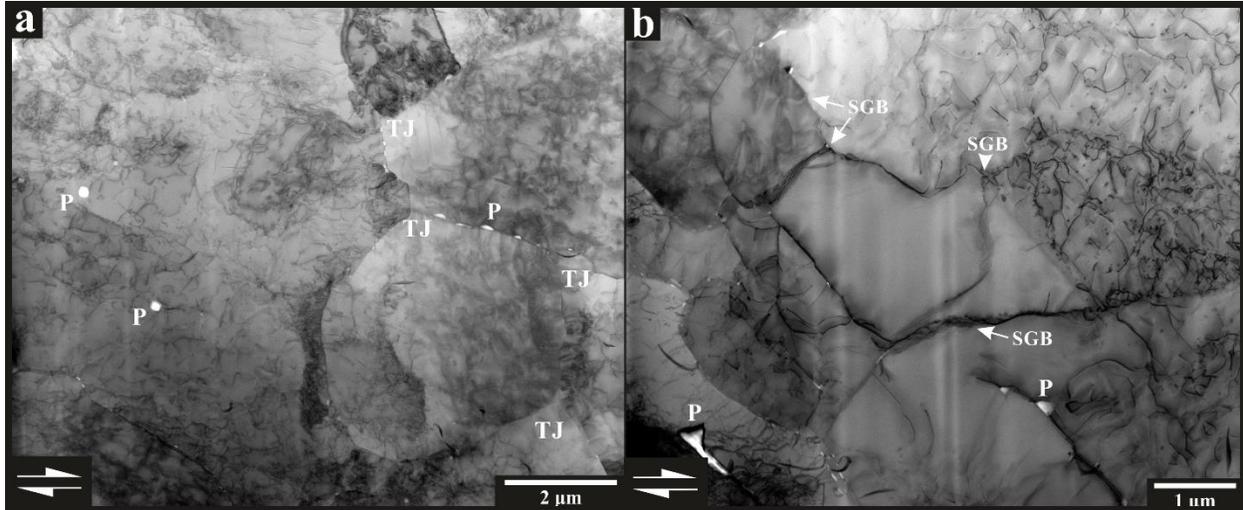
244 TEM investigation of the Arkitsa fault rock also reveals a fine-grained volume situated on
 245 top of coarser grains (Fig. 4a). The first 15–20 μm of material directly below the PSS exhibits a
 246 foam nanostructure. This foam nanostructure consists of grains with approximately equal grain
 247 size and straight grain boundaries that meet in triple junctions with 120° angles (Fig. 4c). The
 248 grains in this zone are commonly sandwiched between, and overprinted by, microstructural

249 discontinuities (e.g., Fig. 4c, white lines) dipping at an angle of about 30° to the slip surface into
250 the nanogranular material. The discontinuities can displace single grains (Fig. 4b and c) or form
251 bands of localised deformation with a sigmoidal appearance, preserving the intact foam
252 nanostructure in between (Fig 4b and d). In Figure 4c, grains with similar diffraction contrast are
253 displaced about 220 nm along such microstructural discontinuities. These discontinuities cannot
254 be traced to the slip surface (Fig. 4c) but terminate in an area with a smaller grain of ~50 nm size
255 below the PSS (Fig. 4c) compared to ~300 nm further away from the PSS (Fig. 6, grain No. 3).
256 Larger grains are occasionally intermingled with the nanogranular material (Fig. 4a). Below the
257 nanogranular material, twinned calcite grains of 3–5 µm in diameter mark the beginning of the
258 cataclasite (Fig. 4a). The grain size at the transition between the slip-surface nanostructure and the
259 larger grains corresponds to the grain sizes observed in the EBSD map (Fig. 4 and d).

260 Figures 5a and b present the nanostructure of the Schinos fault directly at the PSS.
261 Compared to the Arkitsa sample (Figure 4a and c), the grain size is larger, resulting in a less
262 complex nanostructure. The Schinos nanostructure exhibits straight grain-boundary morphology
263 with triple junctions (Fig. 5a) and subgrain boundaries (Fig. 5b). The average dislocation density
264 in the larger Schinos grains is $\sim 1.5 \times 10^{13} \text{ m}^{-2}$. The dislocation density decreases towards the
265 subgrain boundaries but otherwise the distribution is generally homogeneous except for some
266 subgrain interiors that are devoid of dislocations (Fig. 5b).



