

Microstructures, mineral chemistry and geochronology of white micas along a retrograde evolution: An example from the Aar massif (Central Alps, Switzerland)

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Abstract

1 This study investigates gneissic tectonites of different mechanical origins, and aims
2 at unravelling the link between microstructures, white mica chemistry and K-Ar data.
3 The highest temperature mylonitic deformation, which is investigated, is dominated
4 by dislocation creep in monomineralic quartz, and viscous granular flow in
5 polymineralic domains. These mylonites show homogenous mineral chemistry,
6 resulting in robust and consistent K-Ar age data. The lowest temperature deformation
7 investigated, is that of a retrograde brittle deformation, producing cohesionless fault
8 gouges. Within the cohesionless fault gouges, frictional granular flow is considered
9 as the main deformation mechanism. The K-bearing phases inside the fault gouges
10 record no chemical resetting during this brittle overprint, but contain mechanically
11 fragmented K-bearing micas and significant amounts of newly produced smectite.
12 This yields K-Ar ages inside of a gouge that are in the range of adjacent mylonite
13 ages, and may not represent the timing of gouge formation. In an effort to understand
14 these uncertainties in K-Ar ages, detailed observations of the microstructural context
15 of K-bearing white mica were made. Microstructural criteria in combination with
16 quantitative element mapping allowed for the discrimination of five important mica-
17 related processes: (1) fracturing/recrystallization of white mica; (2) (re)-precipitation
18 of new fine-grained white mica inside the deforming rock matrix; (3) pseudomorphic
19 replacement of precursor minerals by white mica and chlorite; (4) pseudomorphic
20 replacement of pre-existing white mica without grain size reduction; and (5) remnant
21 grains. The combination of these different processes led to a high variability in
22 mineral chemistry and isotope data for tectonites, which relates directly to variations
23 in the size of the equilibrium volumes. Despite the samples being highly deformed in
24 the presence of fluids, inherited sheet silicates may survive. If inherited sheet
25 silicates are present they contribute to an increase in both the chemical
26 heterogeneity and the apparent K-Ar ages of a sample.

28
29
30 **Keywords:** *white mica, microstructures, fault gouge, mylonite, K-Ar dating*

31

32 **1 Introduction**

33 Detailed knowledge of chemical and isotopic equilibrium is essential for the
34 reconstruction of temperature-time paths. In this context, the use of chronometers
35 and thermometers depends on the identification of equilibrium volumes and their
36 correlation to specific geological events. The identification of such equilibration
37 volumes is related to the system of interest (e.g., mineral-chemistry, isotopic data,
38 fluid/rock-equilibrium, etc.) and in many cases local bulk and mineral compositions
39 are used to model such equilibria (e.g., Vidal et al. 2006; Agard et al. 2010; Lanari et
40 al., 2012; Lanari and Engi, 2017). In general, larger equilibrium volumes are reported
41 at higher temperatures, in the presence of fluids and in highly deformed rocks (e.g.,
42 Wintsch, 1985; Carlson, 2010; Hobbs et al., 2010). These parameters are not
43 independent, because many re-equilibration processes (i.e., deformation, inter- and
44 intragranular diffusion, pseudomorphic replacement by dissolution-precipitation) are
45 closely linked to temperature and fluid availability (Airaghi et al. 2017). In low-
46 temperature deformation regimes, a high partitioning occurs between brittle quartz-
47 feldspars and viscous deforming sheet silicates. Therefore, the identification of
48 equilibrium volumes is important for pressure-temperature-time estimates. This is
49 well investigated for higher-grade metamorphic conditions using characteristic
50 metamorphic minerals. This study focuses on white micas, which are ubiquitous
51 minerals present over a large temperature interval from amphibolite facies down to
52 very-low-grade conditions (Dubacq et al. 2010 and references therein). They are also
53 of special importance as they strongly influence the rheology of fault rocks (e.g.,
54 Faulkner et al., 2010).

55 Knowledge of equilibration volumes requires precise data on mineral-chemistry in
56 order to identify iso-chemical volumes and to reconstruct physio-chemical conditions
57 (e.g., McAleer et al., 2017; Scheffer et al., 2016; Cantarero et al., 2014; Lanari et al.,
58 2012). In this study, we limit ourselves to the description of sheet-silicates. This
59 mineral group is important as a thermo-barometer (e.g., Vidal and Parra, 2000; Parra
60 et al., 2002; Dubacq et al., 2010) and is useful for dating by the Ar-system (e.g.,
61 Reichenbach and Rich, 1969; Kelley, 1988; Villa, 1998; van der Pluijm et al., 2001;
62 Mulch et al., 2002; Rolland et al., 2009; Zwingmann et al., 2010; Sanchez et al.,
63 2011; Derkowski et al., 2014; Lanari et al. 2014b; Kellett et al., 2016; Viola et al.
64 2016; Mancktelow et al., 2016). The link between microstructures, chemical and
65 isotopic data provides insights in potential equilibrium volumes.

66 For this purpose, the well-investigated Aar-massif (Central Alps, Switzerland; e.g.,
67 Wehrens et al., 2016, 2017; Rolland et al. 2009; Challandes et al., 2008; Kammer,
68 1989) was utilized as a natural laboratory to investigate a link between
69 microstructures, chemical and isotopic data and equilibrium volumes. The Aar-massif
70 has a large distribution of strain and within the exhumed mid-crustal basement there
71 are preserved different tectonites (mylonites, cataclasites and fault gouges). A
72 complementary workflow of, quantitative element mapping, grain size sensitive
73 isotope analysis and SEM-based microstructural investigations were applied to a
74 suite of tectonites samples from upper greenschist-facies mylonites to cohesionless
75 fault gouges. In addition, the results from equilibrium assumptions were compared
76 with independent geological information. Furthermore, the use of structural
77 overprinting relationships provided an independent data set, allowing for the
78 reconstruction of a relative temperature-deformation (T-D) path.
79

80 **2 Geological setting and structural inventory**

81 The Aar massif is one of the largest external massifs of the Alps. It is mainly
82 composed of pre-Variscan polymetamorphic gneisses, which were intruded by post-
83 Variscan granitoids (Labhart, 1977; Abrecht, 1994; Schaltegger, 1994). At the
84 Northern boundary these basement rocks are in contact with autochthonous cover
85 consisting of a Mesozoic sedimentary sequence. The valley “Haslital” in the center of
86 the Aar massif transects through all these different units (Figs. 1 and 2).
87 Multiple stages of deformation, from Ordovician to Alpine, affected the pre-Variscan
88 basement rocks (Stalder, 1964; Steck 1968; Schaltegger, 1993; Schaltegger et al.,
89 2003). Within the post-Variscan intrusives only Alpine deformation has been
90 detected. An Alpine deformation and metamorphic gradient was proposed from north
91 to south (e.g., Frey et al., 1980; Choukroune and Gapais, 1983; Bambauer et al.,
92 2005; Wehrens et al., 2017). The north is characterized by lowest greenschist facies
93 metamorphism, whereas in the south upper greenschist facies conditions prevailed
94 (Bambauer et al., 2005; Bousquet et al., 2012; Challandes et al., 2008; Frey and
95 Ferreiro Mählmann, 1999; Goncalves et al., 2012). The Alpine deformation in the
96 Haslital section of the Aar-massif can be subdivided into four major deformation
97 stages (Figs. 1 and 2; Wehrens et al., 2017): (1) Handegg-, (2) Oberaar-, (3)
98 Pfaffenchoopf-, and (4) Gadenen-deformation. The Handegg-deformation is present
99 over the complete section of the Aar massif and is characterized by steep-reverse
100 faulting (Fig. 2; Herwegh et al. 2017; Wehrens et al., 2016, 2017; Rolland et al.,
101 2009; Kammer, 1989; Steck, 1968). These are dominant SW-NE striking steep

102 reverse/normal shear zones and faults with down-dip lineations. The Handegg
103 structures can be well characterized in the post-Variscan plutons. These shear zones
104 show subgrain-rotation recrystallisation in quartz and newly formed white mica in the
105 intermediate and high temperature range (Wehrens et al., 2017; Rolland et al., 2009;
106 Bambauer et al., 2009). The Handegg shear zones in the central and southern part of
107 the massif cover temperatures in the stability field of biotite, but are also active to
108 lower temperatures. In the southern area, the Handegg deformation is overprinted by
109 the Oberaar deformation. This deformation is characterized by ductile strike-slip
110 deformation (e.g., Rolland et al., 2009). In addition, the Pfaffenchof deformation
111 crosscut the Handegg shear zones in the northern Aar-massif (Fig. 2). The
112 Pfaffenchof structures are characterized by more moderately south dipping thrust
113 planes (Fig. 2; Herwegh et al., 2014, 2017; Kammer, 1989; Labhart, 1966). The
114 Pfaffenchof deformation affects basement and Mesozoic sediments, which result in
115 local carbonate mylonites that show dynamic recrystallization of calcite. In the
116 basement, the shear zones and schistosity are characterized by the frictional-viscous
117 transition. In most examples quartz deformed in a brittle manner, but in some cases
118 local bulging recrystallization can be inferred. The main processes are re/neo-
119 crystallization of white mica and chlorite. The Gaden-structures are represented by
120 brittle faults, gouges and a weak brittle schistosity. They crosscut the Handegg- and
121 Pfaffenchof-deformation, and in the northern area the fault gouges have been
122 investigated in underground facilities. In the area further south, brittle deformation is
123 also frequent, but the crosscutting relationship to the older deformation is not clear.
124 Some of the brittle structures may also represent continuous deformation of the
125 Handegg- and/or Oberaar deformation.

126 It is the P-T variations with time, across of the northern and southern regions of the
127 Haslital area, that are key to understanding recrystallization processes and potential
128 Ar-loss in mica. The southern Aar-massif is well investigated and peak P-T conditions
129 of ~450°C and 6kbar are established there (Goncalves et al., 2012). This is
130 consistent with δO^{18} thermometry (Fourcade et al., 1989) and observations of the
131 transition from microcline to sanidine (Bambauer et al., 2005). The central part of the
132 Aar-massif represents the transition between maximal temperatures of 450°C to the
133 very low-grade conditions at the northern border (see below). This transition is
134 characterized by the “biotite-in” and the “stilpnomelan-out” isogrades (Steck and
135 Burri, 1971; Niggli and Niggli, 1965; Jäger et al., 1961). From these conditions in the
136 center, a steep gradient developed toward the northern end of the massif (Fig. 2).
137 The northern rim is related to a lower maximal temperature of 250-300 °C and a

138 pressure in the range of 2-3 kbar (Frey et al., 1980). The samples selected for this
139 investigation are from south of the “biotite-in” isograd (Fig. 1).
140

141 **3 Sample location and description**

142 Samples from mylonites to fault gouges are located along the above described
143 temperature gradient (Figs. 1, 2 and Table 1). The tectonites can be subdivided into
144 three major groups: (1) intermediate temperature granitoid mylonites, which are
145 dominated by dislocation creep in quartz layers and viscous granular flow in
146 polymineralic domains; (2) gneissic low temperature mylonites, in which there is the
147 brittle deformation of quartz and feldspar and viscous deformation of sheet silicates;
148 and (3) cohesionless fault gouges which are the product of brittle deformation. Along
149 the deformation-temperature gradients, mineralogy, grain size and deformation
150 mechanisms changed (Figs. 2 and 3).

151 The dominant deformation mechanisms are also connected to different deformation
152 stages (see section 2; Fig. 2). The mylonite samples are located in the Central Aar-
153 granite (Fig. 1). Sample GRS05 is related to Handegg deformation and involves
154 dislocation creep in quartz layers and viscous granular flow in a compositionally
155 mixed matrix. At the sample-location the shear zone consists of a 50 cm wide
156 mylonite with a strain gradient toward the undeformed Central Aar granite. The shear
157 zone can be traced for several kilometers along strike. Sample GRS06 originates
158 from a nearby strike slip shear zone of several meters width and kilometers length
159 (Table 1). The schistose host rock is cut by intercalations of bands of (i) mylonitic
160 quartz and (ii) quartz-white mica (Fig. 4). These bands inside the host developed due
161 to hydrothermal activity and are interpreted as synkinematic veins. The host-rock has
162 a granitic composition for these mylonites. To some degree a polymineralic fine-
163 grained matrix has developed consisting of recrystallized quartz, mica, and feldspar.
164 There is a preferential alignment of both white mica and biotite. Dynamically
165 recrystallized grain sizes in the pure quartz bands and the schistose granitoid are
166 comparable. In contrast, the white mica-quartz bands consist of newly formed white
167 mica with grain sizes down to 3 μm (Fig. 4). Handpicked large micas originate from
168 the schistose host and have an unknown history. Fine-grained white mica from both
169 layers were selected for K-Ar dating.

