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Title: Anatexis and fluid regime of the deep continental crust: new clues from melt and fluid inclusions in metapelitic migmatites from Ivrea Zone (NW Italy)

Short running title: Anatexis and fluid regime: clues from melt inclusions

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28 **ABSTRACT**

29 We investigate the inclusions hosted in peritectic garnet from metapelitic migmatites of the
30 Kinzigite Formation (Ivrea Zone, NW Italy) to evaluate the starting composition of the anatectic melt
31 and fluid regime during anatexis throughout the upper amphibolite-facies, transitional and granulite-
32 facies zones. Inclusions have negative crystal shapes, size from 2 to 10 μm and are regularly
33 distributed in the core of the garnet. Microstructural and micro-Raman investigations have shown the
34 presence of two types of inclusions: crystallized silicate melt inclusions (i.e. nanogranitoids, hereafter
35 NI) and fluid inclusions (FI). Microstructural evidence suggests that FI and NI coexist in the same
36 cluster and are primary (i.e. were trapped simultaneously during garnet growth). FI have similar
37 composition in the three zones and comprise variable proportions of CO_2 , CH_4 and N_2 , commonly with
38 siderite, pyrophyllite and kaolinite. The mineral assemblage in the NI contains K-feldspar, plagioclase,
39 quartz, biotite, muscovite, chlorite, graphite and, sometimes, calcite. Polymorphs such as kumdykolite,
40 cristobalite and tridymite were also found.

41 Re-homogenized NI from the different zones and show that all the melts are leucogranitic but
42 have slightly different compositions. In samples from the upper amphibolite facies, melts are less
43 mafic ($\text{FeO}+\text{MgO}$ 2- 3.4 wt. %), contain 860-1700 ppm CO_2 and reach the highest H_2O contents (6.5-
44 10 wt. %). In the transition zone melts have intermediate H_2O (4.8-8.5 wt. %), CO_2 (457-1534 ppm)
45 and maficity ($\text{FeO}+\text{MgO}$ 2.3- 3.9 wt. %). In contrast, melts at granulite facies reach highest CaO,
46 $\text{FeO}+\text{MgO}$ (3.2-4.7 wt. %) and CO_2 (up to 2400 ppm), with H_2O contents comparable (5.4-8.3 wt. %)
47 to the other two zones.

48 Our data suggests that anatexis of these metapelites occurred through muscovite and biotite
49 breakdown melting in the presence of a COH fluid. The fluid is assumed to be internally derived,
50 produced initially by devolatilization of hydrous silicates in the graphitic protolith, then as result of
51 oxidation of carbon by melting of Fe^{3+} -bearing biotite. Variation in the composition of the melts are
52 interpreted to result from combination of higher T of melting, greater contribution of anhydrous
53 reactants (such as plagioclase) and minor role of apatite. The H_2O contents of the melts throughout the
54 three zones are comparable and higher than usually assumed. While the CO_2 contents are highest at

55 granulite facies, and show that carbon-contents of crustal magmas may not be completely negligible.

56 The $a_{\text{H}_2\text{O}}$ of the fluid dissolved in granitic melts decreases with increasing metamorphic grade.

57

58 Key words: melt inclusions, fluid inclusions, anatexis, CO_2 - H_2O contents of melts, Ivrea Zone.

59

60 **1. INTRODUCTION**

61 The rise of temperature during metamorphism occurs in response to tectonic processes that
62 build orogenic and collisional mountain belts of the Earth, or that extend the lithosphere during
63 continental rifting. In particular, the high- (HT) to ultrahigh-temperature (UHT) metamorphism may
64 lead to partial melting of the middle and lower crust. Anatexis, subsequent segregation and transfer of
65 the crustal melt to the upper crust are the mechanisms that control crustal differentiation (e.g.
66 Vielzeuf, Clemens, Pin, & Moinet, 1990; Sawyer, Cesare, & Brown, 2011; Brown, 2013). As a result,
67 the continental crust has become progressively stratified throughout Earth's history, with a more
68 residual and mafic lower crust, enriched in elements such as FeO, MgO, Al_2O_3 , CaO and a more silicic
69 upper crust, enriched in K_2O , SiO_2 and incompatible trace elements.

70 Migmatites and granulites are widespread in the deep crust and, once at the surface, provide
71 fundamental information on how crustal magmas are generated. However, the investigation of melt
72 rich-rocks (i.e. leucosomes, diatexites) is not a straightforward way to access the starting compositions
73 of the melts, especially in terms of volatiles. The main reason is because there are several processes
74 happening simultaneously at the source that modify the melt compositions; for instance, crystal
75 fractionation (e.g., Sawyer, 1987; Milord, Sawyer, & Brown, 2001; Brown et al., 2016; Carvalho,
76 Sawyer, & Janasi, 2016), restite unmixing (Chappell, White, & Wyborn, 1987), entrainment of
77 peritectic or residual phases (Stevens, Villaros, & Moyen, 2007; Clemens & Stevens, 2012; Sawyer,
78 2014), entrainment of less fertile lithologies (Carvalho, Sawyer, & Janasi, 2017) and volatile
79 exsolution and diffusion (White & Powell, 2010).

80 An alternative, but very robust, way to recover the initial composition of the melts is to use
81 melt inclusions (MI) or nanogranitoid inclusions (NI) found in peritectic minerals (Cesare, Ferrero,

82 Salvioli-Mariani, Pedron, & Cavallo, 2009; Cesare, Acosta-Vigil, Bartoli, & Ferrero, 2015, Bartoli,
83 Acosta-Vigil, Ferrero, & Cesare, 2016; Ferrero & Angel, 2018). Peritectic minerals form as result of
84 incongruent melting reactions, and in some cases, they can trap droplets of the coexisting anatectic
85 melt (Cesare et al., 2015) and of the fluid immiscible with it, if present (e.g. Ferrero, Braga, Berkesi,
86 Cesare, & Laridhi, 2014; Tacchetto et al., 2018). These droplets of melt can be preserved as tiny (2-10
87 μm) glassy or crystallized (i.e. nanogranitoid) inclusions and they are the best way to investigate the in
88 situ starting composition of the melt and fluid regime during crustal melting (e.g. Bartoli et al., 2013a;
89 Bartoli, Acosta-Vigil, Ferrero, & Cesare, 2016).

90 In the latest years, the MI and NI have become fundamental tools to investigate crustal
91 petrology and geochemistry. Despite of their small size, which poses analytical challenges, these
92 inclusions have helped to unravel the anatectic history of polymetamorphic terranes (Acosta-Vigil et
93 al., 2016), fluid regime of high grade terranes and initial volatile contents of granitic magmas (Bartoli,
94 Cesare, Remusat, Acosta-Vigil, & Poli, 2014; Ferrero, Wunder, Walczak, O'Brien, & Ziemann, 2015;
95 Bartoli et al., 2016). Composition of MI can also be useful to evaluate melt loss and reconstruct the
96 prograde history of granulite terranes (Bartoli, 2017; 2018).

97 The Ivrea Zone, NW Italy, is a world-renowned section of mid to lower Permian continental
98 crust where the preserved field gradient offers the opportunity to examine the evolution of crustal
99 melting in nature. In this contribution, we describe for the first time the coexistence of primary melt
100 and fluid inclusions in garnet from metapelitic migmatites of Val Strona di Omegna and use them to
101 investigate crustal anatexis of this high-grade terrane from upper amphibolite to granulite facies,
102 thereby contributing to better understand the processes that lead to crustal differentiation. The main
103 points addressed are: (i) compositions of the melts ii) their evolution with increasing metamorphic
104 grade, (iii) their implications for fluid regime of this high grade terrane.

105

106 2. GEOLOGICAL SETTINGS

107 The Ivrea Zone (IZ, NW Italy) is a section of the mid to lower Permian continental crust
108 which comprises very well-preserved high-grade rocks (Schmid & Wood, 1976), and is limited by the
109 Insubric line at the west and by the Cossato-Mergozzo-Brissago Line at east (Fig. 1a).

110 Two main units are recognized in the IZ, the Mafic Complex and the supracrustal Kinzigite
111 Formation. The Mafic Complex comprises gabbroic, noritic and dioritic rocks which have intruded the
112 metasedimentary sequences of the Kinzigite Formation (Sinigoi et al., 1991; Quick et al., 2003). The
113 Kinzigite Formation is mainly composed of metapelites, interlayered with metagreywackes,
114 metabasites, and minor marbles and quartzites (Schmid, 1993; Barboza & Bergantz, 2000; Quick et
115 al., 2003).

116 Metapelites of the Kinzigite Formation show an increase in metamorphic grade from
117 amphibolite facies in the SE to granulite facies in the NW (Schmid & Wood, 1976; Schmid, 1993;
118 Henk, Franz, Teufel, & Oncken, 1997). Some studies have used the term “kinzigite” for amphibolite
119 facies rocks and “stronalite” for rocks showing a granulite facies assemblage (Schnetger, 1994). The
120 Kinzigite Formation of Val Strona di Omegna was subdivided into three zones: amphibolite facies,
121 transition zone and granulite facies (Fig. 1b-d). Rocks from granulite facies zone are considered to
122 have experienced extensive partial melting and melt loss (e.g., Schnetger, 1994, Sinigoi, Quick,
123 Demarchi, & Klötzli, 1994; Redler, White, & Johnson, 2013). *P–T* conditions of metamorphism in Val
124 Strona di Omegna have been widely investigated by conventional thermobarometry (Henk, Franz,
125 Teufel, & Oncken, 1997; Bea & Montero, 1999), phase equilibria modeling (Redler, Johnson, White,
126 & Kunz, 2012; Kunz, Johnson, White, & Redler, 2014; Redler et al., 2013) and trace element
127 thermometry (Luvizotto & Zack, 2009; Ewing, Hermann, & Rubatto, 2013). Maximum *P–T* estimates
128 calculated by Henk et al. (1997) and Bea & Montero (1999) are ≈ 800 °C and ≈ 8 kbar in the granulite
129 facies zone (Fig. 1b). Based on phase equilibrium modeling, Redler et al. (2012) suggested that rocks
130 from Val Strona di Omegna preserve a regional field gradient from 3.5-6.5 kbar at 650-730°C in the
131 upper amphibolite zone, to 11-12 kbar, 900°C at granulites facies zone (Fig. 1b). Peak temperatures of
132 900-950°C are also inferred from Zr-in-rutile thermometry (Luvizotto & Zack, 2009; Ewing et al.,
133 2013).