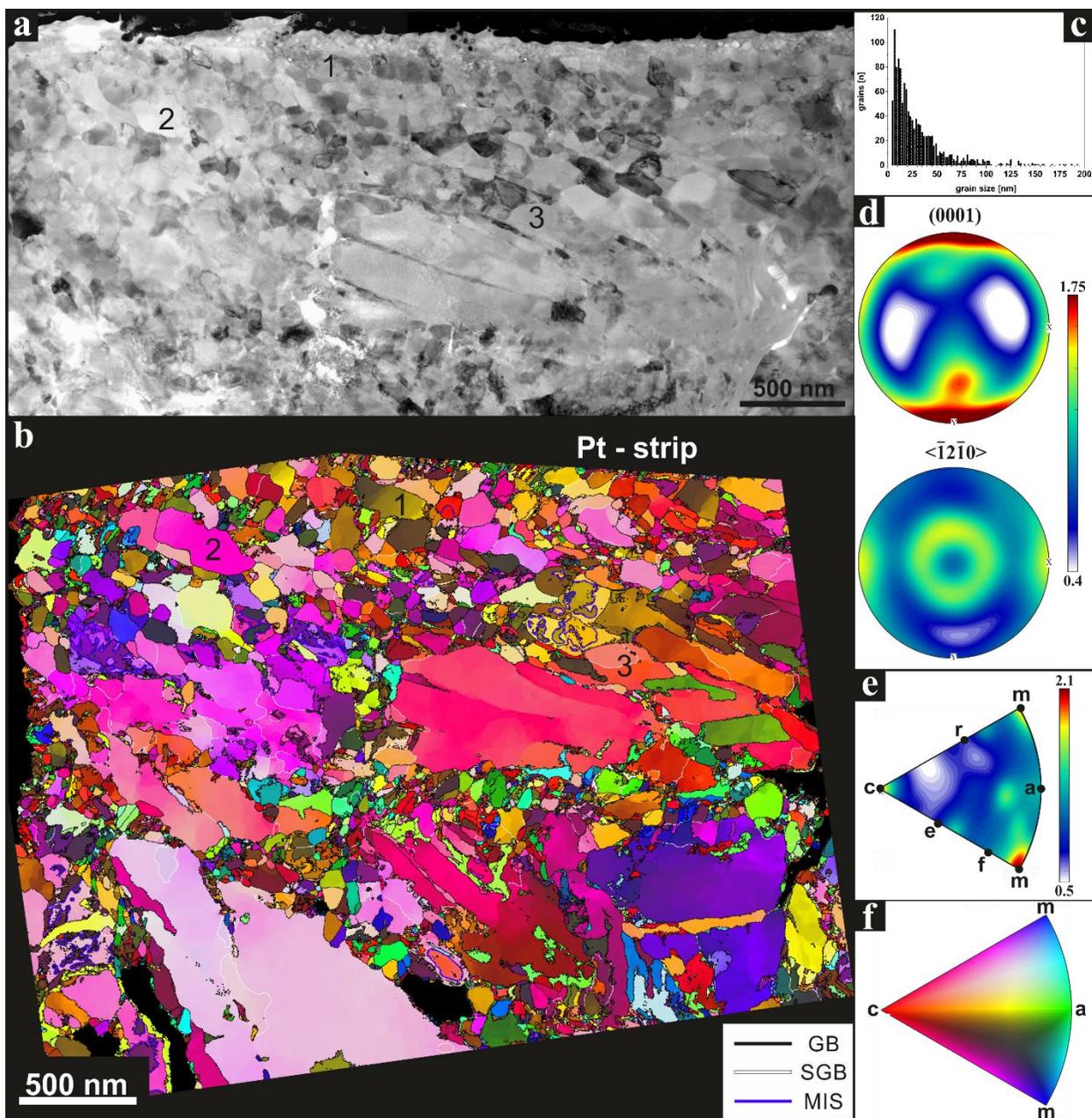
267
 268 **Figure 4:** Arkitsa fault exposure nanostructures. **a:** Bright-field (BF) TEM overview of deformed volume with a sharp
 269 boundary to the footwall cataclasite (dashed line). Larger grains show strong deformation (black arrow). The grain
 270 size of the less deformed grains is about 3–5 μm. **b:** Bright-field STEM image with detailed view of the deformed
 271 volume. Anastomosing boundaries separate alternating domains of deformed and intact foam nanostructure (dashed
 272 lines). **c:** Bright-field STEM image showing intact foam nanostructure with triple junctions and 120° angles adjacent
 273 to the slip surface (black circles). Fractures that dissect grains terminate inside intact foam nanostructure. Fractures
 274 appear to evolve from former cleavage planes (black arrows). Older foam nanostructure is preserved between fracture
 275 planes (white circle). **d:** Bright-field STEM image of detailed view from **b**. Deformed foam nanostructure with former
 276 triple junctions while having a sheet-like structure (white arrows) next to intact foam nanostructure (black arrows).



277
 278 *Figure 5: Schinos fault exposure nanostructures. **a:** BF-STEM image with overview of dislocation structure showing*
 279 *triple junctions (TJ) and grains with dislocation densities of $1.5 \times 10^{13} \text{ m}^{-2}$ and higher. **b:** BF-STEM image with*
 280 *dislocation-free subgrain in the centre surrounded by subgrain boundaries (SGB). Dislocation density of surrounding*
 281 *grain interiors decreases towards the SGBs. P = pores.*
 282

283 4.3 Nanoscale crystal-orientation (ACOM-TEM) data

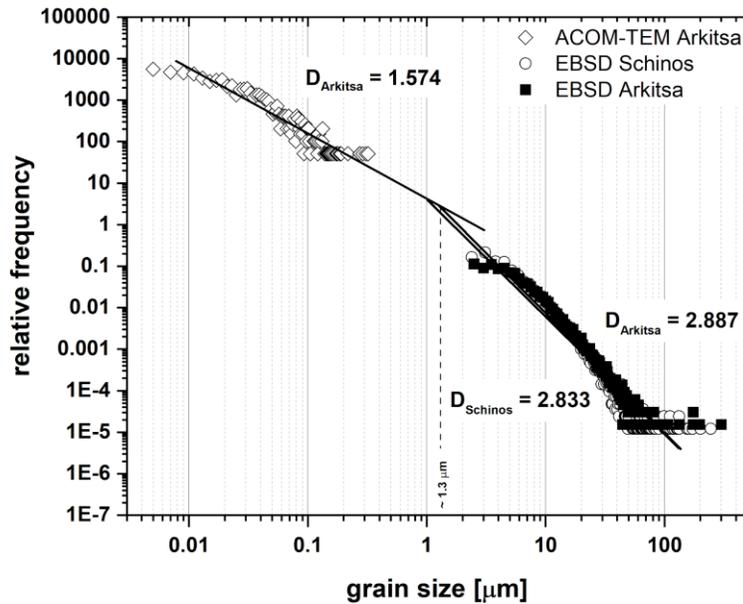
284 Figure 6 presents the ACOM-TEM data acquired on a subset of the same FIB foil shown
 285 in Figure 4c, reproducing the bright field (BF-)TEM nanostructure (Fig. 4c, 6a and b). Pole figures
 286 constructed from the crystal-orientation map exhibit a CPO with (0001) plane-normal densities in
 287 the range 0.4–2.0 MUD in the highly deformed, fine-grained region below the PSS. Some grains
 288 exhibit an orientation spread indicating intragranular misorientation (Fig. 6b). The median grain-
 289 size is 21 nm (Fig. 6c), albeit ranging between 5 to 300 nm. Contoured pole figures (Fig. 6c) reveal
 290 a CPO with [0001] axes oriented perpendicular to the slip surface and $\langle \bar{1}2\bar{1}0 \rangle$ axes clustered sub-
 291 parallel to slip direction. A second clustering of $\langle \bar{1}2\bar{1}0 \rangle$ axes appear as a ring around the centre
 292 of the pole figure. The MIPF of the subgrain misorientation axes exhibits maxima parallel to [c]
 293 and $\langle m \rangle$.



294
 295 *Figure 6: Nanoscale crystal orientation map. Numbers indicate the same grains for comparison between a and b. a:*
 296 *BF-STEM image from Figure 4C. b: Crystal-orientation map colour-coded according to the inverse pole figure in f*
 297 *indicating the crystal direction aligned with the Y-axis of the map. c: Grain-size distribution of map in b. d: Contoured*
 298 *pole figures of (0001) poles and $\langle \bar{1}2\bar{1}0 \rangle$ axes. e: MIPF of misorientation axes associated with misorientation angles*
 299 *in the range 2–10°. f: IPF-Y colour key for map in b. GB = Grain boundary, SGB = Subgrain boundary, MIS =*
 300 *Misindexed grain boundary. Due to the electron-transparent nature of the FIB foil and corresponding diffraction*
 301 *behaviour, grain boundary morphologies are less well defined in the ACOM-TEM data compared to the BF-STEM*
 302 *image.*

303 **4.4 Grain-size distribution**

304 Figure 7 presents a log-log plot of relative frequency as a function of grain size from the
305 EBSD and ACOM-TEM data. A data gap between 350 nm and 2 μm arises from the different
306 spatial resolutions and area coverage of the two techniques. The EBSD-based fractal dimension of
307 the Arkitsa fault exposure is $D = 2.887$ ($R^2 = 0.912$), while the fractal dimension of the ACOM-
308 TEM data is $D = 1.574$ ($R^2 = 0.895$). The EBSD-based fractal dimension of the Schinos is
309 $D = 2.833$ ($R^2 = 0.902$). Extrapolations of the grain-size distributions measured from the two
310 different image datasets intersect at a grain size of approximately 1 μm .