170 The low-T mylonites are related to Pfaffenchoepf deformation, which is very localized
171 (Fig. 2). Two low-T mylonites are investigated, which differ slightly in their mineralogy
172 (Figs. 1, 2, 3 and Table 1). In addition, four fault gouges are selected for analysis.

173 These fault gouges are subdivided into two from the Handegg area (overprinting
174 ductile Handegg shear zones) and two from the Innertkirchen area (close to low-T-
175 mylonites; Fig. 2a).

176

177 **4 Methods**

178 **4.1 Sample preparation**

179 The mylonite samples are crushed by Selfrag mineral liberation (Zwingmann et al. in
180 review; Sperner et al., 2014; Giese et al., 2010). Gouge samples consisted of >0.5 kg
181 cohesionless material providing fine clay like material, sand fraction and mineral
182 aggregates. The fine clay material is associated with the gouge formation, and no
183 artificial mineral liberation was required. Both artificially liberated mineral samples
184 and cohesionless gouges samples were then sieved to obtain particles < 500 µm.
185 Grain size fractions of 6-10 µm and 10-20 µm were separated by gravity settling
186 using Stokes law (Atterberg settling tubes). In addition, grain size fractions <0.8, 0.8-
187 2, 2-5, 2-6 µm were obtained using a swing rotor centrifuge. Large grain sizes (>60
188 µm) were handpicked (Table 2).

189

190 **4.2 Microstructures, element mapping, mineral chemistry and** 191 **thermobarometry**

192 Thin sections were cut from larger rock fragments and aggregates; these sections
193 were then studied with the optical light microscope to confirm the host rock
194 composition. Petrographic investigations involved light and electron microscopy.
195 Scanning electron microscopy was performed on a ZEISS EVO50 using backscatter
196 and charge contrast images. Electron microprobe analyses (EMP) were performed
197 using a JEOL superprobe JXA8200 instrument at the Institute of Geological Sciences
198 of the University of Bern. The analysis of sheet silicates was conducted at 15 kV and
199 5-8 nA, using natural and synthetic standards for major elements (SiO₂, Al₂O₃, FeO,
200 MnO, MgO, CaO, Na₂O, K₂O). X-ray element mapping was conducted using
201 wavelength dispersive spectrometers at 15 kV and 100 nA with dwell time ranging
202 between 100 and 200 ms. The compositional maps were standardized using a series
203 of spot analyses measured on the same area. The data were reduced using the
204 program XMapTools 2.2.3 (Lanari et al., 2014a). The primary data (X-ray counts
205 uncorrected for background) were standardized after the classification using the
206 automatic function provided in XMapTools. Structural formulae calculations (on a
207 basis of 11 and 14 atoms of oxygen for K-white mica and chlorite, respectively) were

208 applied to all the pixels based on the atom site repartition models used in the
209 thermodynamic models (see below). The pixel compositions of K-white mica and
210 chlorite are reported as element maps (in atoms per formulas unit) to highlight the
211 relationships between the compositional zoning and the microstructures (see
212 Scheffer et al. 2016).

213 The chemical subdivisions into distinct groups are based on differences in mineral
214 chemistry related to substitutions. The compositional variability of chlorite in the
215 system $\text{SiO}_2\text{-Al}_2\text{O}_3\text{-FeO-MgO}$ is described by three main substitutions (i) FM: $\text{Fe}^{2+} =$
216 Mg^{2+} , (ii) Tschermak: $\text{Si} + (\text{Mg}^{2+}, \text{Fe}^{2+}) = 2\text{Al}$ and (iii) di-trioctahedral: $(\text{Mg}^{2+}, \text{Fe}^{2+})_3 = 2\text{Al}$
217 (Cathelineau and Nieva, 1985; Lanari et al., 2014c), whereas additional substitutions
218 take place in the ferric system (Trincal and Lanari, 2016; Vidal et al. 2016). The
219 compositional variability of K-white mica is modeled in the system $\text{SiO}_2\text{-Al}_2\text{O}_3\text{-FeO-}$
220 $\text{MgO-Na}_2\text{O-K}_2\text{O}$, based on the definitions of Dubacq et al. (2010), using the following
221 end-members: muscovite ($\text{Si}_3\text{Al}_3\text{K}_1\text{O}_{10}(\text{OH})_2$); Fe/Mg-celadonite ($\text{Si}_4\text{Al}_1(\text{Fe/Mg})_1$
222 $\text{K}_1\text{O}_{10}(\text{OH})_2$); phlogopite ($\text{Si}_3\text{Al}_3\text{Mg}_1\text{K}_1\text{O}_{10}(\text{OH})_2$); pyrophyllite ($\text{Si}_4\text{Al}_2\text{O}_{10}(\text{OH})_2$);
223 and paragonite ($\text{Si}_3\text{Al}_3\text{Na}_1\text{O}_{10}(\text{OH})_2$). The main exchanges led to vacancies in the
224 A-site due to K-free components and to changes in Al(tot) related to celadonite and
225 pyrophyllite exchange reactions (e.g., Parra et al., 2001; Dubacq et al., 2010). This
226 led to the main exchange vectors: paragonite-muscovite exchange $[\text{KNa}_{-1}]$,
227 tschermak: $\text{Si} + (\text{Mg}^{2+}, \text{Fe}^{2+}) = 2\text{Al}$ and pyrophyllite-vector $[(\text{K,Na})_{-1}\text{O}_{-1}]$. P-T estimates
228 of high variance assemblages are generally achieved with the technique of multi-
229 equilibrium (Vidal and Parra, 2000; Vidal et al. 2006). Several calibrations have been
230 developed and the most recent of them are used in this study: Chlorite + Quartz +
231 Water (Lanari et al. 2014) and Phengite + Quartz + Water (Dubacq et al. 2011). A
232 comprehensive description of this technique based on the progressive hydration of
233 the vacancies along a P-T divariant equilibrium curve is provided together with
234 application examples in Lanari et al. (2012). Computations were achieved using the
235 in-house program ChlMicaEqui (Lanari, 2012).

236

237 **4.3 Clay minerals**

238 Oriented sample separates were achieved by settling of the grain size fractions onto
239 silica wafers. Diffractograms were made for all grain size fraction using XPERT PRO
240 PANalytical diffractometer with $\text{CuK}\alpha$ radiation and 40 kV/40 mA. Samples were
241 scanned over the range $2\theta = 4 - 60^\circ$ at $0.0167^\circ 2\theta/\text{s}$. Additional measurements of the
242 smallest grain size fraction for air dried, glycolated and heat treated slides were
243 performed in order to distinguish between smectite, kaolinite and chlorite (Moore and

244 Reynolds, 1989) for which sample separates were sedimented onto glass slides. The
245 diffractograms were obtained using a Philips PW 1710 diffractometer with samples
246 scanned over the range of $2\theta = 2 - 40^\circ$ at $0.02^\circ 2\theta/s$ (Table 2). In selected gouges,
247 the diffraction pattern was Rietveld-refined with TOPAS-Academic V6 (Coelho,
248 2012). For refinement, March-Dollase preferred orientation parameters of both illite
249 structures (1M and 2M) were constrained to the same value (0.565(3)).

250

251 **4.4 K-Ar dating**

252 A standard method, described elsewhere (Dalrymple and Lanphere, 1969), was used
253 for K-Ar dating. The potassium content was determined by atomic absorption.
254 Duplicate K determination on several samples and standards resulted in a pooled
255 error better than 2.0 %. A procedure comparable to the one used by (Bonhomme et
256 al., 1975) was used for Argon isotopic determinations. Several hours of pre-heating
257 under vacuum at 80°C served to reduce the amount of Argon adsorbed onto the
258 mineral surface during sample preparation.

259 Argon was extracted from the separated mineral fractions by fusing samples within a
260 vacuum line serviced by an online ^{38}Ar spike pipette. An on-line VG3600 mass-
261 spectrometer was used to measure the spiked Ar via Faraday cup. The ^{38}Ar spike
262 was calibrated against biotite GA1550 (McDougall and Harrison, 1999). After fusion,
263 the released gases underwent a two-stage purification procedure via CuO and Ti
264 getters. A systematic determination of blanks for the extraction line and mass
265 spectrometer was carried out. The mass discrimination factor was determined by
266 airshots. Argon analyses required around 20 mg of sample material. During the
267 course of this study, the international standards GLO (Odin et al., 1982) and HD-B1
268 (Hess and Lippolt, 1994) were measured several times ($n=5$). The error for Ar
269 analyses is below 1.00 % (Table 2) and the $^{40}\text{Ar}/^{36}\text{Ar}$ value for airshots averaged
270 295.05 ± 0.35 ($n = 5$, Table 3). ^{40}K abundance and decay constants suggested by
271 Steiger and Jager (1977) allowed for K-Ar age calculation. Errors are listed within 2
272 sigma.

273

274 **5 Results**

275 **5.1 Mylonites**

276 Sample GRS05 contains fine-grained deformed layers. The latter consist of biotite,
277 white mica, quartz, epidote and minor amounts of feldspar (Figs. 3, 4; Table 4). The

278 fine-grained biotite and white mica (4-50 μm) are related to syn-deformational
279 (re)crystallization. The mylonite contains large mica flakes (100-400 μm), with
280 unknown pre-deformation history and newly formed grains related to mylonitization
281 (down to 3 μm).

282 The EMP-analyses of white mica from sample GRS06 show one composition (Fig. 5;
283 Table 5). This composition is consistent with the white mica in mylonitic bands as
284 well as in the surrounded schistose material. Sample GRS05 shows only limited
285 spread in white mica composition with an average SiO_2 content of 47 wt%, this is
286 notably independent of grain size. The Na_2O content is below 0.1 wt%. For
287 comparison the white mica from sample KAW2214 is measured (Figs. 1 and 5; Table
288 1). The biotite bearing mylonites contain white mica with low Na content, but a slight
289 temperature effect from north to south can be inferred (Fig. 5). These data overlap
290 with white mica compositions from the literature further south (Challandes et al.,
291 2009; Goncalves et al., 2012). The ductile deformed samples show a consistent K-Ar
292 age range, which is spread between 11.8 ± 0.2 and 13.6 ± 0.3 Ma (Table 6; Fig. 6).
293 The different grain size fractions give similar ages inside one sample and no
294 relationship between grain size and age can be observed.

295

296 **5.2 Low-temperature mylonites**

297 5.2.1 Microstructures

298 Two investigated samples deformed at the frictional-viscous transition (see Table 1).
299 Deformation conditions are characterized by brittle deformation of feldspar and
300 quartz. Quartz grains occasionally show bulging recrystallization. The overall
301 deformation has been classified as ductile, which is caused in the deformation-
302 behavior of the sheet silicates. However, also large white micas and chlorite (100 -
303 400 μm) are observed (Fig. 7b, c). They show bending into strain localization planes
304 (Fig. 7). These planes are characterized by both fine-grained phase mixtures and
305 local chlorite/mica intergrowth. In addition local hydrothermal veins developed, which
306 are related to brittle deformation (see the base of the optical microphotograph in Fig.
307 8a). The micas dominate the deformation. The white micas are designated into the
308 following microstructural groups (Table 7):

309 *mi1*: Several large grains occur as relics in the mylonites. This microstructure is
310 more frequent in sample Gr66 as in sample Ga14-2 (Figs. 7 and 8).

311 *mi2*: This microstructure is characterized by small grains in the direct vicinity of
312 larger mica grains Fig. 7). The process of dynamic recrystallization could be

313 responsible of this microstructure type. However, the single slip-plane of mica
314 makes dynamic recrystallization problematic (e.g., Shea and Kronenberg,
315 1992). Alternatively, fracturing in combination with preexisting stacking faults
316 can produce such a microstructure. This indicates fracturing inside the
317 preexisting grains and their shearing along the (001) plane. The grain size of
318 identified new grains is in the range of 2-5 μm .

319 *mi3*: These micas grew while isolated in the recrystallized quartz or quartz/feldspar
320 matrix by solution/precipitation (Fig. 7). The grain size of this group is
321 relatively constant (2-5 μm ; Table 7). This microstructure is comparable with
322 the microstructure in the intermediate temperature mylonites.

323 *mi4*: Mica pseudomorphs grown at the expense of precursor minerals (Figs. 7 and
324 8). These pseudomorphs are occasionally dominated by white mica, but also
325 chlorite-white mica mixtures occur. In addition, fine-grained alteration of
326 feldspars is observed, producing muscovite and, locally, epidote. The
327 chemical composition of such white micas may have been controlled by the
328 local bulk composition during the reaction that probably differs from the local
329 bulk composition of the matrix (see details in section 5.2.3).

330 *mi5*: This group of mica can only be identified in combination of microstructure and
331 mineral-chemistry data. This process developed patches with variable
332 chemistry (Figs. 7, 8). Such changes of chemistry without obvious change in
333 microstructure requires a process described as pseudomorphic replacement
334 (Putnis, 2002; Airaghi et al. 2017). Such replacement structures can be
335 inferred in single grains as well as in grain aggregates (Fig. 7).