134 The relationship between the intrusion of the Mafic Complex and the granulite facies
135 metamorphism has been largely debated. Some authors considered that the intrusion of mafic magmas
136 may be the cause for the high-grade metamorphism (Sinigoi et al., 1991; Henk et al., 1997). However,
137 other works have shown that mineral assemblages close to the intrusion preserve evidence of low-
138 pressure (contact) metamorphism, and thus argue that the intrusion of mafic rocks occurred rather later
139 (e.g. Barboza, Bergantz, & Brown, 1999; Barboza & Bergantz, 2000; Redler et al., 2012; Ewing et al.,
140 2013). The regional metamorphism of amphibolite to granulites facies in the Ivrea Zone was firstly
141 dated at 299 Ma (Vavra, Schmid, & Gebauer, 1999); later the age of 316 Ma was reported by Ewing et
142 al. (2013). On the contrary, the intrusion of the Mafic Complex occurred during decompression at 288
143 Ma (Peressini et al., 2007) and caused an additional event of contact metamorphism around the
144 intrusion (Barboza & Bergantz, 2000; Redler et al., 2012). A more recent work (Kunz, Regis, & Engi,
145 2018a) has demonstrated a long duration for the high temperature conditions in the Kinzigite
146 Formation (300-240 Ma) and concludes that such long thermal history requires additional heat
147 sources. The preferred geodynamic model suggests the upwelling of the asthenospheric mantle as the
148 most conceivable source (e.g. Schaltegger et al., 2002; Kunz et al., 2018b).

149

150 **3. METHODS**

151 Samples were collected along the Val Strona di Omegna (Fig. 1a) and petrographic analyses
152 conducted in order to select representative rocks from the three main zones (Fig. 1b-d). A detailed
153 study was conducted in inclusions on 12 thin and doubly-polished thick (80-200 μm) sections using a
154 light polarized microscope.

155 Back-scattered electron (BSE) imaging of the inclusions, together with and semi-quantitative
156 energy dispersive spectroscopy (EDS), were carried out using a CAM SCAN MX3000, equipped with
157 LaB6 cathode at the Dipartimento di Geoscienze, Università di Padova (Italy), and a Sigma Zeiss field
158 emission scanning electron microscope (FESEM) equipped with Oxford XMax EDS Silicon Drifted
159 detector at the CNR-IENI, Padova.

160 Major element concentrations of the minerals (garnet, muscovite and biotite) were determined
161 on polished carbon-coated thin sections of 30 μm thickness using the CAMECA SX50 electron
162 microprobe with five wavelength dispersive spectrometers at the C.N.R.-I.G.G. (Consiglio Nazionale
163 delle Ricerche-Istituto di Geoscienze e Georisorse) Dipartimento di Geoscienze, Università di Padova,
164 Italy. The operating conditions were: 20 nA beam current, 20kV acceleration voltage, and 5 μm beam
165 diameter and counting times of 10s on peak and 5s on background. The composition of re-
166 homogenized melt inclusions was determined using a JEOL JXA 8200 Superprobe at the Dipartimento
167 di Scienze della Terra, Università di Milano, using 15 kV accelerating voltage, 5 nA beam current, 10
168 s on the peak and 5 s background, and 1 μm beam diameter to avoid contamination from the
169 surrounding host. Alkali loss was estimated using leucogranitic standards showing a similar
170 composition to the melts, and correction factors were applied for Na, K, Al and Si. Selected
171 microprobe analyses of minerals from representative samples are given in Table 1, and composition of
172 re-homogenized MI are given in Table 2.

173 Micro-Raman measurements were performed on five representative doubly-polished thick
174 sections (STR69, IVT21, STR22, STR28 and STR36) at the Institute of Earth and Environmental
175 Sciences (University of Potsdam), using a HORIBA Jobin-Yvon LabRAM HR 800 equipped with a
176 Peltier cooled multichannel CCD detector and coupled with a petrographic microscope Olympus
177 BX41. Raman spectra were recorded between 100 and 4000 cm^{-1} , using a 100x objective. Integration
178 time of analyses was of 30 s and 3 accumulations, with a spectral resolution of 10 cm^{-1} .

179 Re-homogenization of the nanogranitoid inclusions was performed on garnet chips or cores
180 separated from polished sections of various thicknesses (from 300 μm to 2 mm). Experiments were
181 done using a single-stage piston cylinder at the Laboratory of Experimental Petrology, Dipartimento di
182 Scienze della Terra, Università di Milano, Italy, using the method described by Bartoli et al. (2013b).
183 Experimental re-melting of the inclusions was performed under the following conditions: 820°C – 1
184 GPa, 20h (sample STR36), 840°C – 1.2 GPa, 5h (sample STR28, experiments BC6 and BC9), 850°C
185 – 1 GPa, 20h (sample IVT21, experiments IVT21 and IV21) and 900°C – 1.2 GPa, 5h (sample STR22,
186 experiments BC8 and BC10). Shorter run durations (5 hours) were done in order to avoid interaction
187 of the melt with the host garnet at higher temperatures. Pressures, generally higher with respect to

188 those recently reported for Val Strona di Omega (Fig. 1b; Redler et al., 2012), were chosen in attempt
189 to exceed the internal pressure of inclusions during experimental remelting and avoid their
190 decrepitation and volatile loss upon heating (cf. Figure 9 in Bartoli et al., 2013b).

191 Due to the very small size of the inclusions, the contents of H₂O and CO₂ of re-homogenized
192 glasses were obtained using a CAMECA Nano Secondary Ion Mass Spectrometry 50 (NanoSIMS) at
193 the Muséum National d'Histoire Naturelle of Paris following the procedure of Bartoli et al. (2014) and
194 Créon, Levresse, Remusat, Bureau, & Carrasco-Núñez (2018). Experimental capsules with exposed
195 re-homogenized inclusions were mounted in Indium to improve vacuum in the analysis chamber
196 (Aubaud et al., 2007) and coated with Au. In order to remove the coating and reach sputtering steady-
197 state, sample surface was presputtered by a Cs⁺ primary beam set at 340 pA and rastered over 5×5 μm²
198 surface area before each analysis. Analyses were performed using a 30 pA Cs⁺ primary beam rastered
199 over 3×3 μm² surface area. However, to avoid surface contamination only ions from the inner 1×1 μm²
200 were collected using the “beam blanking” mode. Secondary ions of ¹²C⁻, ¹⁶OH⁻, ²⁸Si⁻ and ⁵⁶Fe¹⁶O⁻
201 were collected in multicollection mode and used to identify the MI. Mass resolving power was set at
202 minimum 6000, sufficient to resolve interferences on ¹⁶OH⁻. A single analysis comprises a stack of
203 200 cycles, each cycle being 1.024 s long. The vacuum in the analysis chamber remained below
204 2×10⁻⁹ Torr during the session of H₂O and CO₂ measurements.

205 The H₂O and CO₂ contents of the glasses (Table 3) were determined through the measurement
206 of ¹⁶OH⁻/²⁸Si⁻ and ¹²C⁻/²⁸Si⁻ ratios, respectively. These ratios were converted into concentrations using
207 calibration curves determined using standards of known compositions. The standards used for the
208 determination of H₂O contents were three leucogranitic glasses: DL reported in Acosta-Vigil, London,
209 Morgan, & Dewers (2003) with H₂O = 6.5 wt.%; glass LGB1 from Behrens and Jantos (2001) with
210 H₂O = 4.9 wt.%; and the almost anhydrous glass B from Morgan and London (2005) with H₂O = 300
211 ± 42 ppm. The standards used for the determination of CO₂ contents were four trachyandesitic
212 standards (STR 9, 10, 11 and 13) from the Stromboli volcano experimentally doped in carbon and
213 water by Bureau et al. (2003). Concentrations and uncertainties were determined using the R program
214 following the procedure described in Bartoli et al. (2014) and Thomen, Robert, & Remusat (2014).

215 For phase equilibria modeling the model chemical system MnNCKFMASHT (MnO–Na₂O–
216 CaO–K₂O–FeO–MgO–Al₂O₃–SiO₂–H₂O–TiO₂) was used with the normalized bulk rock composition
217 in mol.% based on major element XRF analysis. The H₂O content was assumed on the basis of the
218 measured LOI value. Fe₂O₃ was not considered owing to the very low amounts found by means of iron
219 titration of Val Strona di Omegna samples (0.1 mol.%; Redler et al., 2011). To reconstruct a probable
220 prograde history, the melt-reintegration approach has been applied on sample STR28 (Bartoli, 2017
221 and references therein). The amount of melt to be reintegrated has been chosen to bring the solidus to
222 H₂O-saturated conditions at the pressure of 10 kbar. The melt composition, instead, has been obtained
223 from re-melting experiments of nanogranitoids from sample STR28 (details on the procedure are
224 reported in Bartoli, 2018). Calculations were done by the Gibbs energy minimization using the
225 *Perple_X* 6.7.9 software (Connolly, 2009) with the thermodynamic database of Holland and Powell
226 (2011). The solution models used are: melt from White, Powell, & Holland (2007), garnet from
227 Holland and Powell (1998), biotite from Tajčmanová, Connolly, & Cesare (2009), white mica from
228 Coggon and Holland (2002), plagioclase from Newton, Charlu, & Kleppa (1980) and K-feldspar from
229 Thompson and Hovis (1979). An ideal model was used for cordierite and ilmenite.

230

231 **4. RESULTS**

232 *4.1 Samples and petrography*

233 Samples investigated in this study are metapelitic migmatites collected from the three main
234 metamorphic zones of the Ivrea Zone, in Val Strona di Omegna (Fig. 1a): upper amphibolite facies
235 (UA), transition zone (T) and granulite facies (G, see Fig. 1b-d). For further details on the field aspects
236 of the three zones, the reader is referred to Redler et al. (2012), Redler, White, & Johnson (2013) and
237 references therein.

238 In the UA zone, the studied rocks (STR36 and STR77) are metatexites with thin (up to ~5
239 mm) leucosomes (Fig. 2a) as patches or along the folded foliation. The melanosome is lepidoblastic,
240 fine-grained, and comprises biotite (X_{Mg} 0.42; Ti 0.42 a.p.f.u.), fibrolitic sillimanite, quartz,
241 plagioclase, small porphyroblasts of garnet, K-feldspar and minor relict of muscovite (X_{Mg} 0.46; Fig.