311
312 *Figure 7: Fractal dimension plot of grain-size data from both fault exposures. The fractal dimensions of the Arkitsa*
313 *datasets are $D = 2.887$ (EBSD) and $D = 1.574$ (ACOM-TEM). The fractal dimension of the Schinos dataset is $D =$*
314 *2.833 (EBSD).*

315

316

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320 5 Discussion

321 5.1 Grain fragmentation and fractal dimensions

322 Brecciation and cataclasis are important mechanisms of grain-size reduction in fault zones.
323 Whereas intragranular extensional fracturing governs cataclasis during early fault-slip through
324 particle-particle fragmentation, chipping governs the late stages during which grain edges are
325 removed after greater amounts of fault displacement (Billi, 2010; Ferraro *et al.*, 2018). Cataclasis
326 can produce different grain-size distributions with fractal dimensions (D) that provide information
327 on the characteristics of fracturing (e.g., Sammis *et al.*, 1986; Sammis *et al.*, 1987 and Blenkinsop,
328 1991). For example, a fractal dimension of $D = 2.580$ can result from the self-similar fracturing of
329 a three-dimensional object, such as a cube. In addition to obtaining D via linear fitting, one can
330 also determine D via:

$$331 \quad D = \frac{3 \log(f)}{\log(F)} + 3 \quad (1)$$

332 where F is the number of fragments created, and f is the fragmentation fraction, defined as $f = C/F$,
333 with C being the number of fragments that are fragmented further. Fragmentation of a cube
334 produces eight cubes ($F = 8$) of $1/2$ the width of the original cube (Heilbronner and Barrett, 2014).
335 With $F = 8$, it follows that for $f = 8/8$, 100 % of the newly formed grains are fragmented again,
336 which results in a fractal dimension of $D = 3.000$. Our fractal dimensions of $D = 2.887$ and $D =$
337 2.833 (Fig. 7) can be achieved with a fragmentation fraction of $f = 7/8$, giving $D = 2.807$. Such
338 high D -values are reported for natural faults with intense grain-size reduction (Billi and Storti,
339 2004) and are predicted by numerical simulations (Abe and Mair, 2005). The agreement between
340 the theoretical and our measured values suggests that the cataclasite experienced a high degree of
341 fragmentation due to particle-particle interaction. Furthermore, a value of $D = 1.574$ from ACOM-
342 TEM (Fig. 7) may correspond to a low degree of fragmentation with $f = 3/8$ yielding $D = 1.585$

343 (see eq. 1). We propose that the difference in D between the bulk cataclasite and the nanogranular
344 volume arises from a difference in the degree of fragmentation. A lower D of 1.574 may, therefore,
345 indicate a different control on particle size involving a minor degree of particle-particle
346 fragmentation. We suggest that the change in fractal dimension within the same fault rock may
347 reflect a change in fragmentation and thus deformation mechanisms, as also proposed by Keulen
348 *et al.* (2007).

349 **5.2 Nanostructures**

350 The Arkitsa and Schinos faults exhibit different nanostructures in their principal slip zones
351 (PSZs). Whereas the PSZ of the Arkitsa fault is complex directly below the slip surface and
352 includes a layer of nanograins (Figs. 4 and 6), the PSZ of the Schinos fault exhibits a similar grain
353 size as its bulk fault rock (Figs. 3 and 5). We propose the difference in nanostructural complexity
354 are because the Schinos fault represents an earlier stage of fault-rock evolution, while the Arkitsa
355 fault accommodated multiple slip events over an extended deformation history.

356 Slip along the PSS would result in the introduction of plastic strain accompanied by a
357 thermal spike through shear heating (Rice 2006) during a seismic event. The Schinos
358 nanostructure, with a high, free dislocation density and triple junctions (Fig. 5a and b), resembles
359 that of metals subjected to a process known as cold-rolling and annealing (Humphreys and
360 Hatherly, 2004). The procedure involves the introduction of high plastic strain followed by static
361 high-temperature treatment to induce microstructural changes. The typical range for industrial
362 cold-rolling is about 60–180 °C (Hollandt *et al.*, 2010), corresponding to 0.05–0.11 times the
363 melting temperature, T_m , for steel. It is likely that the temperature during the onset of slip of the
364 carbonate faults was at a homologous temperature of about 0.2 T_m (300 °C). Cold-rolling and
365 subsequent annealing is a well-established process in engineering leading to grain-boundary

366 migration and recrystallisation (Humphreys and Hatherly, 2004). Dislocation introduction through
367 strain pulses in the low-temperature plasticity regime can result in strain-hardening effects.
368 Addition of thermal energy through heating enables dislocation climb and solid-state diffusion,
369 leading to recovery or recrystallisation by static grain growth or grain boundary migration. The
370 resulting grain size post annealing is smaller compared to the previous microstructure leading to
371 grain-boundary strengthening and hence, toughening of the material. Such deformation processes
372 followed by annealing of the material are already documented in experimentally and naturally
373 deformed olivine (Druiventak *et al.*, 2012; Matysiak and Trepmann, 2012) and quartz (Trepmann
374 and Stöckhert, 2013; Trepmann *et al.*, 2017). Repeated straining and subsequent annealing can
375 lead to grain-size reduction and may, therefore, pose a mechanism of nanograin formation.