336

337 5.2.2 Mineral chemistry and P/T data

338 The mineral-chemistry of the white mica is divided into the following compositional
339 groups: (A) low Al (high Si) -contents; (B) high Al (low Si) at similar Na contents; and
340 (C) high Al and high Na contents (Fig. 5). The compositional group A is characterized
341 by high Si content (Si \sim 3.3 apfu (=atoms per formula unit)), low Al content (Al \sim 1.5
342 apfu), high Mg content (Mg \sim 0.23 apfu) and low Na content (Fig. 5). This group is
343 systematically associated with newly formed chlorite and is interpreted as newly
344 formed grains. These newly formed white mica indicate temperatures in the range of
345 \sim 300°C (assuming a pressure of 3-4 kbar and $a_{\text{H}_2\text{O}}=1$; Fig. 9). This result is
346 consistent with independent temperature estimates derived from chlorite
347 compositions associated with such white micas (Fig. 9). In sample Ga14-2, rare and
348 isolated calcite grains indicate possible lower water activity (if calcite is reacting at

349 the same time as the white mica). A lower $a_{\text{H}_2\text{O}}$ would slightly decreases the resulting
350 temperature for the white mica by 20-30°C (Fig. 9). The group-C micas are related to
351 alteration of group-A mica and new growth in hydrothermal pockets. These micas are
352 characterized by high, but variable Na-contents. Some of the high Na contents are
353 associated with decreasing interlayer content (increase of the pyrophyllite
354 component, Fig. 10b; see section 6.2).

355

356 5.2.3 Ar-data

357 Six fractions from two samples are measured, which yield apparent K-Ar ages
358 between 19.9 ± 0.4 Ma to 39.8 ± 0.8 Ma (Tables 3 and 6; Fig. 6). The coarse grains
359 are systematically older than the fine fraction (Table 6). However, the absolute values
360 of ages at given grain size differ between the two samples. The fine grain size
361 fraction ($<0.8\mu\text{m}$) of sample Ga14-2 is apparently older than the fraction 2-6 μm in
362 Gr66 (Table 6). However, the selected large grains have apparent ages in the same
363 range (37 and 40 Ma; Table 6; Fig. 6), which is significantly lower than literature data
364 from undeformed samples in the area (compare white mica age of KAW2385 versus
365 Gr66/Ga14-2; Table 1).

366

367 5.3 Fault-gouges

368 Gouges from the southern- and the northern areas are investigated (Figs. 1 and 2;
369 Table 1). Sample He12 from the southern area is collected from a several cm wide
370 fault gouge overprinting a ductile shear zone (Table 1). The symmetric shear zone
371 shows an increase of deformation from weakly deformed host rock over schistose
372 granite to mylonite. Orientation of the fault plane is steeply south dipping. The fault
373 gouge in the centre with a width of several cm consists of fine clay material, where a
374 lineation measured on the clay surface is subhorizontal. This lineation can easily be
375 swiped away by hand and has been established as a slickenside lineation (Doblas,
376 1998). This type of slickenside may develop by the ploughing of harder minerals (i.e.
377 quartz) into the clay matrix during block movements. The gouge is cohesionless and
378 no defragmentation was required prior to clay separation (Table 2). The second
379 sample from the southern area (sample He15; Table 1) is collected from a 10 cm
380 wide fault zone. The shear zone is asymmetric and ranges from weakly schistose
381 granitoid in the south toward mylonite (Fig. 2). The orientation of the fault surface is
382 steeply dipping toward the south. The slickenside lineation on the clay surface is

383 subhorizontal. Also here, collected material consists of fine clay material with a
384 contribution of larger fragments.

385 The investigated fault gouges of the northern area are cross cutting all other
386 structures (Figs. 3 and 4). They are both NE-SW to NEE-SWW striking and steeply
387 dipping toward the south. A lineation plunging down dip was visible on the gouge
388 surface of sample Ik1206, whereas the lineation on the clay surface of Ik1206 is
389 subhorizontal in orientation. These fault gouges are about 10 cm wide. All fault
390 gouges consisted of fine-grained matrix material and fragments. The fragments are
391 relics inside the gouge, which show a composition/microstructure similar to the
392 proximal mylonites. The mylonitic microstructure is preserved in these fragments
393 confirming the overprint of a former a ductile shear zone by the fault gouge. The two
394 northern samples differ in their microstructures from the incorporated clasts. Sample
395 Ik1206 shows in all inspected fragments highly mylonitic structures with fine-grained
396 mica aggregates as described before for the low T-mylonites of the area (Fig. 11).
397 The mineral-chemistry of these micas is comparable with those of adjacent mylonites
398 (Fig. 11). In contrast, blocky quartz and large mica grains characterize sample Ik1202
399 (Fig. 11). The gouge material itself contains smectite, chlorite and illite/white mica
400 and kaolinite (Table 4). The smectite fraction is the highest in the smallest grain size
401 ($< 0.8\mu\text{m}$). Furthermore, the proportion of smectite increases with the decreasing
402 grain size. This indicates that smectite is newly formed in the small grain size fraction
403 (Table 4). This is better evolved in the southern than in the northern samples (Table
404 4). illite, quartz and chlorite/kaolinite were identified in the fine-grained fractions.
405 Rietveld refinements demonstrate the occurrence of both 1M and 2M structures (see
406 section 4.3). XRD-data results point to a ~15% fraction of M1-structures (M2-illite:
407 ~77%, Quartz: ~8%, Kaolinite/Chlorite: ~1%; Fig. 12). As already mentioned for
408 smectite (Table 5), the amount of newly formed low-grade clay minerals is insufficient
409 to date these phases (see below; the potential to unravel the difference in age
410 between the inherited minerals and newly formed minerals, which is short in the
411 investigated examples).

412 The fault gouges of the southern area (sample He12 and He15) show an age spread
413 between 11.3 ± 0.3 - 14.4 ± 0.4 Ma with the exception of the smallest grain size of
414 sample He15 (Fig. 6). The latter data can be excluded, because of the low K-content
415 (Table 6). The fault gouges of the northern area differ between the two sample sites,
416 but in both samples no relationship between grain size and age can be inferred.
417 Sample Ik1202 show ages between 33.0 ± 0.7 and 36.8 ± 0.7 Ma, whereas sample
418 Ik1206 gives results of apparent ages between 18.9 ± 0.4 and 21.1 ± 0.4 Ma (Table

419 6). The different age groups of these two samples correspond well with completely
420 different microstructures of the clasts in these samples (Fig. 11).

421

422 **6 Discussion**

423 The results for mylonites, low-T mylonites and cohesionless fault gouges illustrate
424 changes in the size of equilibrium-volumes, which is related to temperature and
425 deformation conditions. In the following, the different tectonites are first addressed
426 separately and then compared and integrated in a general model (see section 7).

427

428 **6.1 Hydrothermal veins inside mylonites**

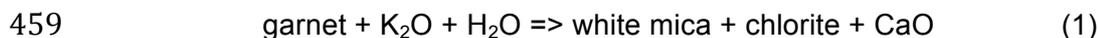
429 Sample GRS06 shows multiple veins consisting of mylonitized pure quartz and white-
430 mica quartz aggregates (Fig. 4). Neocrystallized white mica in the deformed vein with
431 a grain size of 2-6 μm is compared with larger fractions, up to handpicked mineral
432 sizes. The K-Ar ages for the different grain size fractions are, within uncertainty,
433 identical (Fig. 6). In principal, the obtained age could be related to (i) diffusive re-
434 equilibration, (ii) net-transfer reactions and recrystallization, or (iii) inheritance of older
435 deformation / metamorphic phases (e.g., Villa, 1998; Allaz et al., 2011; Villa et al.,
436 2014). Dodson-type cooling ages are based on diffusional length scale and should
437 relate to grain size (Dodson, 1973). This is not the case in the high temperature
438 mylonite samples, and the meaning of the ages as diffusive re-equilibration or
439 "cooling age" (i) can therefore be dismissed (Fig. 6). More recent studies proposed
440 fluid circulation and recrystallization as important processes in closing the isotopic
441 system (Villa and Hanchar, 2013; Allaz et al., 2011; Villa, 1998). Our structural data
442 indicates that the newly formed white mica is the result of recrystallization (section 4).
443 Any inheritance (iii) of an older deformation phase or crystallization would be
444 noticeable, especially in the age of larger grains (Villa et al., 2014), which is again not
445 the case in these samples.

446 The obvious deformational induced recrystallization of the fine fraction and the, within
447 error, similar ages of the large grains indicates contemporaneous deformation and
448 hydrothermal veining (ii). The age of larger handpicked grains may reflect a
449 hydrothermal event with subsequent deformation. This excludes mixed ages for the
450 recrystallized micas in these mylonites. The resulting age in this sample is therefore
451 linked to the hydrothermal activity during shearing of the mylonite.

452

453 **6.2 Mineral reactions, mica petrology and P-T data in low-T mylonites**

454 The low-T mylonites contain different microstructures, some of them are connected
 455 to precursor minerals (e.g., *mi4*: pseudomorphs, *mi5*: pseudomorphic replacement,
 456 Table 7, section 5.2). In these two example microstructures, a net-transfer reaction is
 457 necessary. Two important groups of reactions are inferred from microstructures and
 458 inherited minerals:

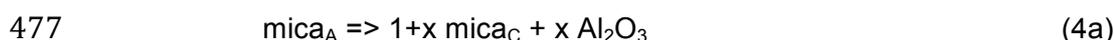


461 Replacement structures and re- or neocrystallization require reaction of the type:

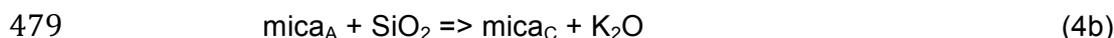


463 In contrast to reaction (1) and (2), reaction (3) does not necessary require the
 464 nucleation of new crystals and can be accommodated by diffusion, dissolution /
 465 reprecipitation and mineral replacement (Putnis, 2002; Putnis and Austrheim, 2013;
 466 Fig. 8). Independent of the inferred reaction, the product compositions are different. It
 467 is important to highlight here that the compositions of the newly formed white micas
 468 define characteristic compositional groups that are observed at the thin section scale
 469 suggesting the achievement of local equilibrium. The local equilibrium is not only
 470 controlled by the local solid phases, but also by the fluid. The occurrences of limited
 471 mica-compositions indicate an important role of the circulating fluid for such local
 472 equilibriums.

473 The combination of microstructures and mineral-chemistry data document mainly a
 474 change from chemical composition of group A to group C. This change in mineral-
 475 chemistry requires an exchange vector of the type $\text{K}_{-1}\text{Al}_{-1}\text{Si}$, which can be written as
 476 an exchange reaction:



478 or:



480 This suggests not only a change in the Si/Al ratio, but also an increase in vacancies
 481 (pyrophyllite component). However, the detected increase in vacancies is
 482 systematically small and the major change is related to K_{-1}Na exchange (Fig. 10).
 483 The change in K/Na ratio in the white mica requires either transport of these
 484 elements in a fluid and/or a reaction involving K/Na minerals (i.e. feldspars).

485 The change in composition between groups A and C includes possible paragonite-,
 486 celedonite- and pyrophyllite exchange vectors (e.g., Livi et al., 1997; Agard et al.,
 487 2001; Dubacq et al., 2010). The K- and Na-data mainly shows a paragonite
 488 exchange (NaK_{-1} ; Fig. 10a). An increased paragonite component in white mica
 489 coexisting with paragonite is related to a temperature increase, which can be derived

490 from the shape of the muscovite-paragonite solvus (e.g., Keller et al. 2005). If
491 paragonite is not present, the absolute Na-concentrations depend on the coexisting
492 phases and fluid, i.e. the composition of the equilibrium volumes (= reactive bulk
493 composition, see Lanari and Engi 2017). Large variations of Na-content and Si/Al
494 ratios occur in sample Gr66, where the micas with higher Na contents (group C)
495 clearly formed along the retrograde path, i.e. at lower temperature than the Na-poor
496 K-white micas.
497

498 **6.3 Relationship between microstructure, mica composition and Ar-data in** 499 **low-T mylonites**

500 The different mica compositions occur at different microstructural positions in the
501 different samples (Fig. 13). For example, the high-Na mica in sample Ga14-2 is
502 observed in the highly deformed domains (Fig. 8f), in the grains of the upper section
503 of Figure 8a, and around the garnet porphyroblast (see Fig. 8a). But this group also
504 occurs as small grains in the matrix (microstructure “mi3”, see Fig. 8), where mineral
505 composition A dominates. In this case, the link between microstructural position and
506 composition is not unique, but again only a limited number of compositional groups
507 can be defined (Fig. 5). For example composition “C” occurs in replacement
508 structures and at locations of recrystallization (Fig. 13). Moreover, the composition of
509 white mica seems to be independent of the local mineral assemblage. The
510 occurrence of the same white mica composition in different microstructures indicates
511 equilibrium with a fluid. It is important to note here that a large part of the rock is not
512 reacting during the formation of the new white mica generation. This process can
513 explain the formation of white mica with the same composition in domains with
514 different unreactive assemblages. The equilibrium volume depends on the type and
515 size of the fluid-mineral exchange volume. In our sample the changes from one to
516 another compositional group suggest change in the P-T-(X) conditions. However, the
517 differing equilibration volumes depend on microstructure and the related equilibration
518 processes between fluid and deforming mica (e.g., Putnis, 2002; Lanari et al., 2014b;
519 Airaghi et al. 2017).