242 3a). Sample STR36 is located in the transition zone from Redler et al. (2012) (Fig. 1b,c), however it is
243 compositionally and microstructurally similar to STR77, sampled at upper amphibolite zone. This is
244 actually in agreement with the amphibolite zone proposed by Schmid (1993), in which case the
245 transition zone is more restricted (Fig. 1d). An important accessory mineral is graphite which occurs in
246 matrix together with ilmenite, apatite, tourmaline, zircon and monazite. Locally, biotite shows
247 resorbed outlines and is associated to films of K-feldspar and quartz. Corroded muscovite may also
248 occur. The garnet occurs as small (up to 1mm), subhedral crystals with abundant FI and NI (Fig. 3b,c),
249 along with quartz, graphite, biotite and sillimanite. No significant zoning is observed in terms of major
250 elements in the garnet, from rim ($\text{Alm}_{77}\text{Sp}_9\text{Gr}_6\text{Prp}_9$, $X_{\text{Mg}} \sim 0.11$) to core ($\text{Alm}_{75}\text{Sp}_6\text{Gr}_4\text{Prp}_{13}$, $X_{\text{Mg}} \sim$
251 0.15).

252 In the T zone, studied samples (STR28 and IVT21) are stromatic metatexites with variable
253 amount of discontinuous, cm-sized, leucosomes which may contain large garnet crystals (Fig. 2b). The
254 melanosome is strongly foliated (Fig. 3d), medium- to coarse-grained, composed of abundant biotite
255 ($X_{\text{Mg}} 0.5$; Ti 0.54 a.p.f.u.), garnet, fibrolitic to prismatic sillimanite, K-feldspar, plagioclase and quartz.
256 Muscovite is not present. Evidence for partial melting are resorbed biotite and sillimanite associated
257 with films of K-feldspar and quartz (Fig. 3e). Accessory phases are graphite, ilmenite, apatite, zircon
258 and monazite. The garnet crystals are much larger (<1 cm) than in UA zone, and contain clusters of FI
259 and NI (Fig. 3d and f) in their core, together with quartz and graphite. From core ($\text{Alm}_{70}\text{Sp}_3\text{Gr}_4\text{Prp}_{22}$;
260 $X_{\text{Mg}} \sim 0.24$) to rim ($\text{Alm}_{78}\text{Sp}_4\text{Gr}_3\text{Prp}_{14}$; $X_{\text{Mg}} \sim 0.2$), the garnet is slightly more almandine-rich.

261 In the G zone, the studied metapelites are residual diatexites (STR22 and STR69), in which
262 the melanosome is mainly composed of coarse-grained garnet (0.5-1.2 cm) and prismatic sillimanite
263 (Fig. 2c and 3g), and also plagioclase, K-feldspar, quartz and subordinated biotite ($X_{\text{Mg}} 0.53$; Ti 0.6
264 a.p.f.u.). Accessories in this case are graphite (relatively less abundant than in the samples from upper
265 amphibolite and transition zones), apatite, zircon, monazite and rutile. Garnet occurs as elongate
266 porphyroblasts with variable amounts of inclusions. FI and NI inclusions occur mostly in the cores
267 (Fig. 3g) together with biotite, sillimanite, quartz, rutile and graphite, however these other phases may

268 also be at rims. No significant zoning was observed from core ($\text{Alm}_{68}\text{Sp}_1\text{Gr}_5\text{Prp}_{26}$; $X_{\text{Mg}}\sim 0.28$) to rim
269 ($\text{Alm}_{72}\text{Sp}_2\text{Gr}_3\text{Prp}_{21}$; $X_{\text{Mg}}\sim 0.23$).

270

271 *4.2 Microstructural characterization of the inclusions*

272 Garnet crystals from the three zones contain abundant inclusions and, as mentioned above, the
273 characterization of the phases inside them was done by BSE imaging, EDS analysis and micro-Raman
274 spectroscopy.

275 Petrography indicates the presence of two types of primary multiphase inclusions in the same
276 clusters (Fig. 3b, c and f), regularly distributed in the cores of the garnet: one darker, with very high
277 birefringence at cross-polarized light and another light-colored to transparent (Fig. 3f), with relatively
278 lower birefringence (Fig. 3c). These are respectively FI and NI. Both types of inclusions have similar
279 size (mostly 2-10 μm) and usually negative crystal shapes (Fig. 3f and 4a-e). Very few of them may
280 show evidence of decrepitation such as small tails and microcracks (Fig. 4f).

281 Nanogranitoids contain aggregates of K-feldspar, plagioclase, quartz, biotite, muscovite,
282 chlorite (Fig. 4a-d), and in some cases calcite (Fig. 4e). Micro-Raman investigation has indicated the
283 presence of polymorphs such as kumdykolite ($\text{NaAlSi}_3\text{O}_8$), cristobalite, trydimite (Fig. 5a-c), and, in
284 one inclusion, kokchetavite (KAlSi_3O_8). Furthermore, a few inclusions may contain CH_4 and N_2 (Fig.
285 5d). Concerning the minerals crystallized from the melt, no variation in mineral assemblage was
286 observed in the samples from the three zones. Common trapped minerals are graphite, apatite, zircon,
287 and ilmenite, whereas rutile is observed frequently in inclusions from G, and rarely from T zone.
288 Glassy inclusions or residual glass in NI were not detected.

289 The FI, darker under transmitted light (Fig. 6a), have very high birefringence under cross
290 polarizers (Fig. 6b), and are composed of fluid and solid phases; the latter comprise both daughter and
291 trapped minerals. These FI are more abundant in the UA metatexites suggesting that the fluid/melt
292 ratio is higher in these rocks, when compared to the T zone and G rocks. Micro-Raman has shown
293 variable proportions of CO_2 , CH_4 and N_2 (Fig. 5e,f); no H_2O was detected. Amongst the most common
294 phases identified in the inclusions are siderite, pyrophyllite (Fig. 6c) and kaolinite (Fig. 5f), and in a

295 few cases calcite, magnesite and graphite. Rutile was observed in the samples from G and likely
296 represents a trapped phase, since some terminations of rutile grains are partially enclosed in the host
297 garnet. The FI throughout the three zones have variable compositions and densities, and different
298 proportions of fluid phases are sometimes observed in the same sample. The relative amounts of
299 components in the fluid were calculated using the method by Dubessy, Poty, & Ramboz (1989). For
300 instance, at UA CO₂ (absent to 94 mol.%), N₂ (22 to 56 mol.%) and CH₄ (absent to 12 mol.%), at T,
301 CO₂ (absent to 92 mol.%), N₂ (5 to 95 mol.%) and CH₄ (absent to 70 mol.%), and at G, CO₂ (absent to
302 86 mol.%), N₂ (18 to 23 mol.%) and CH₄ (absent to 76 mol.%). The CO₂ density of the inclusions was
303 calculated using the method based on the measurement of the Raman intensity of the Fermi diad
304 (Wang et al., 2011) and vary from 0.1 to 0.6 g/cm³. These variable compositions and densities are
305 interpreted as result of the interaction of fluid with the host during the cooling path of the migmatites
306 (e.g. Kleinfeld & Bakker, 2002; Tacchetto et al., 2018; see below).

307

308 *4.3 Microstructures of inclusions after re-melting experiments*

309 The conditions for the re-melting experiments of samples from the three zones were chosen
310 following the phase equilibria modelling from the literature (Redler et al., 2012). As expected all
311 successful experiments showed the presence of homogeneous glass (i.e. melt; Fig. 7a-d). Together
312 with the glass, some inclusions have shown the presence of graphite (Fig. 7e), which represents the
313 most commonly observed phase in the studied samples, and in one sample, euhedral plagioclase (Fig.
314 7f) was also present. These phases were observed in inclusions before the experiments and are
315 interpreted as trapped phases inside the melt inclusions, i.e. they were present together with the melt
316 during entrapment (e.g. Acosta-Vigil et al., 2016). Bubbles may also be observed together with glass
317 (Fig. 7g,h), and may result from incomplete dissolution of the fluid into the melt during the
318 experiment, or may be shrinkage bubbles (Lowenstern, 1995).

319 Incomplete re-melting of some inclusions was also observed, and in that case corroded biotite
320 occurs together with rounded crystals of quartz and glass (Fig. 7i). Interaction with the host was also
321 observed in some cases, as suggested by irregular boundaries of remelted inclusions, formation of new

322 phases such as orthopyroxene (Fig. 7j) and the host garnet showing a different composition in
323 proximity of remelted inclusion (see contrasting shade of grey in the BSE, Fig. 7j).

324

325 *4.4 Composition of anatectic melts*

326 *Major elements and CIPW normative diagrams*

327 Approximately 130 EMP analyses were performed on the remelted MI. Analyses that have
328 clearly shown evidence of interaction/contamination with the host were disregarded. Composition of
329 the preserved inclusions were recalculated to an anhydrous basis and are described below (see Fig. 8
330 and Table 2).

331 The data show that all the melts in the three zones are granitic *s.l.*, peraluminous (Fig. 8a) and
332 have similar ranges of silica (70-78 wt. %), Na₂O (2-4 wt. %) and Al₂O₃ (12.5-16.5 wt. %) in most
333 cases. However, slightly differences in compositions can be noticed. In samples from the UA, melts
334 have restricted K₂O (3.6-5.1 wt. %; Fig 8b), lower Na₂O/K₂O (0.4-0.9), and CaO (0.4-1.2 wt. %, Fig.
335 8c), are peraluminous (ASI ~ 1.1-1.5) and less mafic (FeO+MgO 2-3.4 wt. %; Fig 8d). The melts from
336 T have more spread compositions, are more peraluminous (ASI ~ 1.1-1.8), have higher Na₂O/K₂O
337 (0.1-1.2), and maficity (FeO+MgO 1.2- 3.9 wt. %). The melts at G are peraluminous (ASI ~ 1.2-1.3),
338 have the lowest K₂O (1.8 to 2.5 wt. %) and reach highest Na₂O/K₂O (1.2-1.9), CaO (1.4-2.5 wt. %)
339 and FeO+MgO (3.2-4.7 wt. %).

340 In the CIPW-normative Ab-An-Or diagram (Fig. 9a), the MI form two distinct groups, one
341 including MI from UA and T, which are classified as granite, but show variable Ab/Or ratios and a
342 second, more restricted group, represented by the MI from G which have lower Or contents and higher
343 An contents, and are classified as granodiorites. When compared to other NI from the literature, the
344 MI from UA and T have relatively higher An contents. In turn, the MI from G plot in the same field as
345 MI from Kali Gandaki (Nepal) and Jubrique (Spain).