376 The 120° triple junctions of the Arkitsa nanostructure may indicate annealing by grain
377 boundary migration (Figs. 2 and 4). Static recrystallization involves an initial stage during which
378 deformed grains with high, stored strain energy are replaced by recrystallized grains, which may
379 then continue to grow. To evaluate whether significant grain growth can occur during the
380 postseismic and inter-seismic period, we use the following kinetic model (Covey-Crump, 1997),

$$381 \quad d^{1/n} - d_0^{1/n} = k t = k_0 t \exp(-H/RT) \quad (2)$$

382 where d is the final grain size, d_0 the initial grain size, n is a dimensionless constant, k_0 is a pre-
383 exponential factor, t the duration of grain growth and H is the apparent activation enthalpy. The
384 values of n and H depend on the growth-controlling process. In the case of a grain-boundary
385 controlled system, with no second phases (pure system) $n = 0.5$. For an impure system where
386 coalescence of a second phase occurs by volume diffusion (wet case) $n = 0.33$ and for an impure
387 system where coalescence of a second phase occurs by grain-boundary diffusion, $n = 0.25$ (Covey-
388 Crump, 1997). Assuming fluid-present conditions based on observations that suggest the presence

389 of portlandite ($\text{Ca}(\text{OH})_2$) during deformation (Ohl et al. 2020), we set $n = 0.33$. This interpretation
390 results in the following parameters: $k_0 = 2.514 \times 10^9 \mu\text{m}^{1/n} \text{s}^{-1}$, $1/n \approx 3$ and $H = 173.6 \text{ kJ mol}^{-1}$
391 (Covey-Crump, 1997). To assess the potential for fluid-assisted post-seismic grain growth due to
392 the ambient temperature at depth, we consider the borehole temperatures from the outcrop areas
393 (Metaxas *et al.*, 2010; Papoulis *et al.*, 2013; Lambrakis *et al.*, 2014). We assume a geothermal
394 gradient of 65–75 °C/km and a typical seismogenic crustal depth of 3–5 km (Scholz, 1988)
395 resulting in an ambient temperature of about 300 °C. Annealing of the nanostructure for one year,
396 at a temperature of 300 °C, with $d_0 = 0.1 \mu\text{m}$, leads to a final grain size of $d = 2.3 \mu\text{m}$. Therefore,
397 not only under short-lived, co-seismic temperature spikes but also during the inter-seismic period,
398 grain growth may contribute to the formation and modification of the microstructure. However,
399 the grain-size distribution in Figure 6b contains grains $< 50 \text{ nm}$ in size, illustrating that our grain-
400 growth approximation provides an upper limit. Nonetheless, our assessment of inter-seismic grain
401 growth supports our suggestion that a cohesive nanogranular fault rock may be generated by high-
402 plastic strain deformation and short annealing times.

403

404 **5.3 Deformation mechanisms**

405 **5.3.1 Grain-boundary sliding**

406 GBS has been proposed as a deformation mechanism for fine-grained fault rocks during
407 seismic slip (De Paola *et al.*, 2015). Langdon (2006) describes two possible types of GBS:
408 Rachinger sliding and Lifshitz sliding. Rachinger sliding is defined by the relative displacement
409 of adjacent grains, with strain compatibility maintained by dislocation motion in grain interiors.
410 Therefore, Rachinger sliding is commonly referred to as dislocation-accommodated grain-
411 boundary sliding in the geological literature (Hirth and Kohlstedt, 1995; Hansen *et al.*, 2011). In

412 contrast, Lifshitz sliding is coupled to vacancy diffusion along stress gradients during Nabarro-
413 Herring or Coble diffusion creep. GBS is an essential process that contributes to superplasticity,
414 which is the ability of a material to deform to strains on the order of 1000% without failure
415 (Langdon, 2006; Komura *et al.*, 2001). The term superplasticity does not indicate a deformation
416 mechanism but is a phenomenological description. In experiments on metals (Langdon, 2006) and
417 calcite (Schmid *et al.*, 1977; Rutter *et al.*, 1994) superplastic behaviour is most pronounced in a
418 regime in which strain rate is proportional to approximately the square of both stress and grain
419 size. This mechanical behaviour is associated with Rachinger sliding in materials with grains that
420 are generally too small to host subgrain boundaries (Langdon, 2006). An important consideration
421 for seismogenic faults is that experiments by Komura *et al.* (2001) on metals demonstrate a strong
422 strain-rate dependence for superplasticity, where strain rates $> 1 \text{ s}^{-1}$ reduce the achievable strain
423 from 1000 % down to 100 %. This observation presents a challenge to the interpretation of
424 superplastic behaviour from micro-, or nanostructures in the high-strain rate context of co-
425 seismically produced materials.

426 In many metals, GBS is proposed as a deformation mechanism of nanogranular materials.
427 The in-situ TEM deformation study by Kumar *et al.* (2003) on nanograined Ni with grain sizes $<$
428 30 nm revealed that GBS can be an important deformation mechanism even at room temperature.
429 Those authors report the involvement of dislocations during the deformation process and
430 emphasize the dominant role of dislocation-mediated plasticity. Experimental evidence suggests
431 that at grain sizes of < 20 nm the material strength decreases and produces an inverse Hall-Petch
432 effect (Kumar *et al.*, 2003). Another study by Lu *et al.* (2000) also indicates that GBS may be
433 significant in nanomaterials at lower homologous temperatures. At grain sizes below 10 nm,
434 dislocation activity ceases and GBS dominates. Whether *in situ* nanoscale deformation behaviour

435 within a TEM can be generalised to be representative of bulk deformation behaviour remains a
436 matter of debate (Ma, 2004). Nevertheless, deformation of materials with grain sizes ≥ 30 nm that
437 involves GBS can also involve dislocation activity. The combination of dislocations we observe
438 (Figs. 4 & 5), subgrain boundaries in EBSD (Figs. 2 & 3), and the nanoscale CPO consistent with
439 the activity of known slip systems (Figure 6) suggests that dislocation activity plays an important
440 role during the formation and deformation of the nanostructure.

441 A mechanism that combines GBS and dislocation activity is disGBS and has been proposed
442 as a deformation mechanism for several minerals, including calcite (Walker *et al.*, 1990), olivine
443 (Hirth and Kohlstedt, 1995; Hansen *et al.*, 2011), and quartz (Tokle *et al.*, 2019). Based on the
444 microstructures and mechanical data from their experiments on olivine, Hansen *et al.* (2011)
445 propose a similar disGBS mechanism to the model by Langdon (2006), in which the subgrain size
446 is smaller than the grain size. Dislocation activity during disGBS may be an explanation for the
447 CPO observed by Hansen *et al.* (2011) and may be an alternative interpretation to crystal plasticity
448 for the micro- and nanostructure observed here. Schmid *et al.* (1977) and Walker *et al.* (1990)
449 observed displacements across grain boundaries on the pre-cut surfaces of split cylinders deformed
450 in regimes with non-linear stress dependencies. Rutter *et al.* (1994) use the similarities of stress
451 and grain-size exponents which fit with the later proposed model by Langdon (2006). Likewise,
452 several studies (e.g., Schmid *et al.*, 1977; Walker *et al.*, 1990; Rutter *et al.*, 1994) have measured
453 regimes in which the stress and grain-size exponents of calcite are broadly in agreement with the
454 models of disGBS reviewed by Langdon (2006). Rutter *et al.*, (1994) report a CPO apparently
455 formed during high-temperature creep deformation, where one of the experiments reached a strain
456 of 600–1000 %, representing superplastic flow. Those authors interpreted their results to indicate

457 a contribution from intracrystalline plastic flow involving cyclic dynamic recrystallisation but did
458 not exclude the contribution of GBS.