520 Changing P-T conditions along a given P-T-D-t path involve tectonic processes. This
521 prerequisite has been intensively invoked at other locations with the combination of
522 petrological and Ar investigations (McAleer et al. 2017; Angiboust et al., 2014; Lanari
523 et al. 2014b; Sanchez et al., 2011; Augier et al., 2005; Agard et al., 2002; Cosca et
524 al., 1998; Markley et al. 1998). In in-situ laser ablation studies for instance, the
525 microstructure of micas were successfully linked to Ar isotope ages (e.g., Agard et al.

526 2002, Schneider et al., 2013; Mulch et al., 2002). Some studies extracted Ar from
527 carefully selected areas of multiple grains (e.g., Agard et al., 2002), whereas other
528 studies select large single grains (e.g., Mulch et al., 2002). Several issues in such
529 datasets can be overlooked without direct petrological control (see this study, Kellett
530 et al. 2016). From a theoretical point of view, only the newly formed (and isotopically
531 reset) grains should be analyzed and they remain small in the temperature range of
532 interest (e.g., Gueydan et al. 2003). The complexity of white mica major element
533 chemical zoning has been highlighted in several studies and the re-equilibration
534 commonly involves dissolution-precipitation processes instead of new growth
535 (Scheffer et al. 2016; Airaghi et al. 2017). To obtain isotopic analyses of the newly
536 grown crystals only, one can apply analyses for different grain sizes and select the
537 smallest fraction. Therefore, grain size sensitive analysis may be the optimal tool to
538 unravel such processes with K-Ar techniques. This method has been successfully
539 applied to fault gouges (e.g., Zwingmann et al. 2004; Zwingmann, 2014 and
540 references therein). Conventional K-Ar dating was used instead of $^{39}\text{Ar}/^{40}\text{Ar}$ dating to
541 avoid ^{39}Ar recoil artifacts or encapsulation corrections (Zwingmann, 2014).

542 In the investigated example, recrystallization and fracturing of mica produces
543 isolated, ultrafine grains (Figs. 7, 8, 13, 14 and Table 7). The selection of these fine
544 grains may relate to the dominant deformation stage of the sample. However, as
545 seen in the mineral chemistry also relic chemistry can be retained in small grains, if
546 fracturing is an important process (Figs. 7, 8 and Table 7). Vice-versa, large grains
547 can change their chemistry by replacement processes without changing the grain
548 size (Putnis, 2002; Airaghi et al., 2017). This allows perturbation of isotope
549 information in different grain size fractions, which corresponds with an apparent
550 partial resetting of the large grains in comparison to literature gneiss data (Table 1).
551 In addition, relics of fractured grains are responsible in the fine fraction for apparent
552 ages being too old (Fig. 14). Neither the grain size nor the position of certain micas
553 correlates with chemical groups of micas.

554

555 **6.4 Isotope data in gouges**

556 Dating of gouge formation can be successful if K-bearing phases are neoformed
557 (e.g., Zwingmann, 2014; Bense et al., 2014; Viola et al., 2013; Clauer, 2013; Pleuger
558 et al., 2012; Solum et al., 2005; Zwingmann and Mancktelow, 2004; Zwingmann et
559 al., 2004, 2010; van der Pluijm et al., 2001; Torgersen et al., 2015; Mancktelow et al.
560 2015; Viola et al., 2016). In this context, knowledge of the amount of newly formed

561 illite versus inherited detrital muscovite is important to assess the significance of
562 “mixed” ages. In the investigated gouges, the following points can be highlighted:

563 (1) Gouge samples show no systematic trend between grain size and age (Fig.
564 6).

565 (2) Grain size reduction of the mylonite hosting K-bearing phases resulted in clay
566 size particles during the gouge formation (no crushing were applied).

567 (3) Relative high smectite fractions in the fine fractions indicate formation at very-
568 low temperature.

569 (4) Decreasing K-content with increasing smectite content (sample He12)
570 suggests K-loss via low temperature fluids.

571 (5) Owing to overprinting/reactivation of inherited deformation structures, detritus
572 in the gouges is more likely sourced from precursory mylonites.

573 In this view, the gouge hosts similar processes as the multistage samples. However,
574 due to missing newly formed K-phases during gouge formation, no age information
575 for the brittle deformation can be gained. The K-bearing relics in the gouges can be
576 seen as chemically unaltered fragmented minerals originating from the precursor
577 mylonites. The K-Ar ages of the gouges are therefore similar to the surrounded
578 mylonites, which is the 12-14 Ma isotope ages in the southern area (sample He12
579 and He15) and the apparent 19 Ma data in the Innertkirchen area (Table 6). These
580 ages are most likely inherited from surrounded material, indicating a mechanical
581 grain size reduction processes during brittle deformation (fracturing of sheet
582 silicates). Furthermore, the clay sometimes used mineralogy (e.g., M1-illite; Solum et
583 al. 2005, 2010, Fitz-Diaz and van der Pluijm 2013) does not correlate with apparent
584 ages gained from K-Ar dating in our example (Table 5 and 6). Our results, as data in
585 other studies, indicate that K-Ar dating should be considered in combination with
586 microstructural and chemical analysis (e.g., Mancktelow et al. 2015).

587

588 **6.5 Geological implications for the Aar-massif**

589 The presented Ar-data in the mylonites record ductile deformation in the central part
590 of the Aar-massif (see Tables 2 and 6). The obtained new data are similar and
591 overlap with results further south (Fig. 15; Rolland et al., 2009; Challandes et al.,
592 2008; Rossi and Rolland 2014; Table 1). Considering the uncertainty, the
593 deformation age of the investigated Handegg- and the strike-slip shear zone in the
594 center of the massif are overlapping. This model fits well with the suggested
595 transition of the two phases at ~12 Ma as documented by synkinematic open cleft
596 monazite in the Oberaar area further south (Bergemann et al., 2017). Owing to

597 aforementioned structo-chemical interactions, and the lack of an adequate spatial
598 analysis technique for fine-grained minerals, the K-Ar data in the northern area
599 provides no constraints on the timing of the Pfaffenchof- and Gadmén- deformation
600 phases. However, all data in high strain tectonites differ from literature data, which is
601 derived from less deformed host rocks (Table 1; Dempster, 1986). Therefore, only
602 the detail knowledge of overprinting relationships and geological evolution allow the
603 interpretation of such fault-related data.
604

605 **7 Summary of white mica evolution**

606 Comparing different deformation processes in granitoid to gneissic basement units,
607 both the mechanical role of deformation and the abundance of sheet silicates
608 increase with decreasing temperature. With decreasing metamorphic grade,
609 microstructures of polymineralic assemblages indicate an evolution from viscous
610 granular flow over solution/precipitation to fracturing, finally resulting in frictional
611 granular flow of sheet-silicate-rich fault gouges. This occurs over a range of varying
612 mineral stabilities, from biotite bearing mylonites over low-temperature mylonites
613 containing white mica/chlorite down to smectite-bearing cohesionless gouges (Fig.
614 14). Particularly the deformation behavior of white mica is linked to mineral reactions,
615 which influence both the chemical adaptation processes and the grain sizes. At high
616 temperatures, the micas are chemically completely equilibrated/reset during
617 multiphase viscous granular flow by processes like dissolution, mass transfer and
618 precipitation within the entire aggregate. This resetting allows robust K-Ar dating of
619 such rock types by multigrain analysis. In the low temperature mylonites, fracturing
620 becomes increasingly important, and local mineral equilibria dominates (Fig. 14). The
621 combined variable efficiency of different chemical exchange processes generates
622 white mica in different grain sizes and compositions. At these conditions, kinetics and
623 the resulting local mineral-chemistry depends strongly on the intergranular transport,
624 reactive bulk composition and the solubility of elements in fluids. The reactivity of
625 white mica is related to the exchange rate with a fluid. This exchange (or
626 equilibration) rate depends on grain size and the overall intergranular connectivity,
627 but can generally be said to decreasing with temperature. Although fluid accessibility
628 is enhanced in the fault gouges and chemical precipitation processes still occur, the
629 latter being demonstrated by the formation of smectite, temperatures and K-activity
630 are too low for white mica/illite equilibrium on the retrograde path in our samples.

631 Identifying equilibrated mica at the sample scale requires the combination of micro-
632 structural data and mineral chemistry. The increasing role of fracturing in low
633 temperature mylonites and decreasing exchange volumes with fluids allows for the
634 preservation of different micas along the T-D path of the investigated mylonites. This
635 preservation in combination and the knowledge of local equilibrium is an important
636 aspect for selecting micas for geochronology but can also be used as a chemical
637 proxy for the detection of local equilibria/disequilibria.

638

639

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650

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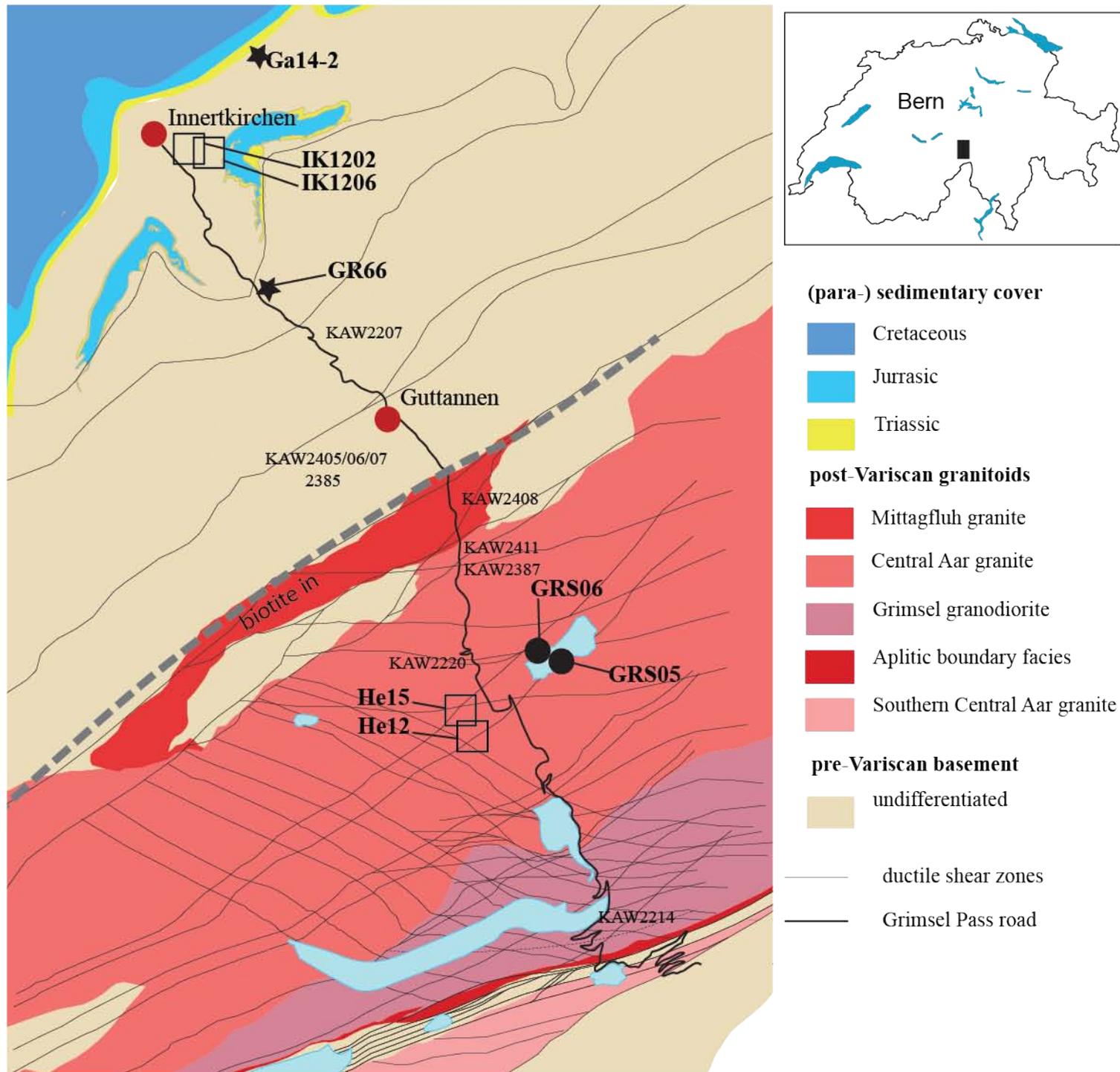


Figure 1:

Map of the Haslital with the Innertkirchen area to the north and the Gelmersee area in the south. Indicated are the surface locations and the tunnels where samples were taken. Also the main ductile shear zones are indicated (Baumberger 2015; Wehrens et al. 2017).