346 In the haplogranite system (Qz-Ab-Or, Fig. 9b), the MI also form two distinct groups. The MI
347 from G have lower Or, and comparable compositions with MI from Kali Gandaki and Jubrique. The
348 MI from UA and T display variable compositions, and are mostly similar to Ojen metatexites and
349 diatexites (Spain). The spread of composition of MI from the three zones is mostly parallel to the Qz-

350 Ab sideline, and could be the effect of sluggish diffusion of Si and Al versus rapid diffusion of alkalis
351 in the granitic melt (e.g. Acosta Vigil, London, & Morgan, 2006).

352

353 *H₂O and CO₂ contents of the melts*

354 The H and C concentrations, then recalculated as H₂O and CO₂, were measured on remelted
355 inclusions from three zones (STR36 from UA, STR28 and IVT21 from T, STR22 from G); results are
356 given in Table 3.

357 MI from all three zones span a wide range of concentrations of CO₂ (Fig. 9). The sample from
358 UA varies from 861 to 1738 ppm (average 1384 ± 373 ppm), in which the lowest values were
359 observed in an inclusion of glass coexisting with a bubble and the highest values were measured in
360 homogeneous glass. Samples have the lowest contents of CO₂ in T, vary from 495 to 1165 ppm
361 (IVT21) and 573 to 1534 (STR28), and have much lower averages (IVT21, 884 ± 267 ppm; STR28,
362 835 ± 329 ppm). In this case, both the lowest and highest values are not easily correlated to the
363 presence of bubbles or crystals. Sample STR22 from G has two contrasting signatures, one similar to
364 the previously described for T (BC8 from 739 to 901 ppm, average 833 ± 61 ppm), and another that
365 displays the highest CO₂ values (BC10 from 1354 to 2444, average 1788 ± 395 ppm). The highest
366 values occur when the glass coexists with graphite, whereas the lowest values occur in inclusions with
367 homogeneous glass.

368 The highest H₂O contents were measured on the sample from UA (10 wt.%), and the lowest in
369 the sample from T (STR28 4.8 wt.%). Average H₂O contents are progressively lower with increasing
370 metamorphic grade: at UA values are slightly higher (8.1 ± 1.4 wt.%), at T, intermediate (7 ± 0.9
371 wt.%) and at G, lower (6.8 ± 0.7 wt.%). However, the ranges of the three zones are relatively similar
372 (UA 6.5 to 10 wt.%; T 4.8 to 8.5 wt. %; G 5.4 to 8.2 wt.%; see Fig. 9).

373

374 **6. Discussion**

375 *6.1 Significance of melt and fluid inclusions*

376 Our study of inclusions of the metapelitic migmatites from the Val Strona di Omegna (IZ) has
377 shown the coexistence of NI and FI inclusions in clusters regularly distributed in the cores of the
378 garnet (Fig. 3). Such microstructural evidence suggests a primary coeval entrapment during the growth
379 of peritectic garnet (Roedder, 1979; Cesare et al., 2015), i.e. these two different types of inclusions are
380 related to the same anatectic event.

381 Considering the mineral assemblages, their behavior during experimental remelting and their
382 chemical composition, NI can be reliably interpreted as former droplets of felsic silicate melt. The
383 presence of polymorphs such as kumdykolite, cristobalite, trydimite and kokchetavite has been
384 previously reported as evidence for the preservation of the original composition of the inclusions (e.g.
385 Ferrero et al., 2016a; see also Ferrero & Angel, 2018) including the volatiles such as H₂O and CO₂.
386 Indeed, these polymorphs are highly susceptible to transformation into their thermodynamically stable
387 phases as result of inclusion decrepitation. Therefore, their occurrence in the investigated NI from Val
388 Strona di Omegna suggests that the inclusions represent a reliable source of information on the starting
389 composition of the anatectic melts and fluid regime of the IZ.

390 Fluid inclusions described in this study are composed of variable proportions of CO₂, N₂ and
391 CH₄, along with siderite, pyrophyllite and kaolinite as daughter phases. This strongly indicates that,
392 although the initial composition was not determined here, the fluid present during anatexis had a
393 COHN composition. The variable compositions and densities of the FI are interpreted as result of
394 post-entrapment re-speciation and reaction between host garnet and the fluid during the cooling path
395 of the migmatites (e.g. Kleinfeld & Bakker, 2002; Ferrero et al., 2014; Tacchetto et al., 2018), which
396 resulted in the crystallization of daughter phases. A possible reaction in the Fe end-member system
397 would be:

398



400

401 Primary in origin, the investigated NI and FI represent, therefore, snapshots of coexisting
402 anatectic melt and C-bearing fluid which were present during partial melting of this section of deep
403 continental crust; i.e., they are indicative of a situation of immiscibility between a carbonic fluid and a

404 crustal melt which may result from the low solubility of carbonic fluid in granitic melts (e.g. Cesare
405 and Maineri, 1999; Tamic, Behrens, & Holtz, 2001).

406 The possibility for peritectic garnet to trap droplets of coexisting melt is highlighted by phase
407 equilibria modelling of a probable protolith composition in which garnet and melt modes rapidly
408 increase after crossing the solidus (Fig. 11a). In a situation of fluid-melt immiscibility, the growing
409 garnet would be obviously able to trap both melt and fluid.

410 The phase diagram of Figure 11a was constructed without considering the presence of a COH
411 fluid. Despite the recent advances in the calculation of partial melting equilibria (White et al., 2011;
412 Palin et al., 2016; White, Palin, & Green, 2017), the suprasolidus behaviour of graphitic systems
413 cannot be adequately modelled owing to the lack of a melt model that considers the occurrence of
414 carbonic species (both CO₂ and CH₄) (see discussion in Bartoli et al., 2016). In this sense, NI and FI in
415 peritectic minerals represent a unique natural laboratory to recover melting mechanisms in deep
416 crustal rocks.

417

418 *6.2 New clues on anatexis of the Ivrea Zone*

419 The Ivrea Zone (NW Italy) is a world-renowned Permian mid to lower crustal section. Crustal
420 rocks have been widely investigated through all possible approaches such as field observations (e.g.,
421 Zingg, 1980; Quick et al., 2003; Redler et al., 2012, 2013), phase relationships (e.g., Schmid & Wood,
422 1976), fluid inclusions (De Negri & Touret, 1978), whole rock geochemistry (major and trace
423 elements and isotopes; e.g., Sighinolfi & Gorgoni, 1978; Baker, 1988; Schnetger, 1994; Henk et al.,
424 1997; Barboza et al., 1999; Bea & Montero, 1999; Alessio et al., 2018), conventional
425 thermobarometry (e.g., Schmid & Wood, 1976; Zingg, 1980; Sills, 1984; Henk et al., 1997; Demarchi
426 et al., 1998; Bea & Montero, 1999; Barboza & Bergants, 2000), geochronology (Köppel, 1974; Vavra
427 et al., 1999; Ewing et al., 2013, 2015; Klötzli et al., 2014; Kunz et al., 2018a, b), trace element
428 thermometry (Luvizotto & Zack, 2009; Ewing et al., 2013; Kunz et al., 2018) and modern phase
429 equilibria modelling (Redler et al., 2012, 2013; Bartoli, 2018). Despite the impressive number of
430 studies devoted to reconstruct the high-temperature and anatexis history of this deep crustal section,

431 the first and clear evidence of carbonic fluid-present melting in the Ivrea Zone is reported only in this
432 study.

433 There is some discrepancy among published P – T estimates for the rocks in Val Strona di
434 Omegna (Fig. 1b). Successful remelting experiments of nanogranioids may provide an additional and
435 realistic constraint on the conditions, in particular the temperature, at which melt was present in the
436 system; i.e., at which rocks were partially melted (Bartoli et al., 2013b; Bartoli, Tajcmanová, Cesare,
437 & Acosta-Vigil, 2013c; Ferrero et al., 2015). Experimental remelting performed in this study indicates
438 that garnet grew and trapped melt inclusions between 820 and 900 °C, proving that UHT conditions
439 were approached in this section on continental crust and confirming the estimates based on trace
440 element thermometry and phase equilibria calculations (Redler et al., 2012; Luvizzotto & Zack, 2009;
441 Ewing et al., 2013). Conversely, conventional exchange and net transfer thermobarometry gave \approx 130–
442 170 °C lower and \approx 2–4 kbar lower values (Fig. 1b; see above). This discrepancy is likely to be related
443 to a significant retrograde re-equilibration of mineral compositions (Pattison et al., 2003; Redler et al.,
444 2012).

445 According to the literature, the rocks from IZ have experienced anatexis through fluid-absent
446 breakdown of muscovite and biotite, and extensive partial melting at the highest temperatures (up to
447 40–50 vol.% melt at 850–900 °C) (Fig. 11b; see also Schmid & Wood, 1976; Schnetger, 1994; Sinigoi
448 et al., 1994). In this study, we have shown that the microstructural evidences for anatexis include the
449 presence of corroded muscovite (restricted to some samples from amphibolite facies), but more
450 frequently biotite with resorbed outlines associated with films of K-feldspar and quartz, and NI in
451 peritectic garnet (e.g. Cesare et al., 2015), which are found from UA to G (e.g. all samples in Fig. 1b).
452 Thus, incongruent melting of biotite in the metapelitic migmatites and the formation of peritectic
453 garnet may have started before the muscovite-out isograd (sample 77, Fig. 1b). Our phase
454 equilibrium modelling (Fig. 11a) shows that continuous melting of biotite $<$ 800°C produces garnet and
455 melt. Another possibility could be that the breakdown of muscovite may also have produced small
456 proportions of garnet with the melt, similar to the migmatites from Ronda (Bartoli, Tajcmanová,
457 Cesare, & Acosta-Vigil, 2013c). Moreover, the primary microstructural arrangement of NI and FI
458 suggests that anatexis of these metapelites occurred through breakdown melting of muscovite and

459 biotite but always in the presence of a COH fluid, as expected from theoretical considerations on the
460 behavior of graphitic systems (see discussion for the origin of the fluid below).