459 High-strain torsion experiments ($\gamma = 20$) by Barnhoorn *et al.* (2005), however, demonstrate
460 that post-deformational annealing can change the microstructural appearance and produce a foam
461 structure where the grain morphologies are indistinguishable from a GBS microstructure. The CPO
462 formed during initial deformation is enhanced with progressive annealing as the axis distributions
463 become tighter. In addition, the calcite deformed by Barnhoorn *et al.* (2005) has microstructural
464 characteristics indicating incomplete reworking of the starting material used and shares similarities
465 with our microstructure (Fig. 2). Specifically, the slightly lobate grain boundaries and not ideal
466 triple junctions of the foam microstructure are comparable. These similarities and a pronounced
467 CPO across different scales suggest that the microstructures of the studied carbonate faults may
468 be influenced by other deformation processes e.g., crystal plasticity, than exclusively GBS.

469 **5.3.2 Crystal-plasticity**

470 The occurrence of CPOs suggests the activation of one or more slip systems in both Greek
471 faults. Multi-scale analysis of crystal orientations (Figs. 2, 3 and 6) reveals that the CPO present
472 at the nanoscale in the PSZ is also present in the adjacent cataclasite. The distributions of (0001)
473 planes and $\langle \bar{1}2\bar{1}0 \rangle$ axes from the Arkitsa fault are consistent with CPOs present in previous
474 carbonates experimentally deformed under both seismic and sub-seismic conditions (Smith *et al.*,
475 2013; Verberne *et al.*, 2013; Kim *et al.*, 2018; Demurtas *et al.*, 2019; Pozzi *et al.*, 2019). However,
476 the experimental studies have not yet provided detailed slip-system analyses. The combined
477 evidence of calcite (0001) planes aligned parallel to the slip plane, $\langle \bar{1}2\bar{1}0 \rangle$ axes aligned parallel
478 to the slip direction and the distribution of subgrain-misorientation rotation axes indicates the
479 activation of the (0001) $\langle \bar{1}2\bar{1}0 \rangle$ glide system (Figure 2d and e). Subgrain-boundary misorientation

480 axes (Figure 2e) parallel [0001] are consistent with the presence of twist boundaries parallel to the
481 (0001) plane and consisting of $\langle \bar{1}2\bar{1}0 \rangle$ screw dislocations whilst misorientation axes around
482 $\langle 10\bar{1}0 \rangle$ are consistent with the presence of tilt boundaries consisting of (0001) $\langle \bar{1}2\bar{1}0 \rangle$ edge
483 dislocations. Both types of boundaries can be produced by activation of the (0001) $\langle \bar{1}2\bar{1}0 \rangle$ glide
484 system. We note that the ring pattern in the centre of the $\langle \bar{1}2\bar{1}0 \rangle$ pole figure (Figure 6c) is likely
485 an artefact arising from diffraction pattern indexing during ACOM-TEM analysis. De Bresser and
486 Spiers (1997) performed a detailed experimental study on calcite single crystals, in which they
487 identified slip systems based on analysis of the traces of slip bands. In their experiments, the
488 (0001) $\langle \bar{1}2\bar{1}0 \rangle$ slip system was activated in the temperature range of 600–800 °C.

489 In contrast to the Arkitsa fault, misorientation axes of subgrain boundaries in the Schinos
490 fault are dominantly parallel to $\langle \bar{1}2\bar{1}0 \rangle$, with only secondary maxima parallel to $\langle 10\bar{1}0 \rangle$ and
491 [0001] (Fig. 3). Misorientation axes parallel to $\langle \bar{1}2\bar{1}0 \rangle$ indicate the presence of subgrain
492 boundaries consisting of edge dislocations on the $f\{\bar{1}012\}\langle 10\bar{1}1 \rangle$ or $r\{10\bar{1}4\}\langle \bar{2}021 \rangle$ slip systems.
493 In the experiments of (De Bresser and Spiers, 1997) the $f\{\bar{1}012\}\langle 10\bar{1}1 \rangle$ slip system was activated
494 at temperatures between 600–800 °C, while $\{r\}$ slip was activated over a broader temperature
495 range of 300–800 °C. These two slip systems also exhibit different critical resolved shear stress
496 (CRSS). At temperatures > 600 °C, the CRSS for $f\langle 10\bar{1}1 \rangle$ is less < 20 MPa and for $r\langle \bar{2}021 \rangle$ is
497 ≤ 10 MPa. Overall, we suggest that the misorientation axes around $\langle \bar{1}2\bar{1}0 \rangle$ (Figure 3a and e) most
498 likely originate from edge dislocations on the $r\langle \bar{2}021 \rangle$ slip system as the CPO indicates that this
499 system is more favourably aligned for slip than is the $f\langle 10\bar{1}1 \rangle$ system. The change from rotation
500 around $\langle a \rangle$ to additional rotation around [0001] and $\langle m \rangle$ indicates the activation of more than one
501 slip system, in particular the additional activation of (0001) $\langle \bar{1}2\bar{1}0 \rangle$. The high temperatures
502 indicated by the misorientation analyses are in agreement with our previous estimates for these

503 faults of 600–800 °C, but < 1000 °C, based on the degree of sp² hybridisation of partly-hybridised
504 amorphous carbon (Ohl *et al.*, 2020). Whether or not the potential high-temperature signals are
505 diagnostic for deformation at co-seismic velocities warrants further investigation. Because a
506 systematic experimental study of slip systems in sub-seismic and seismically deformed carbonate
507 fault rocks is lacking, more experiments are required to investigate potential differences in CPOs,
508 including between dry and wet environmental conditions.

509 To evaluate whether changes in slip systems indicate shear-heating induced temperature
510 gradients, we analysed EBSD subsets over a range of distances from the PSS to test for systematic
511 variation in the temperatures associated with the recorded slip systems (De Bresser and Spiers,
512 1997). We find that overall, the Arkitsa (Fig. 2a) and Schinos (Fig. 3a) fault rocks do not exhibit
513 systematic changes in misorientation axes and hence slip systems or associated temperatures with
514 distance from the PSZ. If the faults experienced seismic slip, a temperature gradient was not
515 recorded. However, the Schinos fault does display a non-systematic variation in the intensities of
516 misorientation-axes maxima parallel to $\langle\bar{1}2\bar{1}0\rangle$ and [0001], suggesting variation in the
517 contributions of *r*-slip and (c)<a>. The underlying cause for these non-systematic changes in
518 misorientation axes remains unknown and warrants further investigation. Nevertheless, if we can
519 reliably apply the slip system-temperature correlations from De Bresser and Spiers (1997), the
520 common feature of both faults is the high temperatures suggested by the activation of specific slip
521 systems. However, we note that the experiments carried out by De Bresser and Spiers (1997) were
522 performed at $3 \times 10^{-5} \text{ s}^{-1}$ and extrapolation of the results to higher strain rates should be undertaken
523 with caution.