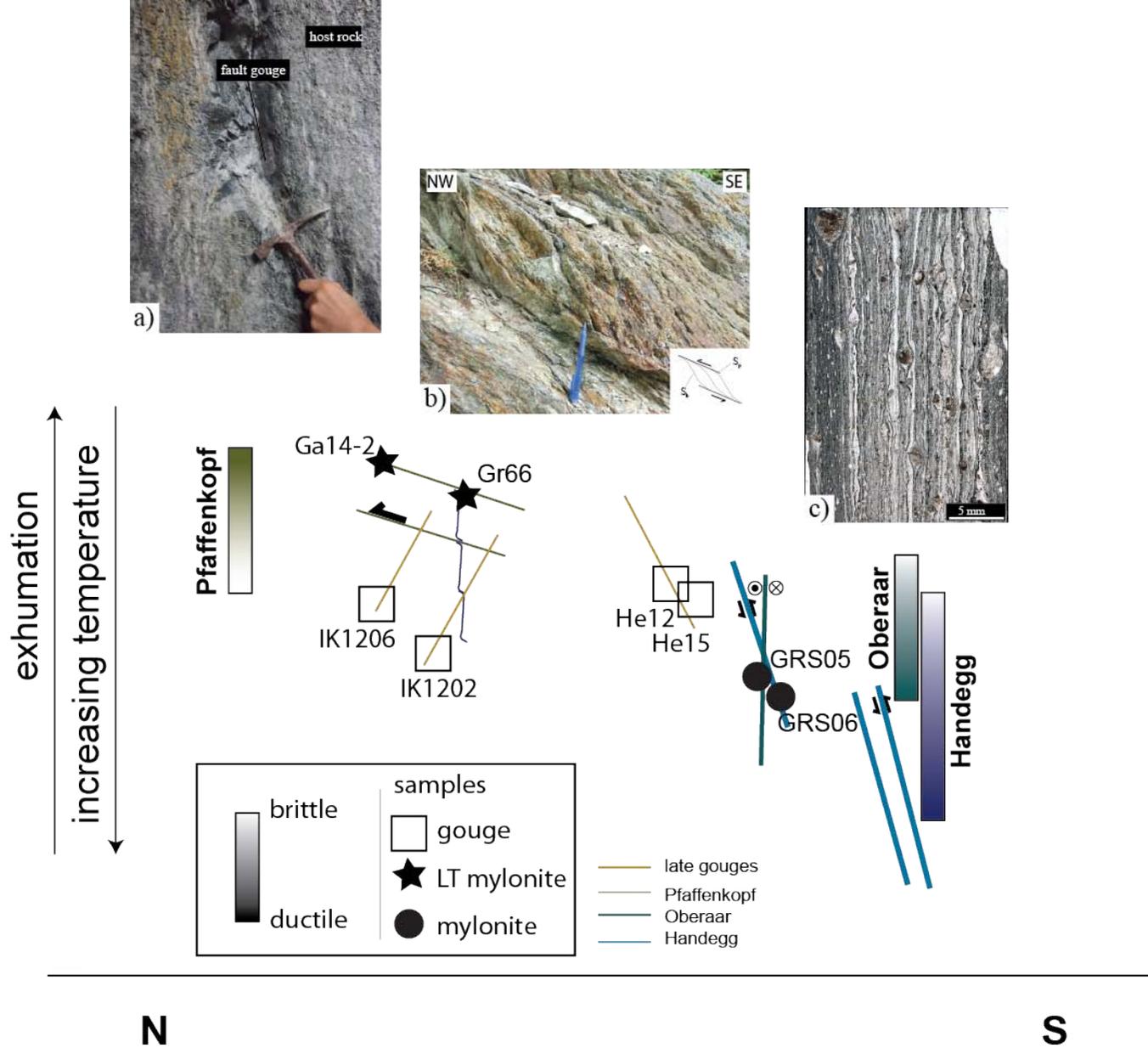


Figure 2: Sketch of structural evolution and the selected samples along a N-S section along the Haslital. (a) Photo of outcrop showing the 10 cm wide fault gouge (b) Outcrop showing the Handegg foliation (S_h) and the Pfaffenkopf foliation (S_p), inset (lower right corner) shows the geometric relation. (c) Micrograph showing the mylonitic fabric of the intermediate T mylonites. (d) Sketch indicating the shear zone orientations and the main conditions of deformation. Note the colors of the Handegg, Oberaar and Pfaffenkopf phase are shown in the orientation and the type of deformation.

Type	mylonite	Low-T mylonite	gouges
sample	GRS05 GRS06	Gr66 Ga14-2	He4/6 IK1202/06
quartz + feldspars	—————		
white mica	—————		
biotite	—————		
chlorite		—————	
smectite			—————
Quartz plasticity	————— ■ ■ ■		

Figure 3:

Mineral distribution in the analysed tectonites.

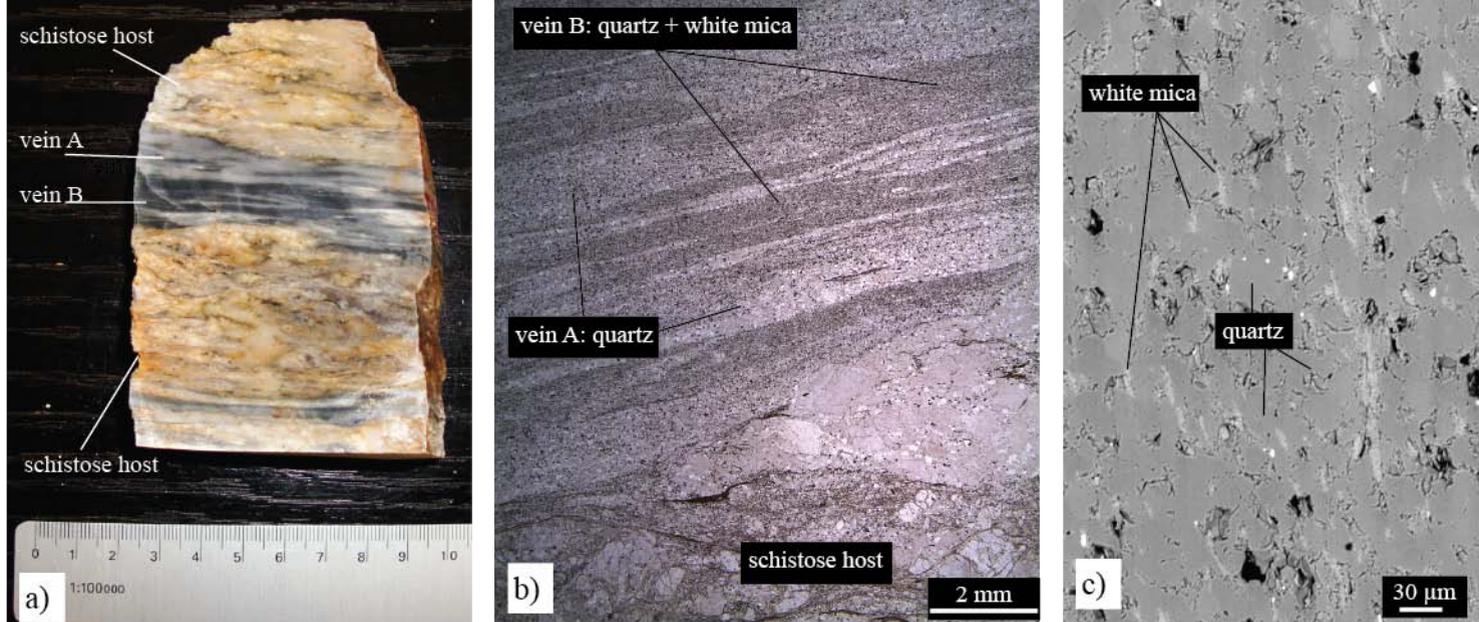


Figure 4:
(a) Hand specimen of sample GRS06 again showing the mylonitized monomineralic quartz veins (vein A) and quartz-mica bands (vein B) within the weakly schistose host. (b) Micrograph of GRS06 showing in the bottom of the image weakly schistose granitoid. In the top half of the image the quartz and white mica veins and pure quartz veins are visible. (c) Electron backscatter image of the white mica quartz vein. Note the aligned and homogeneously distributed white mica grains with sizes down to a few μm .

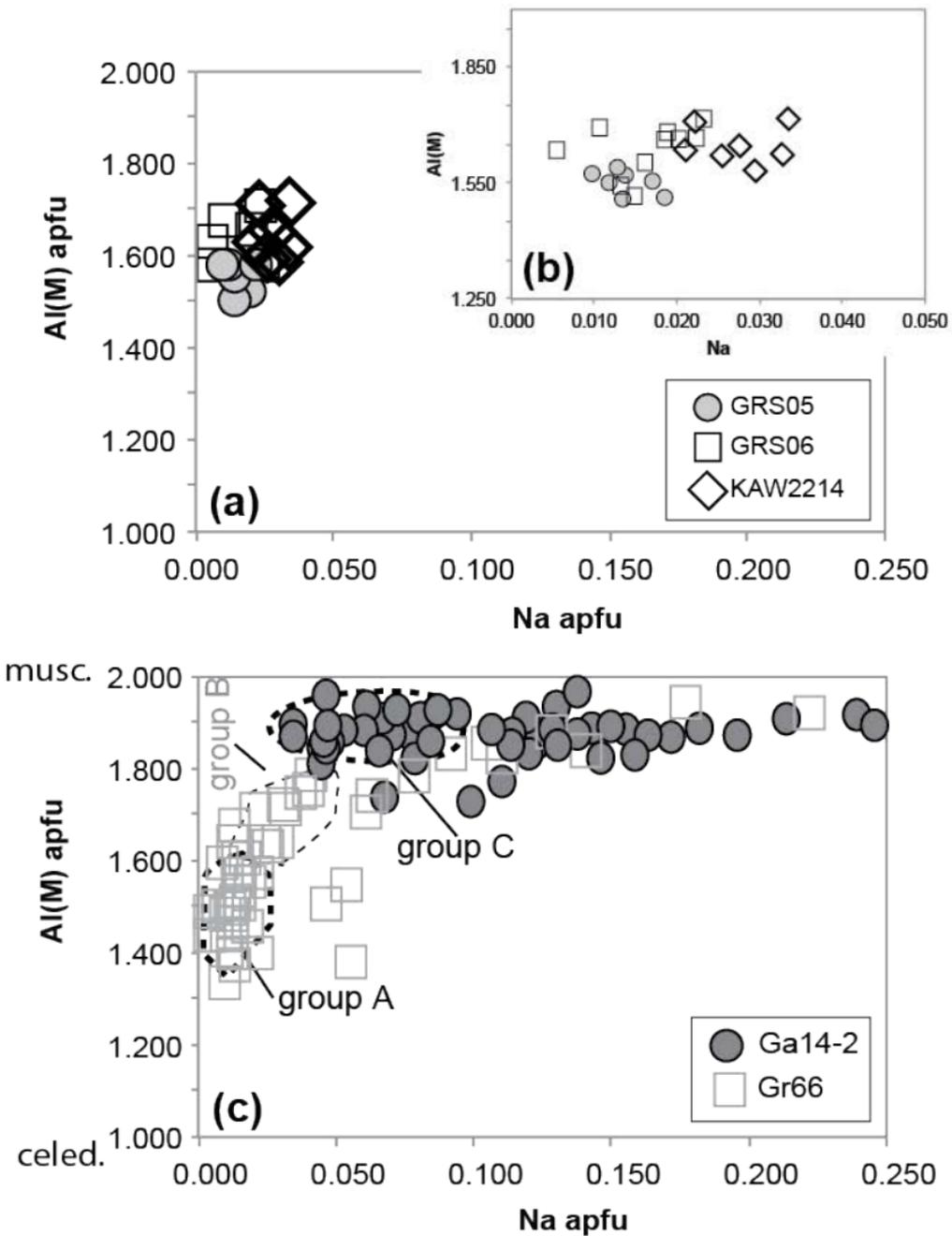


Figure 5:

Mineralchemical data in Al (apfu) versus Na (apfu) diagrams. The Al is recalculated on the M-site. (a) data from the mylonites. Sample KAW2214 is from the south and for comparison only (see Table 1). (b) details of part (a). (c) data from the low-T mylonites (Gr66, Ga14-2).