461 The composition of the melts from UA and T are similar and consistent with melting of
462 biotite, in agreement with microstructural observations and phase equilibria constraints (Figs. 3 and
463 11). On the other hand, the melts from G display higher contents of CaO, lower K₂O (and
464 consequently higher normative An and lower Or contents) and higher FeO+MgO (Figs. 8, 9).

465 Most of the MI analysed in this study have FeO+MgO contents <3 wt.% similar to other MI
466 from the literature (see Cesare et al., 2015). However, 40% of MI from the T and all from G have
467 FeO+MgO from 3 to 4.7 wt.%. Same ranges are observed in MI from terranes where anatexis occurred
468 at 850-900°C (e.g. El Hoyazo, Bohemian Massif and Kerala Khondalite belt). This is consistent with
469 experimental works which suggest that temperature controls the FeO+MgO and CaO of the melts
470 (Johannes & Holtz, 1996; Montel & Vielzeuf, 1997; Gao et al., 2016).

471 Higher CaO and FeO+MgO in MI might indicate that melts have interacted with the host, and
472 therefore they could not represent pristine compositions. These cases can be detected if the host garnet
473 has higher contents of Ca and positive correlation of FeO+MgO and ASI in the MI is observed (e.g.
474 Cesare et al., 2015). However, contents of Ca in the garnet are virtually the same in the three zones
475 (see Table 1), furthermore, correlation between FeO+MgO and ASI does not occur. Therefore,
476 compositions of remelted MI from G are interpreted to be original, and not related to interaction
477 between melt and host. The contrasts in the compositions of the MI from UA facies and T versus G
478 can be related to several additional factors, we discuss some of them below.

479 All remelted inclusions from this study have higher normative An contents than most MI from
480 the literature (see Fig. 9a). The highest An contents are observed in the melts from G, which are
481 compositionally granodiorites, and plot together with MI from Kali Gandaki (Nepal) and Jubrique
482 (Spain). More calcic compositions of crustal melts have previously been interpreted as a result of H₂O
483 present-melting (Patiño Douce & Harris, 1998; Frost & Frost, 2008), and likewise these two
484 occurrences mentioned above were interpreted to have been formed by fluid present melting
485 (T<800°C) (Carosi et al., 2015; Acosta-Vigil et al., 2016). This is coherent in the case of MI from
486 Jubrique, which have high H₂O contents (7 to 13 wt.%) and low FeO+MgO (Acosta-Vigil et al.,

487 2016). However, the MI from G are unlikely to have been produced under similar conditions. Indeed,
488 the H₂O contents of melts are lower despite the higher P of formation, and there is no evidence, in
489 terms of FI, of the presence of free water at suprasolidus conditions.

490 Garcia-Arias (2018) recently showed a mismatch between the Ca contents and maficity of
491 anatectic melts and S-type granites, and concluded that preferential non-stoichiometric entrainment of
492 plagioclase could be a viable process to increase the Ca of granitic magmas. However, our data,
493 coming from MI that cannot have been influenced by entrainment, show that under some
494 circumstances, anatexis may produce primary melts with higher Ca and maficity.

495 Alternatively, Ca-rich compositions could result from changes in the reactants, for example, a
496 greater contribution of (Ca-rich) plagioclase at granulite facies. A similar scenario, in which rims of
497 plagioclase may melt congruently producing a more Ca-rich melt, was considered by Taylor et al.
498 (2014) in order to explain composition of the K-poor, Ca-rich leucosomes of the Southern Marginal
499 Zone migmatites, Limpopo Belt, South Africa. Thus, we believe that the MI from granulite facies may
500 represent an additional evidence for the origin of Pl-rich leucosomes usually found in granulite
501 terranes (e.g. Sawyer et al., 1999; Taylor & Stevens, 2010). However, detailed study of the
502 microstructure of leucosomes is necessary in order to evaluate if the low K and high Ca was not
503 produced by cooling driven processes, for instance, framework of plagioclase may indicate loss of
504 interstitial (fractionated) melt (e.g. Carvalho et al., 2016).

505 The breakdown of apatite (and monazite) during suprasolidus metamorphism of metapelites is
506 the main supplier of phosphorus, as well as LREE, to the anatectic melt (see Yakymchuk, 2017 for a
507 review). Due to the high contents of Ca in apatite (~55 wt. % CaO), its dissolution could also have a
508 influence on the contents of Ca of the melts. A detailed study of the accessory phases (monazite,
509 zircon, apatite and xenotime) in the metapelitic migmatites from Val Strona di Omega (Bea &
510 Montero, 1999) showed that the grain size and modal abundance of apatite “decreases dramatically” in
511 the rocks from granulite facies (so called stronalites by those authors; see their Table 1 and Figure 4),
512 whereas in rocks from amphibolite and transition zones the grains are large and fairly abundant. Those
513 authors interpreted this as result of elevated solubility of apatite in peraluminous melts. Thus, apatite
514 may have played a (restricted) role and contributed to the Ca-budget (together with P₂O₅ and LREE)

515 of the melts at G. Additional information on the trace elements and P₂O₅ of the melts (which were not
516 analysed in this study) are required to further investigate this hypothesis.

517 Ca-rich and K-poor crustal melts were documented as leucosomes in the Kinzigite Formation
518 of IZ. However, these rocks are restricted to the 2–3 km wide aureole around the Mafic Complex and
519 are in textural equilibrium with cordierite and hercynitic spinel (Barboza et al., 1999; Barboza &
520 Bergants, 2000). Instead, the investigated NI clearly coexist with peritectic garnet and come from
521 metapelitic migmatites of Val Strona di Omegna which lack any clear evidence for a metamorphic
522 overprint caused by the Mafic Complex (Redler et al., 2012). Therefore, the K-poor melt inclusions
523 described in our study could likely represent crustal melts produced during HT-UHT regional
524 metamorphism.

525

526 *6.3 Immiscible carbonic fluids and the origin of CO₂-bearing fluid*

527 We have shown that throughout the investigated anatectic rocks of the Ivrea Zone, peritectic
528 garnet contains FI coexisting with NI, where the fluid is dominated by CO₂, CH₄ and N₂, without
529 detectable H₂O. As suggested above, the original composition and density of such immiscible fluid
530 within FI has been modified by both interaction with the host (e.g., Tacchetto et al., 2018) and re-
531 speciation (e.g., Cesare, 1995) during cooling, leading to the formation of carbonates, graphite and
532 phyllosilicates. Regardless of the actual fluid composition at the time of entrapment - which is beyond
533 the scope of this research - a carbonic component must have been present (and abundant).

534 In addition, the contents of CO₂ (500 to 2500 ppm) found in the remelted inclusions from IZ
535 are in agreement with the experiments of Tamic et al. (2001), and also represent a strong indication of
536 the presence of a COH fluid during anatexis of the studied terrane. A similar scenario has been
537 previously described by Cesare et al. (2007) in graphitic enclaves from El Hoyazo, where the fluid
538 was considered internally derived. Another example of immiscibility was reported by Ferrero et al.
539 (2014), however this case the fluid was interpreted to have derived from the crystallization of
540 mantellic magmas (e.g. Touret, 1992).

541 The occurrence of CO₂-rich fluid inclusions in granulites is not an uncommon feature and the
542 origins of the fluid, if externally or internally derived, have been described in several works (e.g.
543 Huizenga & Touret, 2012, and references therein).

544 Frequently proposed sources of CO₂ in granulitic terranes are the degassing mantle and the
545 crystallizing mantle-derived magmas which intruded the lower crust; i.e., externally derived CO₂
546 (Newton, 1980; Jackson, Matthey, & Harris, 1988; Touret, 1992; Santosh & Omori, 2008). In the IZ
547 mafic magmas and deep crustal rocks coexist and the emplacement and crystallization of the Mafic
548 Complex could have promoted the CO₂ flushing into the overlying Kinzigite Formation. However, the
549 container of NI and FI (the peritectic garnet) is thought to represent part of the mineral assemblage
550 related to the regional metamorphism whose thermal peak predates the emplacement of mafic magmas
551 (Barboza et al., 1999; Barboza & Bergantz, 2000).

552 One could argue that the relationship between Mafic Complex and high-grade metamorphism
553 of the Kinzigite Formation has long been debated and that some doubts still exist (Sinigoi et al., 1991;
554 Henk et al., 1997; Vavra et al., 1999). Baker (1988) conducted an oxygen and carbon isotope study of
555 minerals and rocks from Val Strona di Omegna and concluded that there is no evidence for significant
556 transfer of CO₂-rich fluids from mantle to crust. Whatever the role of Mafic Complex is in the thermal
557 history of the Kinzigite Formation, the infiltration of CO₂ from crystallizing mafic magmas is,
558 therefore, highly improbable.

559 The metapelitic migmatites from IZ contain considerable amounts of graphite, therefore an
560 internal origin for, at least, part of the carbonic component in the fluid is expected. Such COH fluid is
561 initially produced by devolatilization of hydrous silicates (implied by the presence of graphite in the
562 protolith, e.g. Connolly & Cesare, 1993), before or during amphibolite-facies metamorphism,
563 explaining the higher fluid/melt in the rocks ratio from UA. With temperature increasing at
564 suprasolidus conditions, a possible additional internal source might be the Fe³⁺ reduction during biotite
565 melting accompanied by the oxidation of carbon and formation of CO₂ (Hollister, 1988; Cesare, Meli,
566 Nodari, & Russo, 2005). The role of decarbonation in the marbles intercalated in the Kinzigite
567 Formation cannot be completely excluded, however, it is probably unrelated (e.g. De Negri and
568 Touret, 1978).

570 *6.4 Implications for fluid regime of a high-grade terrane*

571 Fluid regime during high-temperature metamorphism and crustal anatexis has been part of a
572 heated ongoing debate in the literature. In the 1970s and early 1980s some seminal studies presented
573 the concepts of “dehydration melting” and “fluid-absent melting” (Eggler, 1973; Thompson & Tracy,
574 1979; Thompson, 1982). These two terms imply that the melting process and formation of anatectic
575 melt occur in absence of a fluid phase, through melting reactions involving the breakdown of hydrous
576 minerals such as micas and amphibole. Fluid-absent conditions have been long considered to prevail
577 during granulite-facies metamorphism, anatexis of the deep crust and formation of large volumes of
578 crustal magmas (e.g., Clemens and Vielzeuf, 1987; Clemens & Watkins, 2001; Brown, 2013).