524 Combined numerical models and deformation experiments by Demurtas *et al.* (2019)
525 indicate that a temperature-increase of approximately $\Delta T = 620 \text{ °C}$ decays to about 50 °C over a

526 thermal diffusion distance of 2 mm inside carbonate fault gouge with 1 second. Assuming a single
527 shear-heating event, the resulting temperature diffusion front could be captured as a change in
528 activated slip systems and associated CPOs. However, the absence of differences in slip systems
529 with decreasing temperature away from the PSS may suggest a later thermal overprint of the
530 cataclasite by more than one event. Based on the microstructures in Fig. 4a, this overprint may
531 lead to annealing of the microstructure and a loss of an apparent temperature diffusion profile.
532 Consequently, the analysed cataclasite could contain several slip surfaces which are no longer
533 discernible. The agreement between CPO and subgrain misorientations suggests that crystal
534 plasticity was the main deformation process to produce the CPO rather than other, more exotic
535 CPO-formation mechanisms such as surface energy interactions (Toy *et al.*, 2015) or coupled
536 solution and growth (Power and Tullis, 1989). Overall, our results show that crystal plasticity
537 played a role within the whole fault rock volume.

538 Water can have an influence on crystal-plastic deformation. It is known for quartz that a
539 higher water content can result in a transition of active slip systems from slip in the $\langle a \rangle$ directions
540 to slip in the $[c]$ direction (Blacic, 1975) and a similar trend is observed by (Tokle *et al.*, 2019)
541 where added water can result in a different stress exponent. The temperature threshold for the
542 transition between different dislocation creep regimes in quartz can also be lowered by about 100
543 °C by the addition of water (Hirth and Tullis, 1992). However, Stipp *et al.* (2002) point out that
544 the regimes identified by Hirth and Tullis (1992) may correspond to different types of dynamic
545 recrystallisation. The effect of water content on fabric transition is also known from experiments
546 on olivine where for example type-B ((010)[001]) and type-C ((100)[001]) CPOs are more
547 common with higher water content, whereas type-A ((010)[100]) is most common without water
548 (Jung and Karato, 2001). Deformation experiments on wet calcite at seismic velocities show a

549 more significant drop in friction coefficient compared to dry experiments (e.g., Violay *et al.*, 2014;
550 Chen *et al.*, 2017) and the development of a similar CPO to the one reported here (Demurtas *et al.*,
551 2019). It has been inferred that the presence of water can promote hydrolytic weakening and
552 influence dislocation glide and climb in calcite (Liu *et al.*, 2002). We speculate that the above-
553 mentioned examples of water influencing crystal-plastic deformation may also have an influence
554 on the activity of specific glide systems and its activation temperature in crustal carbonate faults.
555 The addition of water could explain why De Bresser and Spiers (1997) consider the (c)<a> slip
556 system to be of minor importance in their experiments, which are performed dry and at low strain
557 rates. The potential influence of water on crystal-plastic deformation suggests that the proposed
558 temperature range for the activation of (c)<a> (600–800 °C) and *r*-slip (300–800 °C) may be
559 different or lower in other situations and may explain the absence of a temperature gradient in
560 Figure 2 and 3: essentially no temperature gradient was produced. In such a case, the syn-
561 deformational temperature would evolve along the water-vapour transition as suggested by Chen
562 *et al.* (2017).

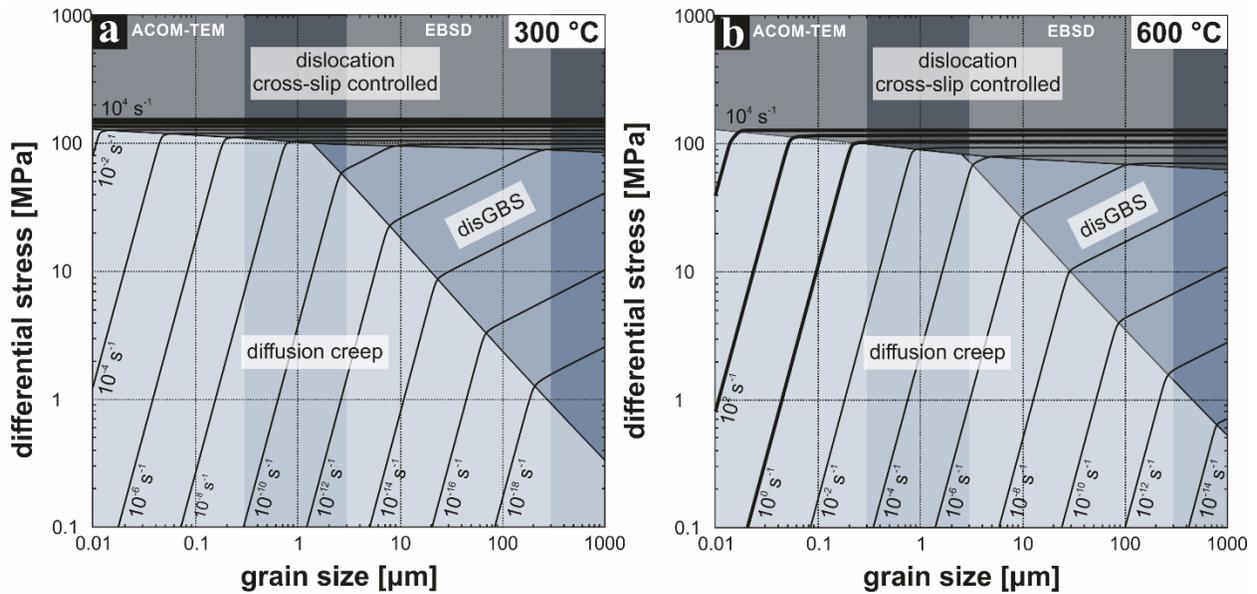
563 The development of CPOs has been reported in natural carbonate faults before. For
564 example, Smith *et al.* (2013) and Kim *et al.* (2018) report a similar CPO and Kim *et al.* (2018)
565 speculate about the contribution of crystal plasticity during deformation. Our subgrain
566 misorientation analysis matches the inverse pole figures presented by displaying a rotational
567 maximum around [0001] close to the slip surface (Kim *et al.*, 2018). The authors report that the
568 intensity of the maximum weakens over 10 cm away from the slip surface. This may indicate that
569 temperature is not the main governing factor for the activation of the (c)<a> glide system because
570 temperature diffusion would reach background values after about 2 mm (Demurtas *et al.*, 2019).
571 In contrast to the fault rocks of Kim *et al.*, (2018), our analyses do not show a pronounced region

572 of plastic deformation. In addition, high dislocation densities are reported from numerous studies
573 of natural faults, e.g. (Colletini *et al.*, 2014) who also shows free dislocations, as well as
574 nanometric, dislocation-free subgrains comparable to our observations in Figure 5.

