

# Skeletal quartz and dendritic biotite: Witnesses of primary disequilibrium growth textures in an alkali-feldspar granite

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1	Skeletal quartz and dendritic biotite: witnesses of primary disequilibrium
2	growth textures in an alkali-feldspar granite
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# 30 Highlights

- 31 Cathodoluminescence imaging reveals the presence of two generation of quartz
- 32 The first generation corresponds to both rounded and skeletal grains
- 33 The second generation (overgrowths) led to partly equilibrated texture
- 34 Biotite shows variable morphology, including skeletal and dendritic habits
- 35 These textures are interpreted to result from primary disequilibrium growth

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37

## 39 Abstract

40 We describe a case of S-type alkali-feldspar granite (quartz/K-feldspar/plagioclase/

41 biotite/garnet ~ 30/35/20/13/2 vol. %) showing skeletal and dendritic textures. It consists of

42 three nested units distinguishable by the size of biotite crystals: a first-emplaced unit

43 characterized by small biotite flakes (0.4–9 mm); a second unit with cm-sized biotite platelets

44 (0.3–28 mm) outlining a syn-magmatic foliation; and a third unit formed of large biotite

45 plates (0.7–850 mm) showing dendritic and radial growth patterns.

46 Quartz occurs mostly as rounded to euhedral grains, but also shows complex shapes 47 (serrated outline, re-entrants, etc.). Cathodoluminescence imaging reveals that grains consist 48 of both rounded and skeletal bluish cores (quartz-1) rimmed by reddish overgrowths (quartz-49 2) that lead locally to the coalescence of neighbouring grains and equilibrated texture. Biotite 50 is poikilitic enclosing dominantly quartz grains but also in lesser amount plagioclase and 51 garnet. In the third unit, biotite shows textures typical of dendritic growth (bifurcating, 52 radiating or branching structures, skeletal core). Plagioclase (An<25) occurs as small euhedral 53 to subhedral grains scattered in the matrix or forming glomerophyric aggregates. K-feldspar 54 (Or<sub>74–100</sub>) mostly forms an almost equilibrated mosaic of anhedral perthitic crystals; it also 55 appears in association with albite as a secondary phase replacing plagioclase. The 56 crystallization sequence started with garnet, then proceeded with quartz, plagioclase, biotite 57 and K-feldspar, ended with cordierite, and was followed by subsolidus reequilibration. 58 The variability in size and morphology of quartz and biotite is interpreted in terms of 59 variable undercooling depending on temperature of the host-rocks and on more or less intense 60 degassing of magma batches with variable melt water content. Here, rapid crystallization 61 prevented significant textural reequilibration and allowed the initial crystallization textures of 62 quartz to be preserved. Overall, this study shows that behind the common "coarse-grained" 63 texture of quartz in granite, subsist primary disequilibrium crystallization textures.

**Keyword**: texture; dendritic growth; granite; undercooling

## 67 **1. Introduction**

68 During their thermal evolution, igneous rocks tend to approach textural equilibrium to 69 minimize excess surface energy. This is more particularly visible in plutonic rocks owing to 70 mass redistribution within the crystal pile (e.g. Lesher and Walker, 1988), which leads to 71 textural coarsening (e.g. Higgins, 2011), a process well illustrated by the mafic layered 72 igneous complexes (e.g. Cawthorn, 1996). Even though the thermal history and related 73 textural maturity can, to some extent, be deciphered from the study of dihedral angles 74 (Holness, 2005; Holness et al., 2007a, 2007b, 2007c) or crystal size distribution (Cashman 75 and Marsh, 1988; Higgins, 2011; Magee et al., 2010; Marsh, 1988), studies of igneous rocks 76 are often facing with the issue to know to what extent the textures we observe are 77 representative of the initial stages of crystallization.