579 This point of view has been recently challenged by some works, which have highlighted the
580 potential role of free H₂O and/or highly saline solutions (brines) during melting of the continental
581 crust: water-fluxed melting (Weinberg & Hasalova, 2015a) and brine-assisted melting (Aranovich et
582 al., 2014). Not surprisingly, these studies have added fuel to the fire. To have an idea of the heated
583 debate in the scientific community, the reader may refer to Clemens and Stevens (2015) vs. Weinberg
584 and Hasalova (2015b), and Aranovich Makhlof, Manning, Newton, & Touret (2016) vs. Clemens,
585 Buick, & Stevens (2016). It is important to highlight the fact that despite there seem to be two
586 opposite views on fluid regime during crustal melting, the fluid-present and -absent processes aren't
587 mutually exclusive, and it is quite likely that during natural anatexis a transition from fluid-present to
588 fluid-absent conditions often takes place (e.g., Acosta-Vigil et al., 2016). This view may hold in non-
589 graphitic protoliths (see below).

590 An older, but still ongoing, controversy on the fluid regime in the deep continental crust is
591 related to the existence of CO₂-rich fluids as well as their active role during high-grade
592 metamorphism. The concept of “carbonic metamorphism” was firstly proposed by Newton et al.
593 (1980). This theory derived from the observation of CO₂ fluid inclusions in many granulitic terranes
594 (e.g., Touret, 1971) and invokes the flushing of CO₂-rich fluids to generate dry mineral assemblages
595 typical of the granulitic crust. Despite the growing list of high-grade metamorphic terranes containing
596 CO₂ fluid inclusions (Santosh & Omori, 2008; Huizenga & Touret, 2012; Touret & Huizenga, 2012;

597 and references therein), late relative ages of some inclusions (Lamb, Valley, & Brown, 1987),
598 experimental constraints (Clemens, 1990) and the low mobility of CO₂ along grain boundaries
599 (Watson & Brenan, 1987) raised serious doubts for this model. For example, Stevens & Clemens
600 (1993) state that "...even when CO₂-rich fluid inclusions do record primary fluid compositions, this is
601 not necessarily evidence for fluid-present conditions. Rather, in anatectic rocks, these inclusions
602 represent the unreactive volatile dregs that would have occurred as high-dihedral angle bubbles in the
603 granulite-grade environment". Clemens et al. (2016) reaffirm the same concept: "the presence of
604 inclusions of such a fluid does not represent evidence for a truly fluid-present condition, as the rocks
605 in which they occur would have behaved as fluid-absent bulk systems".

606 Because the IZ experienced high-grade metamorphism, up to granulite-facies conditions, and
607 extraction of large volumes of crustal melt (reference here), it is considered an ideal natural laboratory
608 to study melting processes which led to crustal differentiation and S-type granite formation (e.g.,
609 Sighinolfi & Gorgoni, 1978; Schnetger, 1994; Bea & Montero, 1999; Ewing et al., 2014; Alessio et
610 al., 2018).

611 The CO₂-bearing inclusions investigated in this work are clearly primary in origin and related
612 to the prograde history of these rocks (see above). The coexistence of NI and FI in peritectic garnet,
613 their presence throughout the three zones and the measured CO₂ contents in NI unequivocally prove
614 that the fluid regime during high-temperature metamorphism and anatexis in this Permian crustal
615 section was characterized by the presence of a CO₂-bearing fluid phase. The fluid had progressively
616 lower aH₂O from UA to G. Consistently, the contents of H₂O decrease in the melt, whereas CO₂
617 increases from UA to G (see above). The H₂O contents of the melts throughout the three zones in IZ
618 indicate that aH₂O decreases with increasing metamorphic grade. AT UA, melts reach the highest
619 values (6.5 to 10 wt.%), and decreases through T (4.8 to 8.5 wt.%) to G (5.4 to 8.2 wt.%). The melts
620 are clearly H₂O-undersaturated, nevertheless, these values are slightly above the minimum water
621 contents estimated for haplogranite systems at similar P-T conditions (respectively, 5, 4.5 and 4 wt.%;
622 Holtz et al., 2001) which is usually assumed as initial H₂O contents of anatectic melts.

623 Although the study of nanogranitoids is still in its infancy (Cesare et al., 2015), the
624 coexistence of NI and carbonic FI has been already documented in other regional, granulite-facies

625 metamorphic terranes. Ferrero et al. (2016b) found silicate melt inclusions coexisting in the same
626 cluster with carbonatitic inclusions and COH fluid inclusions in garnet from Bohemian Massif,
627 indicating conditions of primary immiscibility between two melts and a fluid during the Variscan
628 anatexis. Instead, Tacchetto et al. (2018) proved the presence of a CO₂-rich fluid during the
629 Neoproterozoic anatexis of the continental lower crust exposed in the Athabasca granulite terrane
630 (Canada). Additional examples of C-bearing fluid-melt immiscibility during prograde melting are
631 documented in crustal enclaves from El Hoyazo (Spain) and La Galite (Tunisia) (Cesare et al., 2007;
632 Ferrero et al., 2011, 2014).

633 Note that in many of these occurrences (Ivrea zone, Athabasca granulite terrane and El
634 Hoyazo), plus several more unpublished (F. Ferri, unpublished data), graphite is present in the
635 metasedimentary protolith. Owing to the differential partitioning fluid species (mainly H₂O, CO₂ and
636 CH₄) between coexisting fluid and silicate melt, equilibrium thermodynamics impose that a rock
637 undergoing melting will always show the presence of a free COH fluid, in variable amounts and with
638 variable $a_{\text{H}_2\text{O}}$ depending on pressure, temperature and the bulk composition of the system (that can be
639 expressed also as f_{O_2} ; Connolly & Cesare, 1993). It follows that not surprisingly, graphitic
640 metasedimentary rocks behave in a fluid-present manner during anatexis, but that such fluid cannot be
641 pure H₂O but rather a COH fluid. Consequently, in the same rocks the source of carbon is, at least in
642 part, necessarily internal to the system, and that a possible mantle contribution, to be constrained by
643 isotopic means, may also be an additional carbon source.

644 The role of COH fluids during high-temperature metamorphism and formation of granitic
645 magmas has been largely discredited. Following Stevens & Clemens (1993), Clemens & Watkins
646 (2001) state that “it is perfectly possible to have fluid-absent partial melting in the presence of
647 graphite. For a treatment of this situation see Holloway et al., (1992)” and that “... graphite is unlikely
648 to be ubiquitous in crustal protoliths of granitoid magmas. At granulite-facies temperatures, it is only
649 likely to be stable in certain metapelitic rocks.”

650 However, the study of Holloway and coworkers finds little, if any, applicability to the
651 evolution of graphitic metapelitic systems which undergo breakdown of hydrous minerals. Holloway
652 et al. (1992) presented an experimental study considering a hydrogen-free basaltic system in which the

653 CO₂ is produced by reaction between the graphite capsule and the Fe₂O₃ in the melt. Clearly a topic
654 unrelated with the melting of metasediments in nature. Concerning the second issue highlighted by
655 Clemens & Watkins (2001) - the scarcity of graphitic granulites, - we think that such a statement is in
656 apparent contrast with evidence coming from graphitic granulite terranes worldwide, as attested by the
657 vastness of literature on this topic indexed in the major publication databases.

658 We conclude that carbonic fluid-present melting of the deep continental crust may be more
659 common than previously assumed and, together with breakdown melting reactions, it can represent an
660 important key process in the origin of crustal anatectic granitoids. It follows that the nature of fluid
661 regime during formation of granitic magmas may be variable and needs to be reevaluated. In order for a
662 reliable thermodynamic modelling to be possible, a more rigorous theoretical background and a
663 comprehensive experimental dataset on the suprasolidus partitioning of H₂O, CO₂ and CH₄ between
664 fluid and silicate melt are necessary and need to be implemented in the existing modelling softwares.

665

666 **8. Conclusions**

667 The study of nanogranitoid and fluid inclusions suggests that anatexis of the migmatitic
668 metapelites from Ivrea Zone occurred through muscovite and biotite breakdown melting in the
669 presence of a COH fluid. The fluid is assumed to be internally derived, produced initially by
670 devolatilization of hydrous silicates in the graphitic protolith, then as result of oxidation of carbon by
671 melting of Fe³⁺-bearing biotite.

672 Composition of the melts vary with increasing metamorphic grade, and are interpreted to
673 result from combination of higher T of melting, biotite melting in the three zones, but with greater
674 contribution of anhydrous reactants (such as plagioclase) and minor role of apatite at granulite facies.

675 The aH₂O of the fluid dissolved in granitic melts decreases with increasing metamorphic
676 grade. However, the H₂O contents of the melts throughout the three zones are comparable and slightly
677 above than the minimum H₂O contents assumed. The CO₂ contents are highest at granulite facies, and
678 show that carbon-contents of crustal magmas may not be completely negligible. Our data shows that

679 the thorough investigation of melt inclusions may provide significant new information of well-studied
680 terranes.