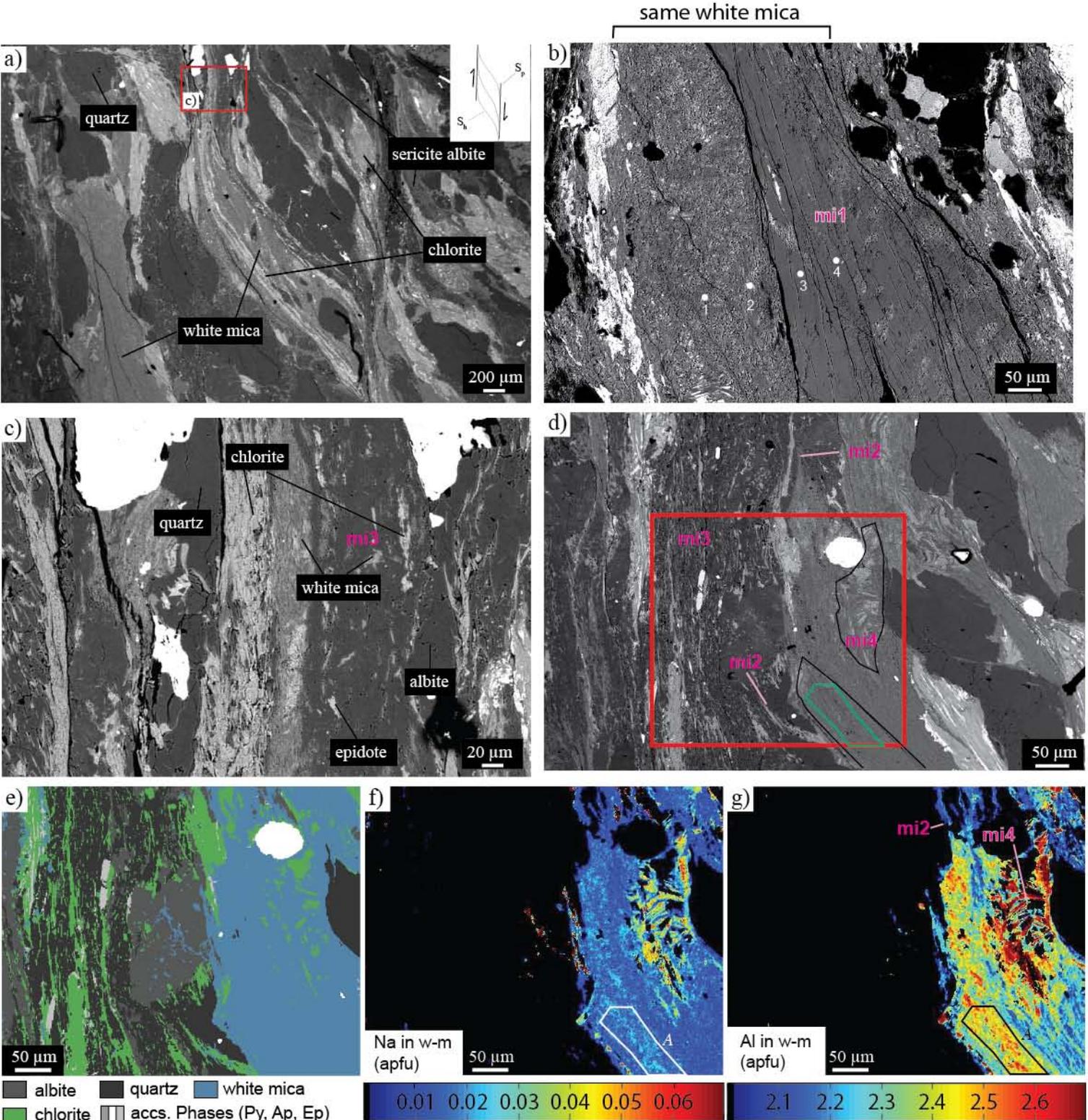


Figure 6:

Microstructures and quantitative maps of sample Gr66 (a-d) Backscatter electron images showing the different textures. (a) overview showing the the different planar fabrics; (c) zoom in of (a) showing the newly formed white mica in the S_p with a grain size $\sim 5\mu\text{m}$. (b) large grain of white mica, which is surrounded by fine grained material. Analytical points are given. (d) bending and recrystallisation of white mica into the S_p . Red angular is the position of quantitative mapping shown in (e-g). (e) phase distribution of the map (f) Na in white mica (unit: atoms per formula unit). (g) Al in white mica (apfu). The different microstructural types (mi1-mi5) are marked in the different images.

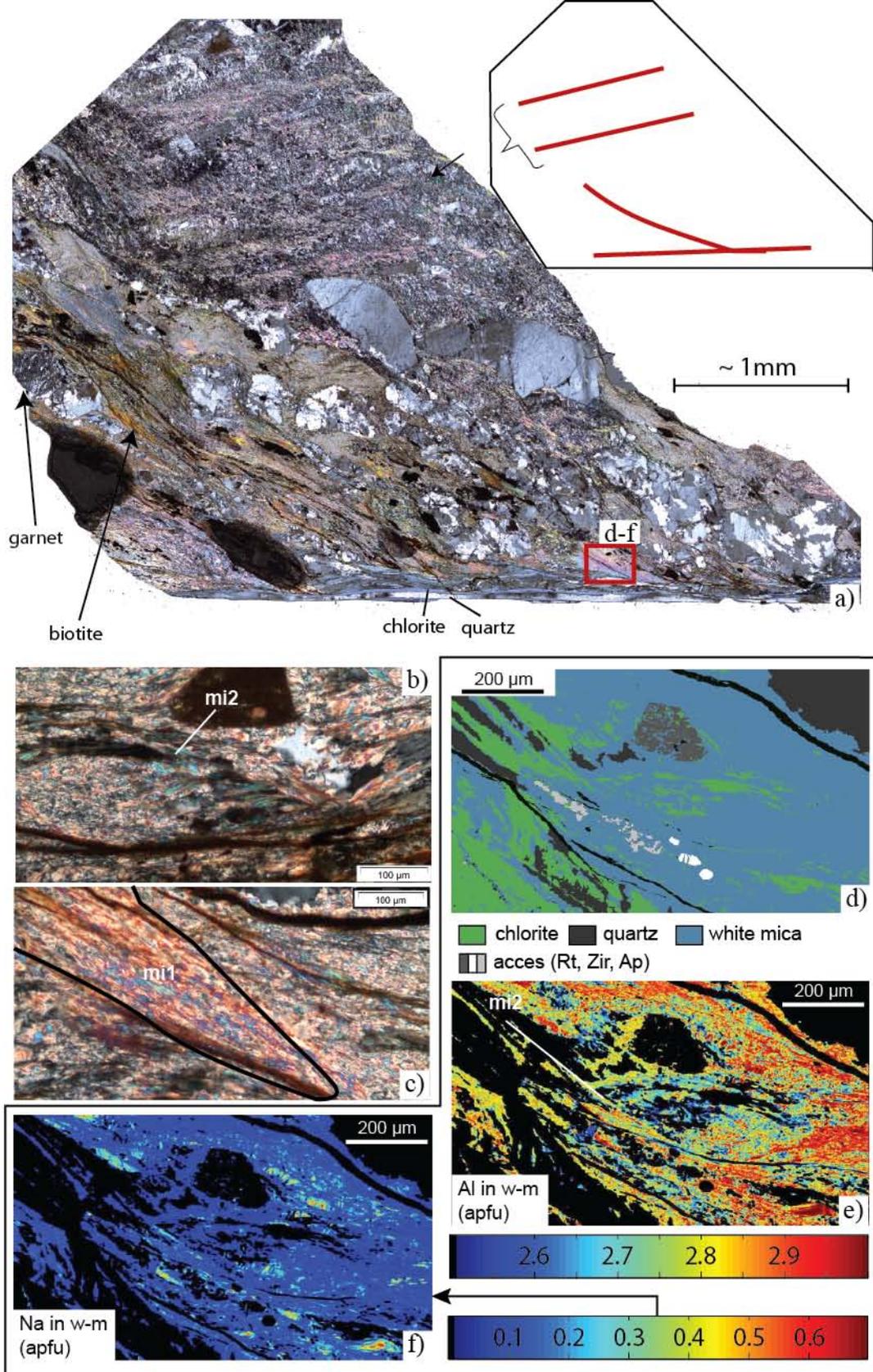


Figure 7: Microtextural map of sample Ga14-2. (a) optical microphotograph of the complete section Red angular is the position of quantitative mapping shown in (b-f). (b) , (c) . (d) phase distribution of the map (e) Al in white mica distribution (unit: atoms per formula unit, apfu). (f) Na in white mica (apfu). The pixels in red are paragonite, whereas the pixels in green (Na = 0.3-0.5 apfu) are mixing analysis between K-white mica and paragonite.

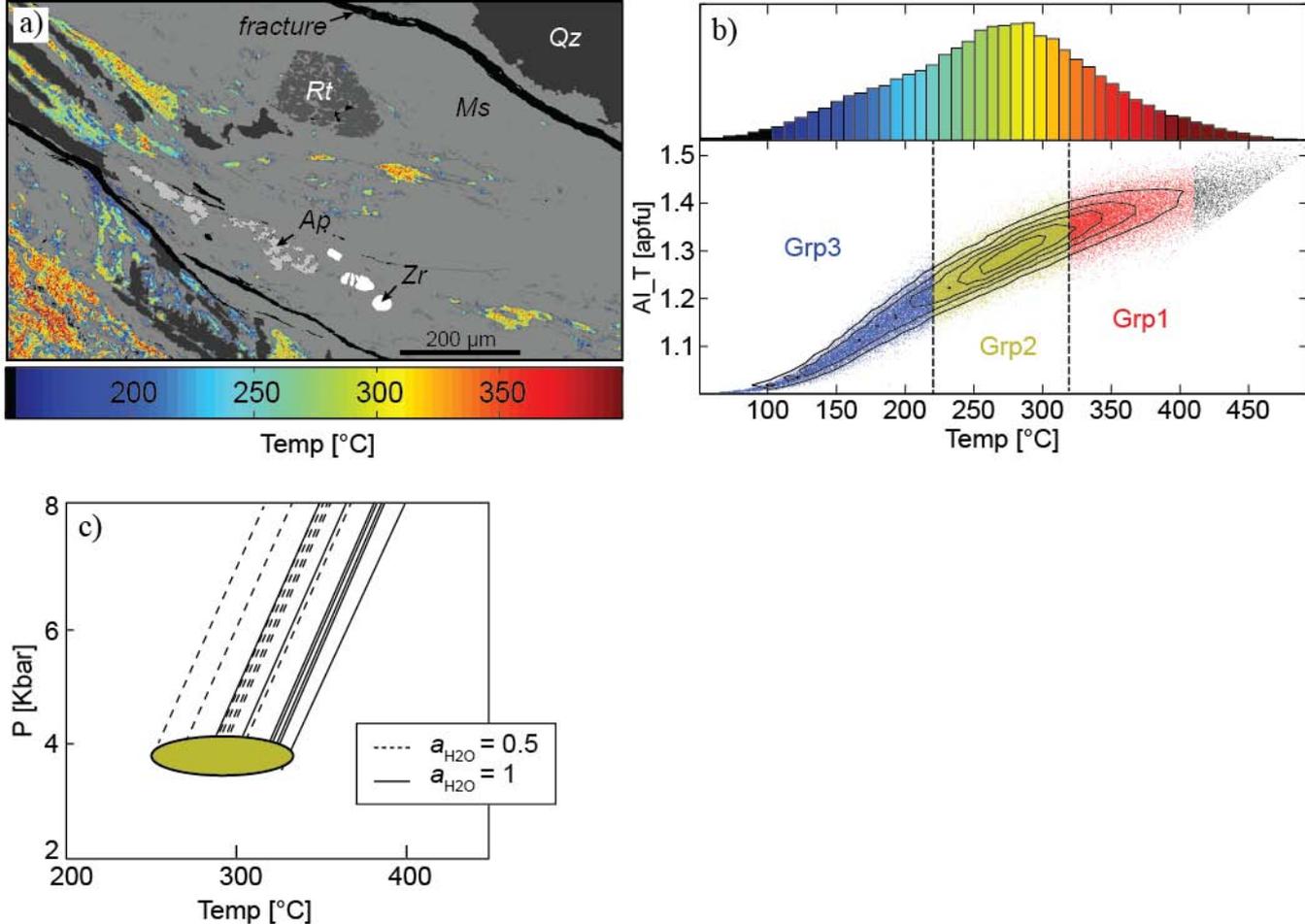


Figure 8:

Chlorite and white mica thermobarometry. (a) Temperature map of chlorite calculated using the calibration of Lanari et al. (2014) at a fixed pressure of 5 kbar. (b) The Temperatures are reported again the amount of Al in tetrahedral position (Al_T) expressed in atom per formula unit (apfu). Three groups are distinguished based on the temperature map and they are reported in (b). The colors of the bars of the temperature histogram are based on the colormap of the temperature map. (c) P-T equilibrium lines between white mica (mi2, see Fig. 7), quartz and water calculated for $a_{\text{H}_2\text{O}} = 1$ (continuous lines) and $a_{\text{H}_2\text{O}} = 0.5$ (dashed lines) using the calibration of Dubacq et al. (2010). The green ellipse show the best intersection between the chlorite temperature (Grp2, see text) and the white mica equilibrium lines.