78 Experimental works led to a better understanding of crystal morphology in relation with 79 the dynamics of magma crystallization in rapidly cooled rocks (e.g. Lofgren, 1980). For 80 instance, shapes of plagioclase in basalts or of olivine in spinifex-textured komatiites, first 81 attributed to quenching, was shown to be related to cooling rates (Donaldson, 1976, 1982; 82 Lofgren, 1974; Lofgren & Donaldson, 1975). Later, Faure et al. (2003, 2006) determined 83 experimentally the respective role of undercooling, cooling rate and thermal gradient on the 84 shape and orientation of olivine crystals. Phosphorus zoning patterns in olivine from diverse 85 volcanic rocks and meteorites have also revealed that olivine phenocrysts are cored by 86 dendrites (Libourel et al., 2018; Milman-Barris et al., 2008; Welsch et al., 2013, 2014), a 87 characteristic attributed to primary variations in crystal growth rate (Milman-Barris et al., 88 2008). This observation has been extended to olivine from Mid-Atlantic Ridge gabbro and 89 from mafic-ultramafic intrusions (Welsch et al., 2014; Xing et al., 2017). This led Welsch and 90 co-workers to conclude that diffusion-controlled crystal growth is an important feature of 91 primary textures in igneous rocks. Others authors also emphasized that high supersaturation

and high growth rate are not uniquely driven by rapid cooling, but could also be enhanced by
shift in the liquidus temperatures related to change in melt water content (Donaldson, 1974;
Vernon, 1985; Almeev et al., 2007; Médard and Grove, 2008).

95 Concerning silicic plutonic rocks, crystallization experiments show that quartz exhibits 96 different morphologies as a function of the degree of undercooling (MacLellan and Trembath, 97 1991; Swanson and Fenn, 1986). At small  $\Delta T$ , quartz develops subhedral to euhedral shapes 98 by interface-controlled growth mechanisms; whereas at larger  $\Delta T$ , diffusion-controlled 99 growth mechanisms promote the formation of skeletal shapes in relation with compositional 100 gradient at the melt-crystal interface. Under longer experimental runs, skeletal quartz 101 becomes rounded by the reduction in the degree of faceting and the partial infilling of skeletal 102 crystals. Besides, preservation of primary compositional growth patterns in quartz has been 103 found in dacites and rhyolites (Ackerson et al., 2018; Ahmed, 2011; Götze et al., 2001; Seitz 104 et al., 2016; Watt et al., 1997); and disequilibrium growth textures (skeletal and dendritic) are 105 also known in some specific silicic plutonic rocks, such as pegmatites (e.g. Frindt and 106 Haapala, 2004; Haapala and Lukkari, 2005; Ivanov and Grozdanov, 2001; London, 2009; 107 Müller et al., 2002; Shannon et al., 1982; Swanson and Fenn, 1986) and orbicular granites 108 (e.g. Decitre et al., 2002; Smillie and Turnbull, 2014). 109 Overall, it can be expected that the common "coarse-grained" texture of granite might 110 conceal primary disequilibrium crystallization textures partly hidden (if not erased) by the 111 subsequent thermal evolution. The aim of this study is to address this issue from an 112 interesting case of alkali-feldspar granite. After description of phase assemblages and 113 chemical compositions, we focus on the characterization of textures to demonstrate that this 114 granite preserves primary disequilibrium growth textures recorded in the morphology of

115 quartz and biotite.

116

## 117 **2. Samples**

118 The granite studied here is located in the Central Vosges domain, eastern France (Fluck et al., 119 1991; Skrzypek et al., 2014). The outcrop, ca. 250 m long, is located along the road D42, 2 120 km to the NW of Eloyes ( $48^{\circ}06'21.50'' \text{ N} - 06^{\circ}35'30.50'' \text{ E}$ ). A cross-section (Fig. 1) shows 121 that the alkali-feldspar granite body forms the most part of the road cut, being bounded 122 (faulted contacts) to the NW by intrusive coarse-grained porphyritic granite (Remiremont 123 granite belonging to the Central Vosges granite association; Tabaud et al., 2015), and to the 124 SW by migmatitic biotite gneisses (metatexites to diatexites). Therefore, its exact extension 125 and geometry, seemingly a dike, remain unknown. A presentation of its geological context 126 and emplacement age is given in Supplementary material SM.1. 127 The granite corresponds to a pale pink rock, consisting of a fine- to medium-grained 128 quartzofeldspathic matrix with limited amounts of ferromagnesian minerals. Considering the 129 matrix grain size (average of ca. 0.25 mm), the rock appears to be at the limit between granite 130 and microgranite according to the grain size classification of Gillespie & Styles (1999). The 131 matrix consists of quartz (30-40 vol.%), K-feldspar (30-40 vol.%) and plagioclase (20-25 132 vol. %), whereas the ferromagnesian mineral phases consist of garnet ( $\leq 7$  vol. %), biotite (5– 133 15 vol. %) and scarce late-magmatic cordierite (± tourmaline). Accessory minerals are zircon, 134 monazite, ilmenite and Ti-oxide. Secondary phases are muscovite, albite and K-feldspar replacing primary feldspars; green biotite, chlorite, and hematite replacing garnet; and chlorite 135 136 and prehnite replacing biotite. The variable size of biotite led us to distinguish three magmatic 137 units (Fig. 1): 138 - Unit-1 forms the external parts of the igneous body. It contains medium-grained, 139 randomly distributed, biotite flakes outlining a faint syn-magmatic fabric (Fig. 2a, b). The