681

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693

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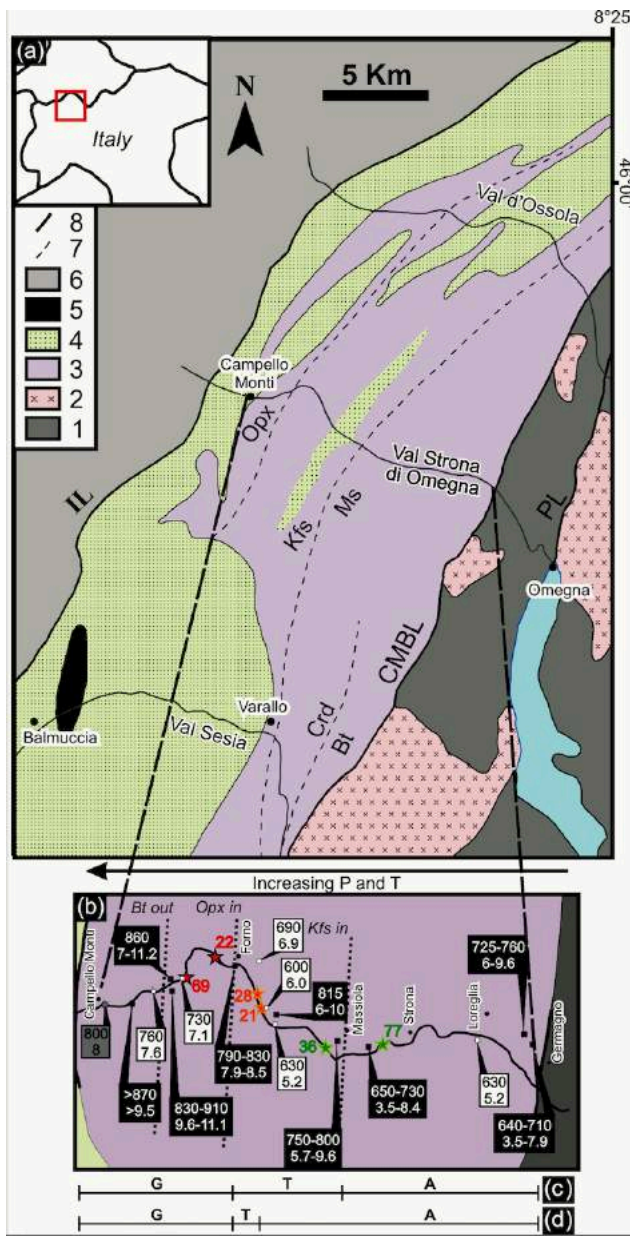
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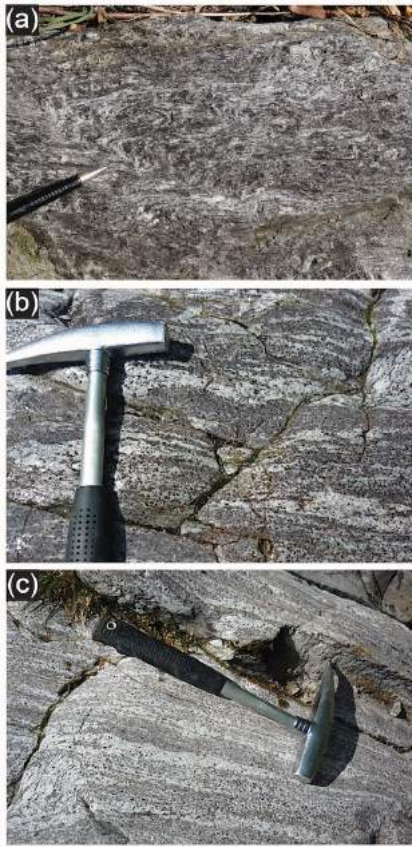
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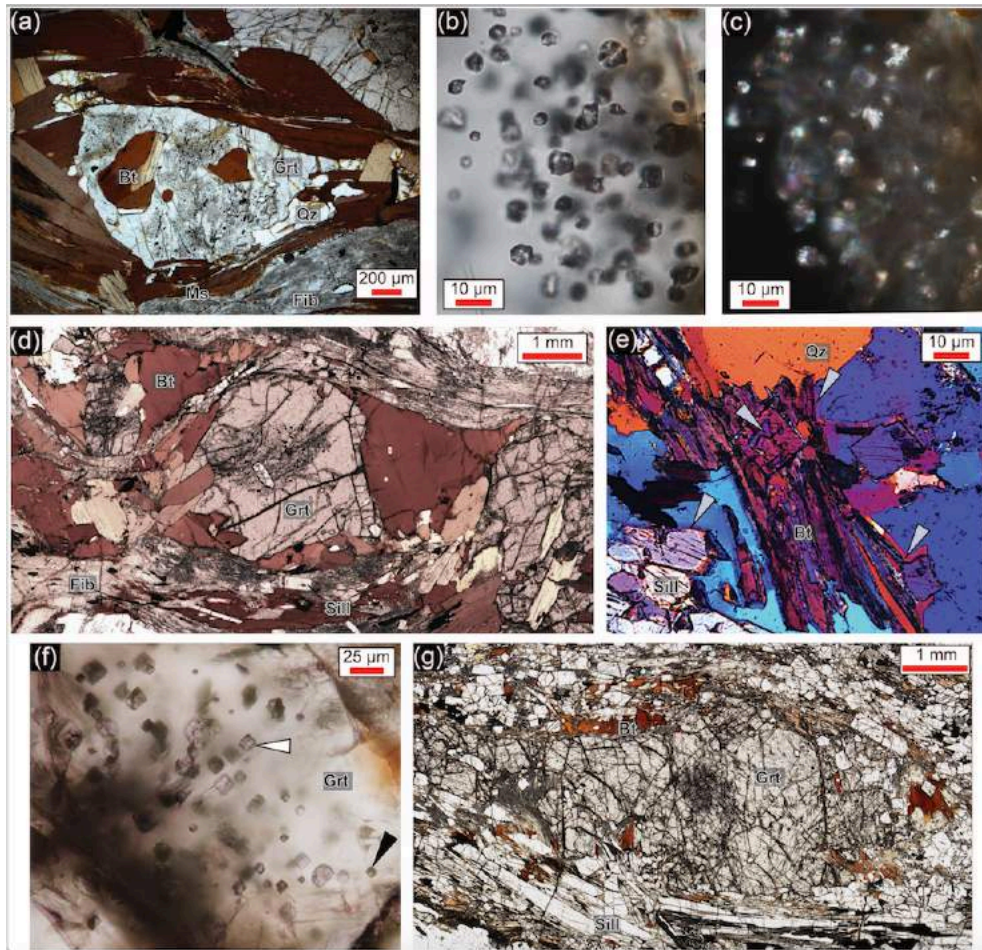


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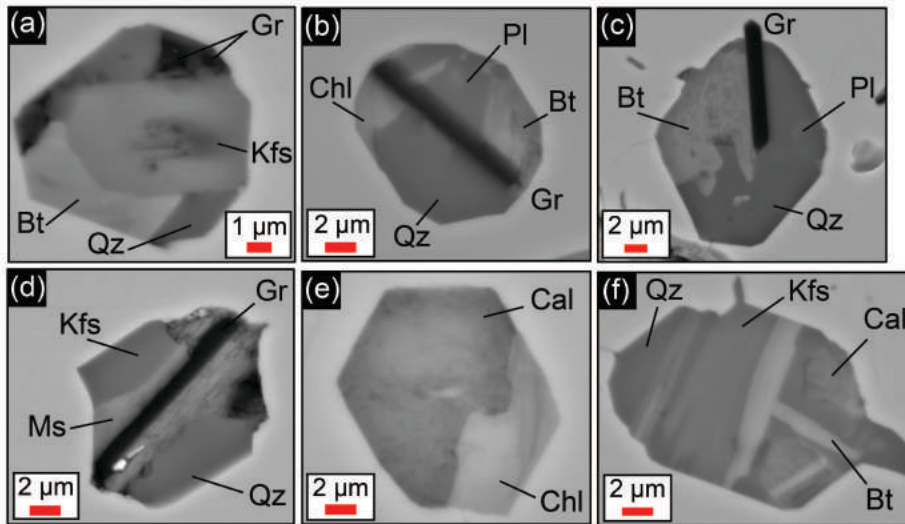