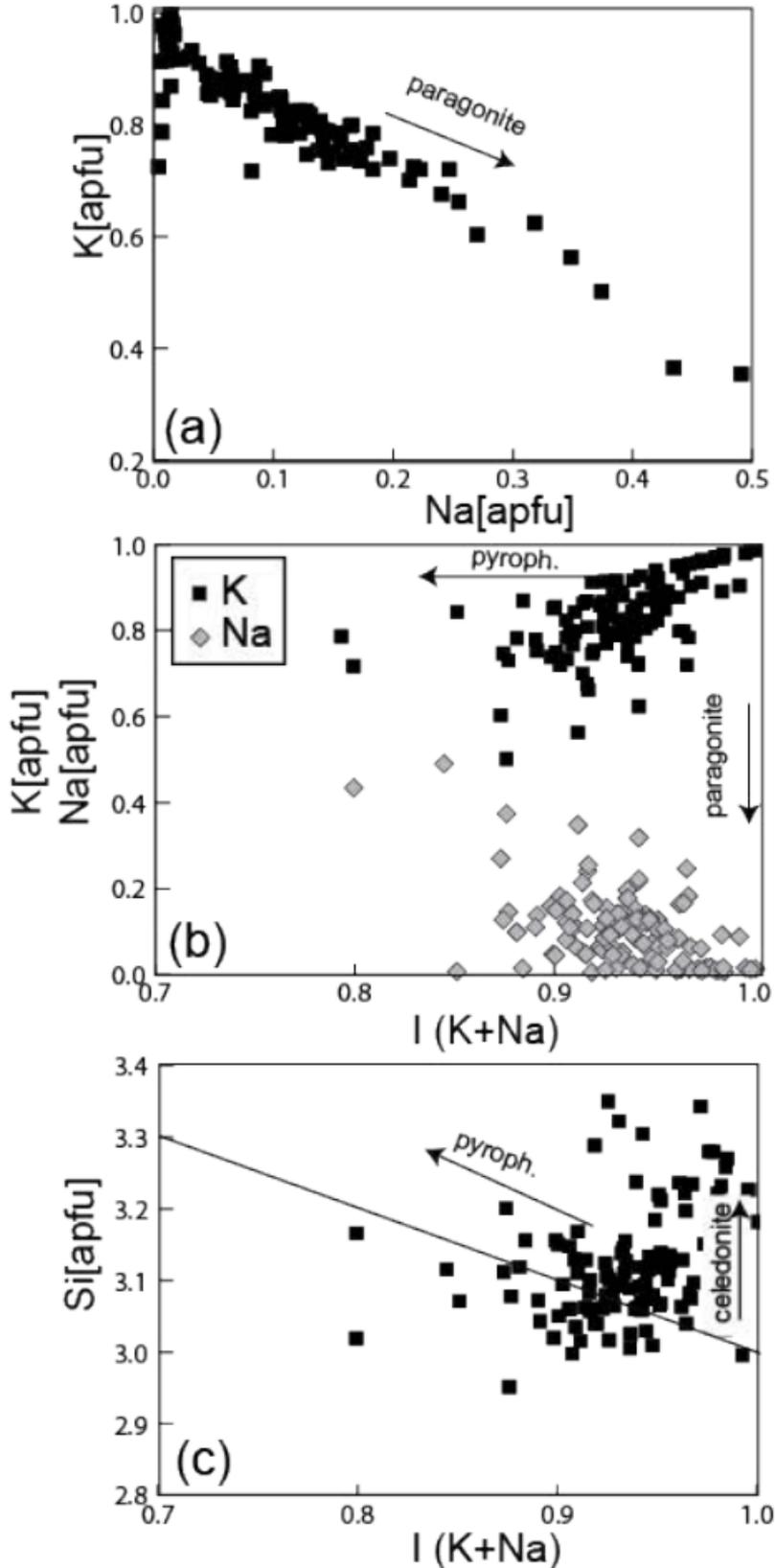


Figure 9:

Mineral chemistry of white mica in low-T mylonites. (a) Na versus K per formula unit, the paragonite exchange vector is indicated. (b) amount of K+Na versus K and versus Na; (c) K+Na versus Si content. The pyrophyllite vector and limits are indicated.

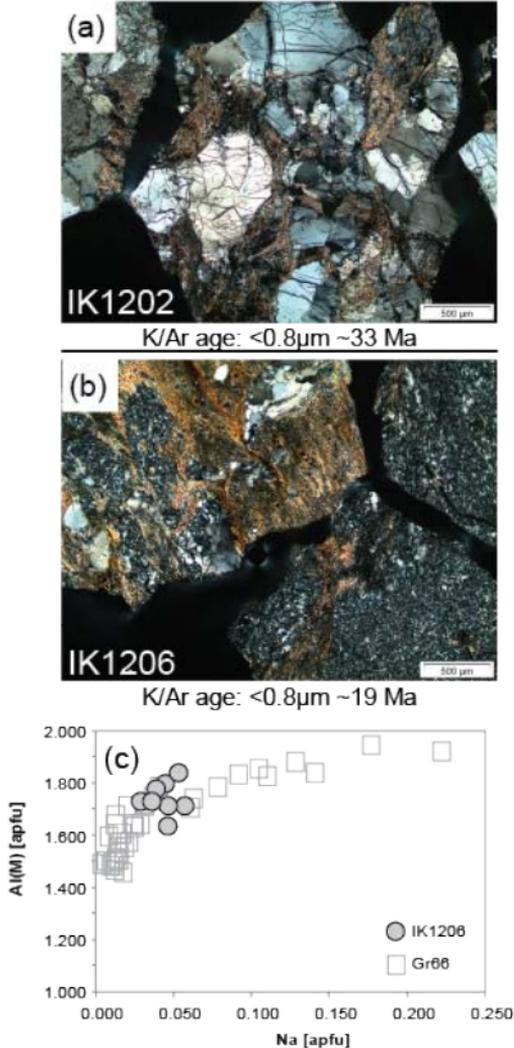


Figure 10:

The connection of fragments in gouges and their evolution. (a+b) Microphotograph of the two southern investigated gouges. Note the different recrystallisation of the quartz and the related apparent ages. (c) Microprobe data of the micas inside the fragments of sample IK1206.

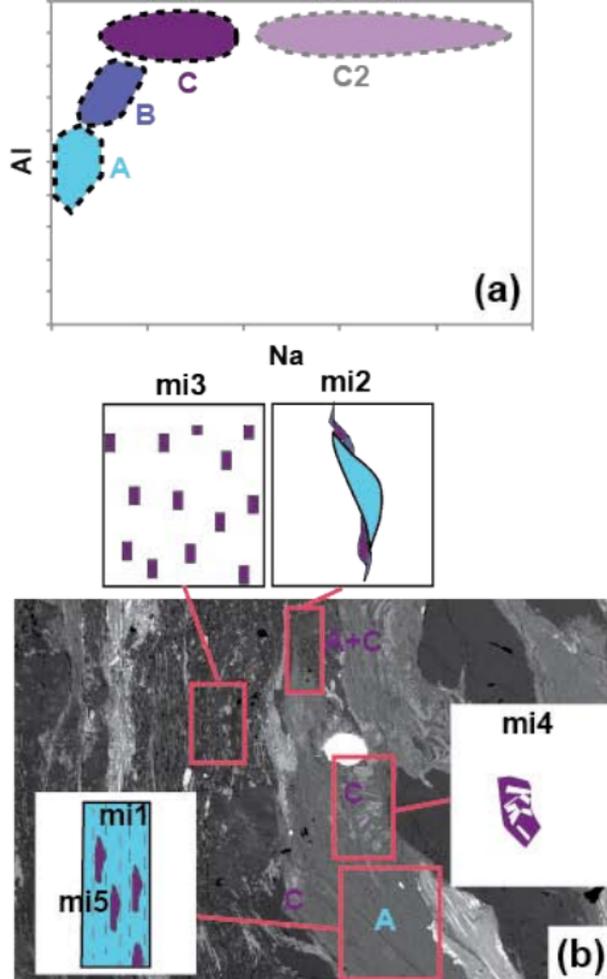


Figure 11:

Relationship between microstructural- (*mi1-mi5*) and chemical groups (A, B, C). (a) Scheme of the mica composition, simplified from figure 5; (b) microphotograph from figure 6 with schematic microstructural groups (Table 7). Capital letters indicate the chemical mica groups (see figure 6f+g).

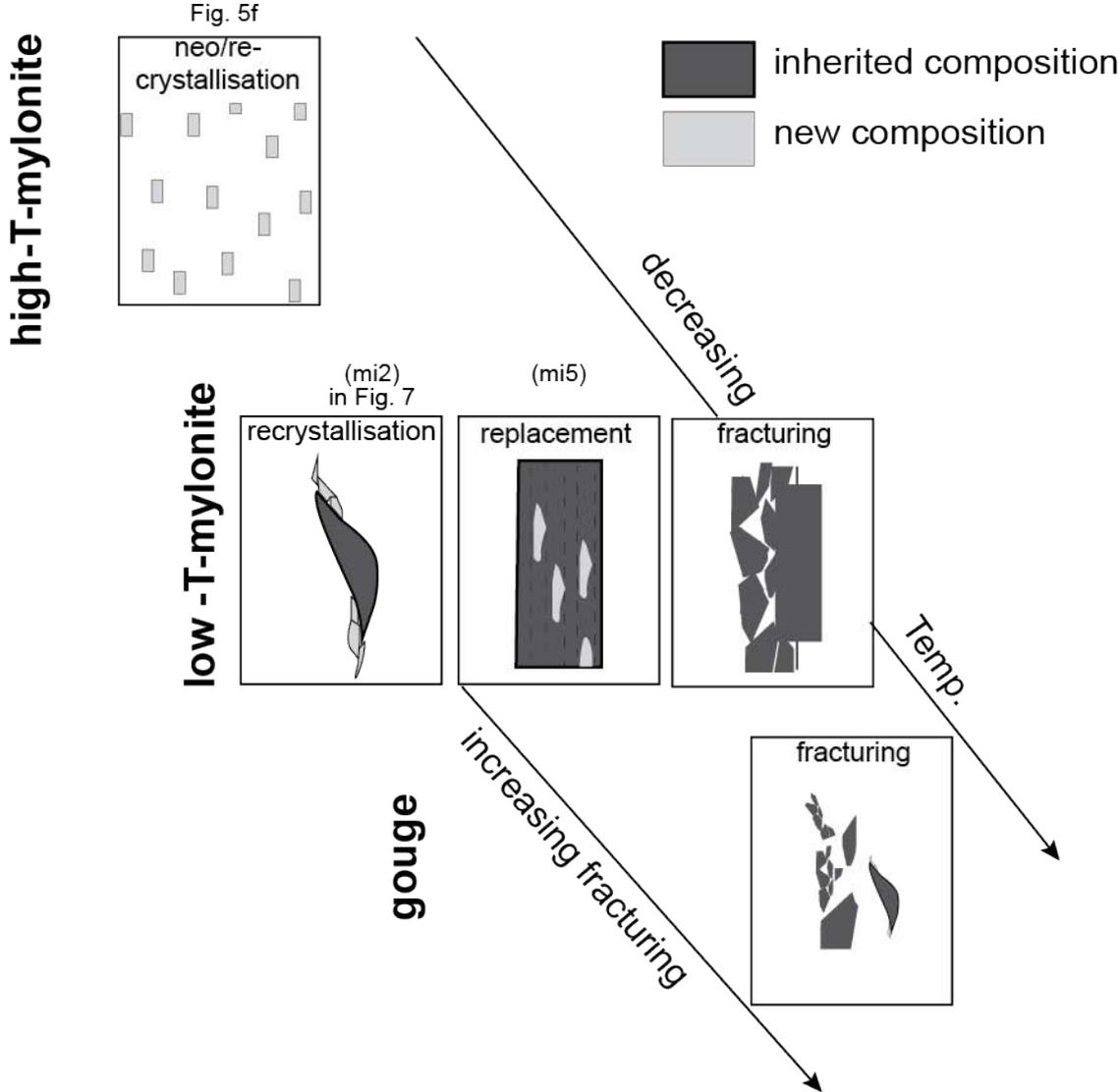


Figure 12:

Sketch of different microstructures described along the temperature evolution

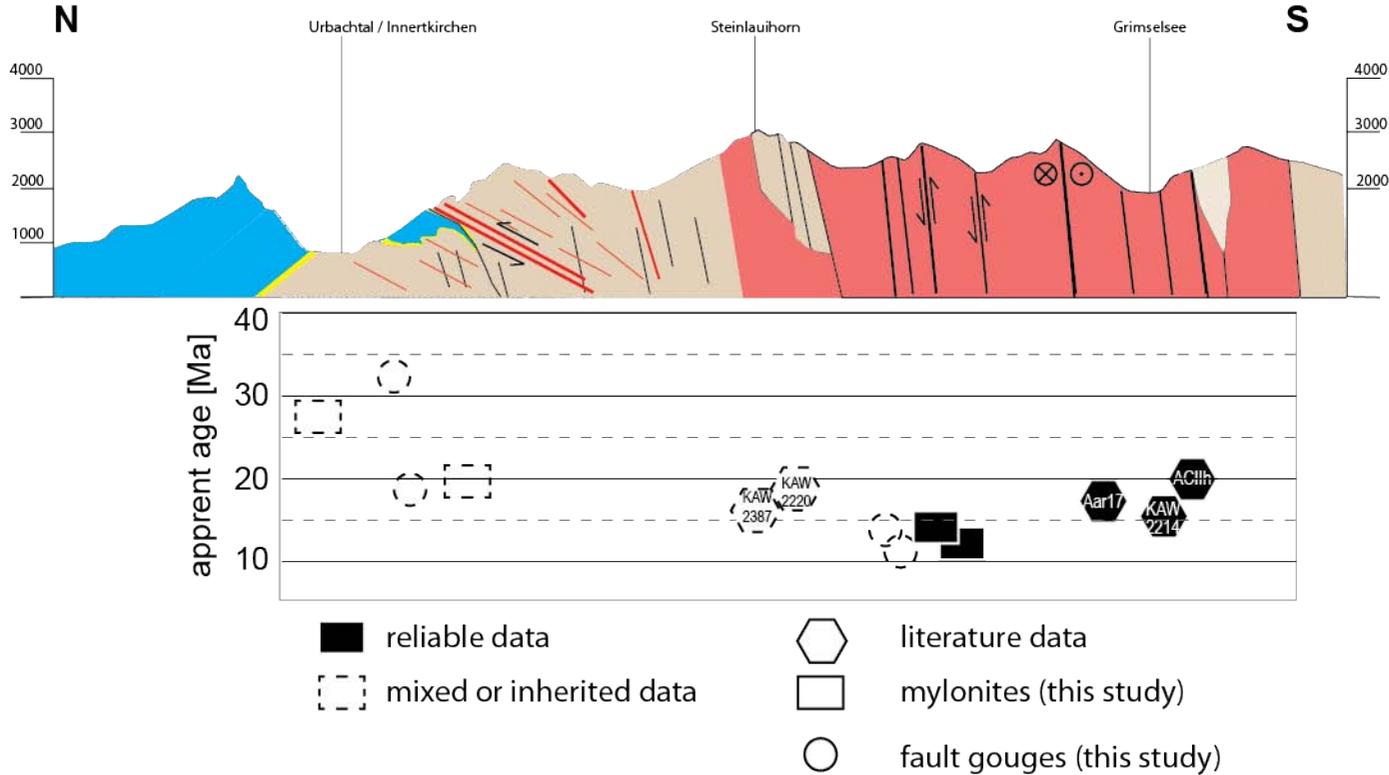


Figure 12:

Distribution of white mica Ar-ages along the Halsital. Data are from Table 1.

Table 1: Available K/Ar data sorted from N to S and coordinates for the investigated samples

	Sample	coordinates	rock-type	biotite	white mica		Reference	
				K/Ar	K/Ar age smallest fraction	Ar age oldest age		
N	Ga14-2	670190 176980	B		27±0.5	37±0.8	this study	
	KAW2207	661631 170978	D	246±3			D86	
	Ik1202	661316 172478	C		33±0.7	37±0.7	this study	
	Ik1206	661254 172514	C		19±0.4	21±0.4	this study	
	Gr66	662470 169634	B		20±0.4	40±0.8	this study	
	KAW2404	664131 168058	D	171±3			D86	
	KAW2385		D	78±2		~180	D86	
	-	KAW2407	665971 166358	D	112±2			D86
		KAW2408	666200 166180	E	54±1			D86
		KAW2387	666000 164400	E			16±1	D86
	KAW2220	666400 163000	E	23±0.5		18±1	D86	
	He12	666470 161622	C		12±0.3	11±0.3	this study	
	He15	666362 161794	C		-	14±0.4	this study	
	GRS05	668125 162697	A		12±0.3		this study	
	GRS06	667629 162940	A		14±0.3		this study	
	Aar17	667900 160450	E			17±0.1	C08	
	KAW2214	668900 158500	E	19±0.5		15±1	D86	
	ACIih	668500 158400	B			19±0.1	C08	
	Aa0351	668810 157290	B			14±0.1	R09	
S	S123	664240 155480	B			13±0.1	R09	

A: mylonite, B: low temperature-mylonite, C: fault gouge, D: basement gneiss, E: granitic gneiss
D86: Dempster 1986; C08: Challandes et al. 2008; R09: Rolland et al. 2009

Table 2: Overview of the separation and analytical methods for each sample and fraction

Sample	grain size fraction (μm)	Selfrag	Grain-size separat.	Mineralogy
GRS05	2- 6	x	C	X
GRS05	6- 10	x	A	X
GRS06	2- 6	x	C	X
GRS06	6- 10	x	A	X
GRS06	10- 20	x	A	X
GRS06	63- 500	x	p	TS
GR66	2- 6	x	C	X
GR66	63- 500	x	P	TS
Ga14-2	sep.		P	TS
Ga14-2	2-6	x	A	TS
Ga14-2	2-0.8	x	A	TS
Ga14-2	<0.8	x	A	TS
He12	< 0.8		C	X,G,H
He12	0.8- 2		C	X,G,H
He12	2- 5		C	X,G,H
He15	< 0.8		C	X,G,H
He15	0.8- 2		C	X,G,H
He15	2- 5		C	X,G,H
Ik1202	< 0.8		C	X,G,H
Ik1202	0.8- 2		C	X,G,H
Ik1202	2- 5		C	X,G,H
Ik1206	< 0.8		C	X,G,H
Ik1206	0.8- 2		C	X,G,H
Ik1206	2- 5		C	X,G,H

Abbreviations: C: centrifuge, A: Atterberg, P: hand-picked, X: XRD, G: glycolated, H: Heat treated, TS: thin-section

Table 3: Standards measured during the course of this research

Standard ID	K [%]	Rad. 40Ar [mol/g]	Rad. 40Ar [%]	Age [Ma]	Error [Ma]	Error to reference [%]
GLO-154	6.55	1.1196E-09	94.35	95.96	1.49	+0.98
GLO-157	6.55	1.1093E-09	94.54	95.09	1.50	+0.06
HD-B1-121	7.96	3.3294E-10	90.46	23.97	0.37	-0.99
HD-B1-124	7.96	5.4850E-11	92.58	24.31	0.37	+0.41
HD-B1-125	7.96	3.3574E-10	91.97	24.17	0.36	-0.17

Airshot ID	40Ar/36Ar	+/-
AS116-AirS-1	295.69	0.36
AS117 AirS-1	296.86	0.31
AS118-AirS-1	295.17	0.56
AS120-AirS-1	295.55	0.29
AS121-AirS-1	296.96	0.24

Table 4: Representative white mica analyses

sample	KAW2214	KAW2214	GRS05	GRS06	Gr66	Ga14-2	Gr66	Ga14-2
					groupA	groupC	groupC	groupC2
SiO ₂	47.17	47.44	45.97	48.22	50.19	45.68	47.41	45.85
TiO ₂	0.04	0.04	0.17	0.05	0.04	bd	0.02	0.03
Al ₂ O ₃	33.37	30.39	30.15	31.05	27.49	35.44	32.52	36.22
FeO	2.88	4.19	4.73	2.80	2.89	1.69	1.66	1.40
MnO	bd	0.16	1.00	bd	bd	0.01	0.04	0.03
MgO	0.94	1.88	2.05	2.78	2.98	0.94	1.95	0.49
CaO	0.08	bd	bd	bd	bd	bd	0.03	0.14
Na ₂ O	0.64	0.19	0.10	0.04	0.08	0.65	0.29	1.65
K ₂ O	10.97	11.45	11.68	11.69	11.48	10.26	10.63	8.22
sum	96.09	95.74	95.85	96.63	95.15	94.67	94.56	94.02

Table 5: Composition of the analyzed samples and grain size fractions. Note that the distinction between chlorite and smectite has only been made for the finest grain sizes of the gouges. The amount of each phase is estimated and indicated from minor to dominant with "x" to "xxxx" respectively. The "(x)" refers to a phase which could not unequivocally be determined.

Sample	grain size fraction (μm)	smectite	kaolinite	illite/ muscovite	biotite	quartz	k-feldpsar	albite	chlorite
GRS05	2- 6			xxx	xxx	xx	x	x	x
GRS05	6- 10			xxx	xxxx	xx	x	xx	x
GRS06	2- 6			xxx		xxx		xx	
GRS06	6- 10			xxx		xxx		xx	
GRS06	10- 20			xxx		xxxx		xx	
GRS06	63- 500			x					
He12	< 0.8	xxxx	x	xx			(x)		
He12	0.8- 2	xxx	x	xxx			(x)		
He12	2- 5	xx	x	xxxx			(x)		
He15	< 0.8			xx					x
He15	0.8- 2	xxxx	x	xx					
He15	2- 5	xxx	x	xxx					x
GR66	2- 6			xxx		xxx		x	xxxx
GR66	63- 500			x					x
lk1202	< 0.8	xxx	xx	xxxx					x
lk1202	0.8- 2	xx	xxx	xxxx					x
lk1202	2- 5	xx	xxx	xxxx					x
lk1206	< 0.8	xx	x	xxx				x	
lk1206	0.8- 2	xx	x	xxxx				x	
lk1206	2- 5	xx	x	xxxx				x	

Table 6: K-Ar data for all measured samples and fractions

Sample	tecto. group *	grain size fraction (μm)	K [%]	Rad. 40Ar [mol/g]	Rad. 40Ar [%]	Age [Ma]	Error [Ma]
GRS05	A	2- 6	5.31	1.1295E-10	82.18	12.2	0.3
GRS05	A	6- 10	5.80	1.1947E-10	81.74	11.8	0.2
GRS06	A	2- 6	5.16	1.2108E-10	87.79	13.5	0.3
GRS06	A	6- 10	4.39	1.0081E-10	92.18	13.2	0.3
GRS06	A	10- 20	4.61	1.0679E-10	89.83	13.3	0.3
GRS06	A	63- 500	6.65	1.5696E-10	94.72	13.6	0.3
He12	C	< 0.8	3.70	7.531E-11	34.15	11.7	0.3
He12	C	0.8- 2	4.55	1.0152E-10	40.95	12.8	0.3
He12	C	2- 5	5.23	1.0238E-10	34.66	11.3	0.3
He15	C	< 0.8	1.73	4.8349E-11	13.18	16.0	1.0
He15	C	0.8- 2	2.28	5.538E-11	35.65	14.0	0.5
He15	C	2- 5	3.62	9.0493E-11	31.96	14.4	0.4
Gr66	B	2- 6	5.80	2.0111E-10	91.38	19.9	0.4
Gr66	B	63- 500	3.55	2.4762E-10	88.62	39.8	0.8
Ga14-2	B	63- 500	3.18	1.989E-10	84.5	35.7	0.7
Ga14-2	B	2-6	4.89	3.178E-10	90.4	37.1	0.8
Ga14-2	B	2-0.8	5.03	2.629E-10	89.3	29.9	0.6
Ga14-2	B	<0.8	4.96	2.298E-10	86.6	26.5	0.5
Ik1202	C	< 0.8	6.30	3.6361E-10	84.98	33.0	0.7
Ik1202	C	0.8- 2	6.78	3.8377E-10	90.79	32.3	0.6
Ik1202	C	2- 5	6.49	4.1885E-10	89.35	36.8	0.7
Ik1206	C	< 0.8	6.68	2.1974E-10	74.26	18.9	0.4
Ik1206	C	0.8- 2	6.14	1.9892E-10	65.18	18.6	0.4
Ik1206	C	2- 5	6.76	2.4872E-10	77.51	21.1	0.4

A: mylonites, B: LT-mylonites, C: fault gouges

Table 7: Microstructural groups of low T mylonites

Group	description	grain size	Figure
<i>mi1</i>	relic grains	large	6
<i>mi2</i>	fractures/recrystallized	interm.	6, 7
<i>mi3</i>	precipitated in qtz/fsp matrix	small	6
<i>mi4</i>	pseudomorphs	variable	6, 7
<i>mi5</i>	replacement	large	6