575 **5.4 Deformation mechanism maps**

576 In the following, we compare our microstructural observations and interpretations of
577 deformation mechanisms with theoretical considerations. We constructed deformation-mechanism
578 maps (DMMs) (Fig. 8) (Ashby, 1972) for both the approximate ambient temperature conditions of
579 300 °C during the inter-seismic period and onset of slip at a depth of 3–5 km and the potential
580 high-temperature conditions of 600 °C attained by seismic shear heating, constrained by the
581 observed CPO and sp² hybridisation of partly-hybridised amorphous carbon (Ohl *et al.*, 2020). The
582 general parameters utilised are A as a material-dependent factor, n as the stress exponent, p as the
583 grain-size exponent, B as a temperature-dependent constant and Q as the activation energy. For
584 the flow laws in Figure 8 we utilised the values $A = 10^{7.63}$, $n = 1.1$, $p = 3.3$, $Q = 200 \text{ kJ mol}^{-1}$ for
585 diffusion creep (Herwegh *et al.*, 2003); $A = 10^{4.93}$, $n = 1.67$, $p = 1.87$, $Q = 190 \text{ kJ mol}^{-1}$ for disGBS
586 (Walker *et al.*, 1990) and for cross-slip-controlled plasticity we used a power law approximation
587 with $A = 10^{16.65}$, $B = 2.431$ and $Q = 584 \text{ kJ mol}^{-1}$ according to Verberne *et al.*, (2015), based on the
588 initial work by De Bresser (2002). As a first approximation, we only consider flow laws for
589 materials with grain sizes on the order of 10^{-8} – 10^{-3} m, comparable to the grain sizes of our faults.
590 We also investigated a flow law derived for water-assisted grain-boundary diffusion described by
591 Verberne *et al.* (2019) and found that it produced the same slope of strain rate contours but
592 predicted lower strain rates than the flow law by Herwegh *et al.*, (2003). Hence, we include the
593 flow law by Herwegh *et al.*, (2003) because it is more reasonable for a high-strain rate

594 environment. Future investigations will also need to determine the impact of flaw laws explicitly
 595 derived for nanogranular materials (Mohamed, 2011).



596
 597 *Figure 8: Deformation-mechanism maps for calcite at (a and c) 600 °C and (e and d) 300 °C. Light-shaded areas*
 598 *indicate the grain-size ranges from crystal orientation mapping by ACOM-TEM and EBSD. a and b: Deformation*
 599 *mechanism maps with three domains: diffusion creep (Herwegh et al., 2003), disGBS (Walker et al., 1990) and cross-*
 600 *slip controlled dislocation glide (De Bresser, 2002). Bold lines represent relevant strain rates.*
 601

602 Figure 8 displays DMMs calculated for temperatures of 300 °C and 600 °C representing
 603 the onset of seismic slip and potential peak deformation conditions, respectively. The difference
 604 in temperature has little influence on the position of the field boundaries but has a significant
 605 impact on the predicted strain rates. The constraints on the grain sizes in this study are good, but
 606 we lack reliable estimates of the stresses. At lower stresses, ≈ 100 MPa, more typical of shallow
 607 faults (e.g., Behr and Platt, 2014) the material is predicted to deform by diffusion creep and/or
 608 disGBS, depending on grain size. Close to a field boundary, dislocation activity may contribute to
 609 the total strain even within the diffusion creep field. Figure 8a suggests that at strain rates of > 1 s⁻¹
 610 and a temperature of 300 °C, approximating the onset of seismic slip, calcite would deform by
 611 cross-slip controlled dislocation glide. Figure 8b indicates that at 600 °C diffusion creep following
 612 the flow law of (Herwegh et al., 2003) can accommodate a strain rate of > 1 s⁻¹, in material with

613 grain sizes of < 100 nm at stresses < 10 MPa. At 600 °C, coseismic strain rates ($1-10^4 \text{ s}^{-1}$) can be
614 accommodated in the PSS by either diffusion creep or plasticity depending on the differential
615 stress. In general, the DMMs predict that seismic strain rates could be accommodated by cross-
616 slip-controlled dislocation glide at stresses >100 MPa.

617 We have shown that crystal-plasticity played a role during the deformation of fault rocks
618 within the vicinity of principal slip surfaces. Although the DMMs in Figure 8 predict the operation
619 of deformation mechanisms known to not produce a strong CPO, our micro-, and nanostructural
620 observations indicate the activation of several slip systems resulting in CPO development. Future
621 studies need to further evaluate the competition between crystal plasticity and GBS processes
622 during the seismic cycle. Advances may be made by combining microstructural observations and
623 DMMs as we have, and by considering dynamic coseismic changes of different deformation
624 mechanisms.

625

626 **5.5 Rheological considerations**

627 **5.5.1 Piezometric equilibrium and dynamic recrystallisation**

628 The analysis above indicates that crystal plasticity and recrystallisation are feasible even
629 under upper-crustal conditions in the brittle regime. Nevertheless, crystal plasticity and GBS
630 processes will be cooperating mechanisms during fault rock deformation. To further decipher the
631 physical nature behind co-seismic deformation processes, Pozzi *et al.*, (2019) proposed the
632 establishment of a piezometric equilibrium during dynamic recrystallisation between GSI and GSS
633 deformation mechanisms. The authors propose that this equilibrium promotes rheological
634 weakening during seismic slip due to cycles of grain-size reduction and thermally driven grain
635 growth. We can further assess the piezometric relationship for recrystallised calcite grains with the
636 relation proposed by Platt and De Bresser (2017):

637
$$D = K \sigma^{-p} \quad (3)$$

638 where D is the recrystallised grain size in μm , $K = 1243$, σ is the differential stress in MPa, and p
639 $= 1.09$. For the cataclasite region with grain sizes of approximately 5–2 μm , Eqn. 3 predicts
640 differential stresses in the range 158–365 MPa. This range of differential stress would plot inside
641 the field of dislocation cross-slip controlled deformation in Figure 8, regardless of grain size, and
642 suggests that crystal plasticity was the major contributor to accommodate strain. For the foam
643 nanostructure (Fig. 6) with grain sizes of approximately 200–20 nm, Eqn. 3 predicts differential
644 stresses in the range 3–24 GPa. While the differential stresses for the cataclasite are plausible on a
645 fault plane, the potential differential stresses estimated for the foam nanostructure are implausibly
646 high and demonstrate that the piezometric relationship of Platt and De Bresser (2017), which was
647 calibrated for much coarser grain sizes, is not applicable in this context. Either the piezometric
648 relationship has a different slope at these finer grain sizes or the nanograins formed by mechanisms
649 other than dynamic recrystallisation. In such a case, static recrystallisation may be able to reach
650 such small grain sizes as a formation mechanism for nanograins and would not reflect differential
651 stresses during deformation.