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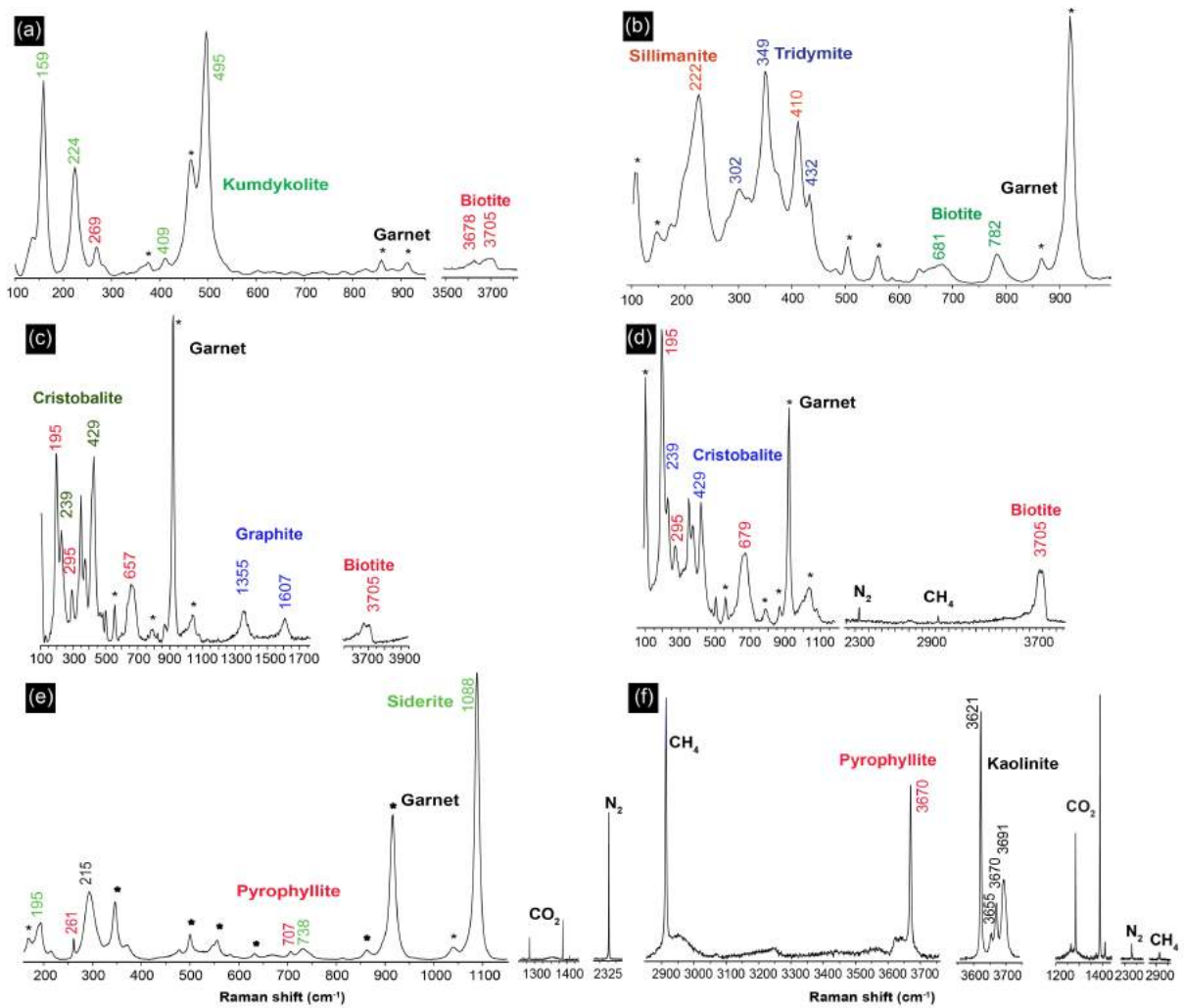
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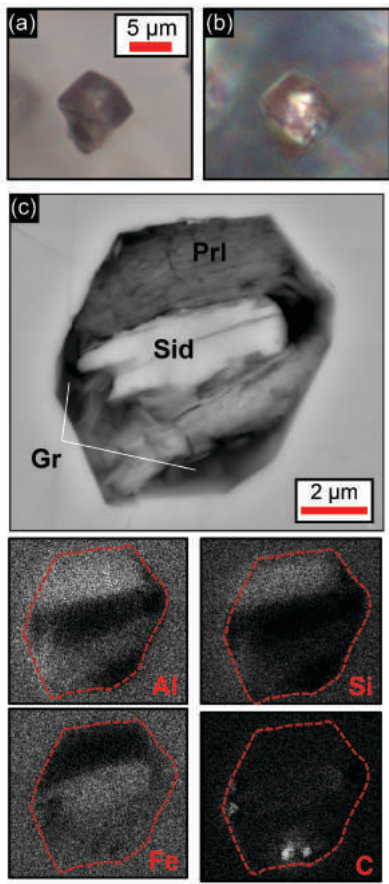
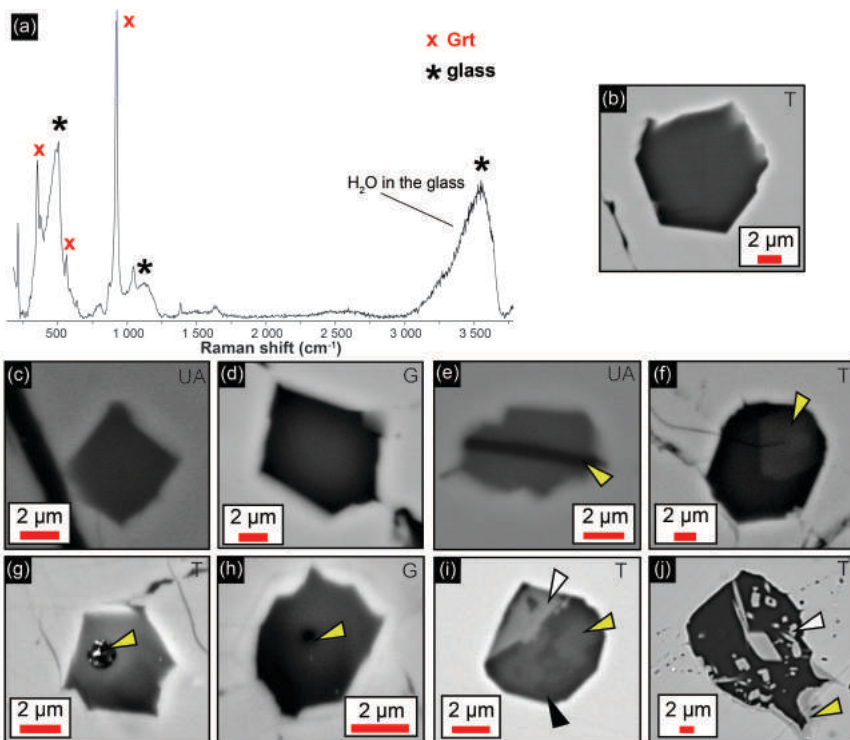
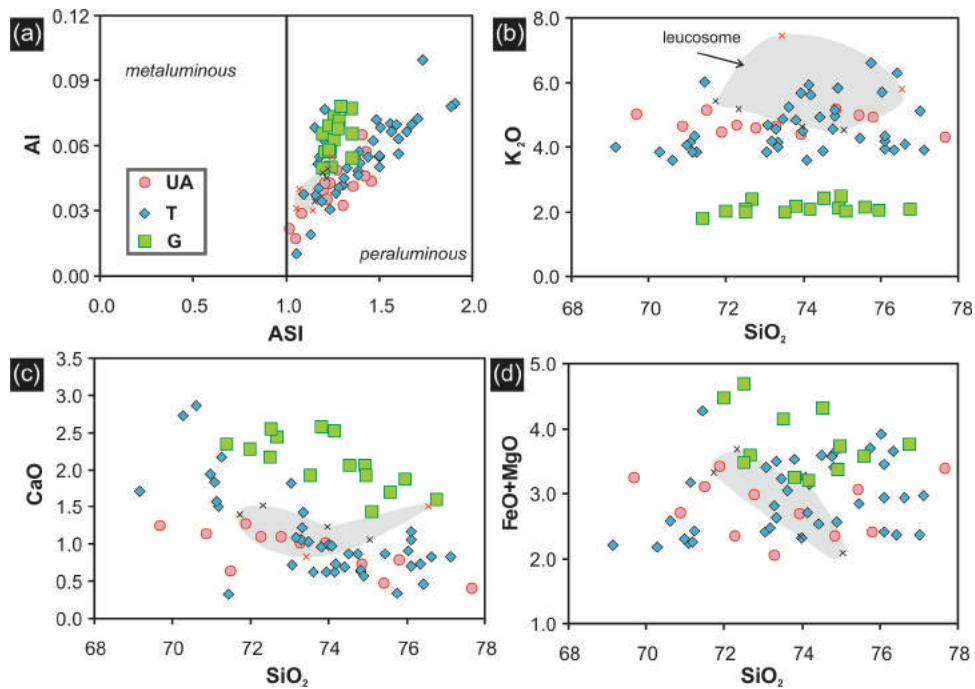


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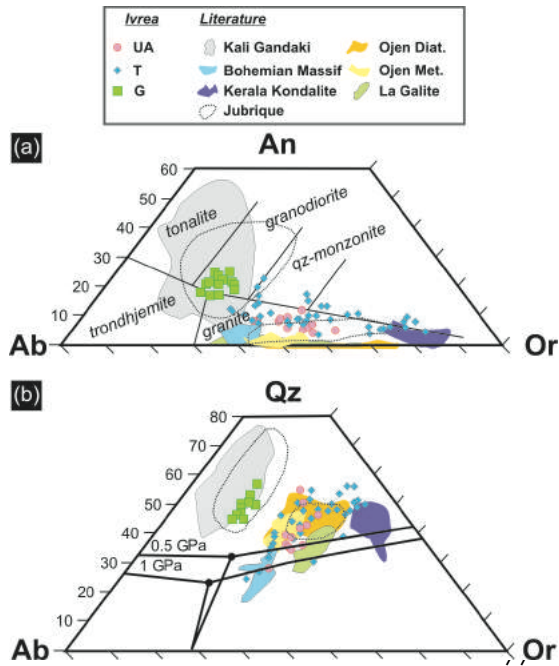


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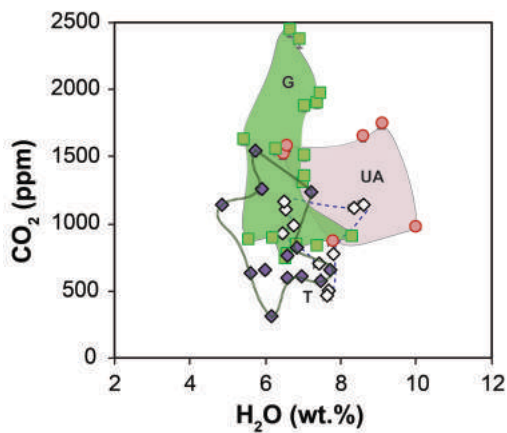
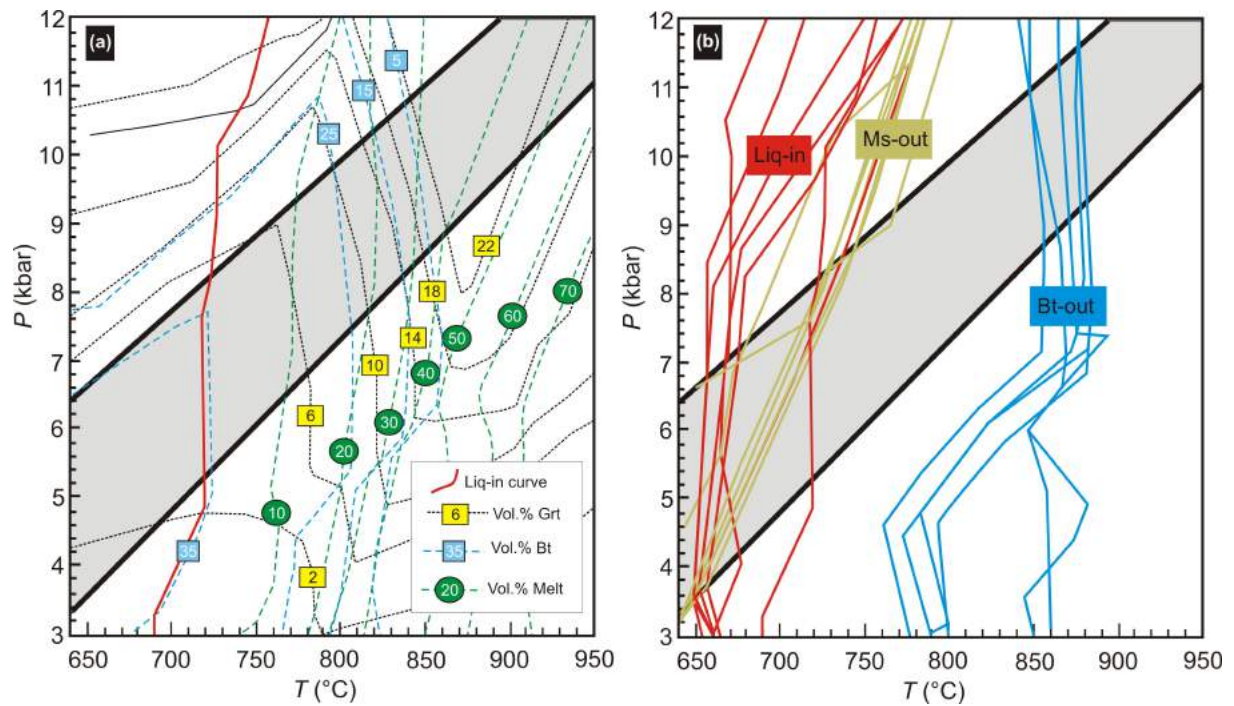


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