652 **5.5.2 Post-seismic annealing and fault rock strength**

653 Our observations of the grain-boundary morphology within the Arkitsa nanostructure (Fig.
654 4c) suggest that post-seismic annealing occurred via static recrystallization and grain growth
655 through grain-boundary migration. We define two foam nanostructures, old and new, depending
656 on the overprinting relationship. The older foam nanostructure lies at a greater distance from the
657 PSS (Fig. 4c, white circle), while the new foam nanostructure borders the PSS (Fig. 4c, black
658 circles). We interpret apparent traces of discontinuities that displace grains (Fig. 4c) as fracture
659 planes originating from the PSS. These fractures cross-cut grains of the interlocked nanostructure

660 overprinting the old foam structure (Fig. 4c). Larger grains within the old foam nanostructure (Fig.
661 4c, white circle) are truncated by fractures that cannot be traced back to the PSS but terminate
662 within the new foam nanostructure (Fig. 4c, black circle), instead. The resulting cross-cutting
663 relationships suggest fault reactivation after static recrystallisation. Angular relations indicate that
664 the fractures are Riedel shears (Verberne *et al.*, 2013) and suggest that slip along the PSS may
665 have also taken place during an advanced stage of nanostructural evolution.

666 To assess the influence of grain size on the strength of the PSZ, we calculate the required
667 minimum shear stress, σ_s , to fracture a grain of size d [m] using a modified Hall-Petch equation
668 (Sammis and Ben-Zion, 2008):

$$669 \quad \sigma_s = Y/2 = \frac{2 C K_{Ic}}{\sqrt{d}} \quad (4)$$

670 where $C = \sqrt{\frac{2}{3}}$ and $K_{Ic} = 0.39 \text{ MPa } \sqrt{m}$ (calcite, Broz *et al.*, 2006). For grain sizes of
671 approximately 5–2 μm , Eqn. 4 predicts minimum shear stresses in the range 285–450 MPa. For
672 the median grain size of 21 nm from ACOM-TEM, Eqn. 4 predicts a minimum shear stress of 4.4
673 GPa. Given the spread of the grain-size distribution, we also determine σ_s for a grain size of 200
674 nm (Figure 6a and b) and obtain 1.4 GPa. Based on these calculations, it is evident that with
675 decreasing grain size, slip localization onto the PSS increases because the required shear stress to
676 fracture grains increases. A potential explanation is the decreasing distance between dislocation
677 pinning points leading to a grain-size dependant increase in yield stress with decreasing grain
678 diameter (Kato *et al.*, 2008). The modified Hall-Petch equation, which we note is derived from
679 fitting empirical data from Al_2O_3 spheres and uses an empirically derived value for K_{Ic} from
680 microindentation, should be applied with caution. Alternatively, either local grain-scale stresses
681 may be higher than the overall average stress state of the fault during slip or fractures develop
682 preferentially along zones of weakness, such as cleavage and twin planes.

683 The localisation of slip can be observed over six orders of magnitude (μm – m) and suggests
684 a repeated toughening of the microstructure by grain-boundary strengthening. Our microstructural
685 observations coupled to DMM predictions suggest that at small grain sizes diffusion creep and
686 dislocation creep were active. Deformation by GBS would result in stretching and elongation of
687 the host-rock clasts (Figure 2a and 3a) but the initial shape of the fragments is preserved despite
688 showing an internal, polygonal structure expected to promote GBS. This example is further
689 illustrated by another clast in a transition stage consisting half of a fine-grained microstructure and
690 half of a single crystal (Fig. 2a, white lasso). These examples show that the internal structure is
691 not diagnostic for GBS. Grain-size reduction by deformation and annealing suggests that with
692 evolving localisation, the fault plane becomes progressively stronger with every annealing step.
693 This proposition supports the existence of a grain-boundary strengthening effect within the fault
694 rock volume. Figure 1d shows the presence of a secondary slip surface which develops inside the
695 Schinos cataclasite and we propose that its formation was the first microscale evidence for the
696 locking of the fault rock volume immediately below the PSS. This interpretation is consistent with
697 photographs of the fault exposures (Fig. 1b and d) that show various late-stage fault planes which
698 crosscut inside the wider fault damage zone. Multiple slip surfaces like those typical in any fault
699 zone may be the macroscopic expression of a repeated grain-boundary strengthening effect.
700 Ultimately, the grain size along the fault plane may reach a critical limit, prompting the fault plane
701 to jump and localise elsewhere inside the damage zone leading to the formation of multiple slip
702 surfaces.

703 6 Conclusion

704 The subgrain misorientations and the matching crystallographic preferred orientations
705 across different scales indicate that crystal plasticity played a role during fault rock formation in

706 the Arkitsa and Schinos fault. Although the precise nature of slip systems at sub-seismic velocities
707 are unknown, our results suggest that the slip systems inferred from subgrain misorientation
708 analysis potentially indicate high temperatures during co-seismic deformation or the influence of
709 water. Nevertheless, future studies need to further evaluate the applicability of slip-system
710 analyses as paleoseismicity indicators, especially comparing dry and wet deformation. Plastic
711 straining and tempering, described as cold working and annealing, offers an alternative mechanism
712 to produce a cohesive nanogranular material. Paleopiezometric estimations based on grain sizes
713 immediately below the slip surface suggest that either dynamic recrystallization did not take place
714 or at least did not follow the piezometer calibrated by low-strain rate experiments. The cyclic
715 repetition of plastic strain, annealing and static recrystallization via grain-boundary migration
716 produces a grain-boundary strengthening effect until the grain size reaches a critical minimum.
717 This strengthening effect forces the fault plane to relocate inside the fault damage zone, resetting
718 the deformation cycle.

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726 Data availability

727 All datasets found in this manuscript will be made available open access through the European
728 Plate Observing System at <https://public.yoda.uu.nl/geo/UU01/A77O7X.html>.

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