140 grain size of the matrix lies mostly between 0.3 and 0.6 mm, though ranging from 0.05 mm to

141 1.7 mm for the largest K-feldspar crystals (Fig. 2c).

142 - Unit-2 forms the core of the body. Biotite is mostly medium- to coarse-grained and 143 outlines a widespread syn-magmatic foliation (Fig. 3a-c), trending ENE-WSW, steeply 144 dipping (ca. 80°) and locally accompanied with a weak lineation. The grain size of the matrix 145 lies between 0.3 and 0.8 mm (Fig. 3d). Small mafic and autolithic enclaves (6-8 cm in 146 diameter), wrapped up by the foliation, are observed locally (Fig. 3c). 147 - Unit-3 corresponds chiefly to a ca. 2-metre-thick dyke-like body, but also to scarce small 148 veins within Unit-2. It is characterized by the presence of large biotite crystals showing a 149 specific textural organization (Fig. 4a-c) described farther. The grain size of the matrix lies 150 mostly between 0.2 and 0.6 mm (Fig. 4d), though some K-feldspar crystals may reach 1.5 cm 151 in length. A faint foliation occurs locally in connection with small shear-zones. 152 Internal contacts between the three units are sharp with no chilled margin and no crystals 153 nucleation on the contacts; they are marked mainly by change in the biotite size and locally by 154 loss of the fabric (between Unit-2 and Unit-3). Textures and mineral compositions are 155 presented in more detail in section 4.

156

# 157 **3. Whole-rock compositions**

158 Whole-rock major and trace element compositions are given in Table 1 and analytical 159 conditions in the Supplementary material SM.2. The rock is granitic ( $69.7 < SiO_2 < 75.2$  wt. 160 %), with a normative composition classifying it as alkali-feldspar granite (Q' = 34.9-42.4; 161 ANOR = 0.5-5.4) according to Streckeisen and Le Maitre (1979). It is peraluminous 162  $([Al_2O_3]/[CaO+Na_2O+K_2O] = 1.22-1.38)$ , with Mg# values ranging from 33 to 41 (total iron 163 as  $Fe^{2+}$ ), plotting close to the ferroan line in the classification diagram of Frost et al. (2001) 164 (not shown). Trace element contents are low, with the notable exception of Ba, Rb and Zr 165 (331–689, 211–264 and 72–149 ppm, respectively). Trace element discriminant ratios (Whalen et al., 1987) corroborate its S-type chemistry (Zr+Nb+Ce+Y = 139-246 ppm; 166

167	$10^{4*}$ Ga/Al = 2.1–2.4), corresponding to low-Ca unfractionated granite (X <sub>Sr</sub> < 0.15 and X <sub>Rb</sub> <
168	0.35) according to the nomenclature of El Bouseily and El Sokkary (1975). Zircon saturation
169	thermometry (Watson and Harrison, 1983) yields temperatures in the range 740–800 $^{\circ}$ C. Sr
170	and Nd isotopic compositions (Table 2) show ${}^{87}$ Sr/ ${}^{86}$ Sr(330 Ma) initial ratio of 0.726, $\epsilon$ Nd(330 Ma)
171	value of -8.2, and Nd model age ( $T_{DM}$ ) of 1865 Ma consistent with zircon U–Pb data attesting
172	to the presence of inherited core at 1.9 Ga (see Supplementary material SM.1).
173	Multi-element plot normalized to Primitive Mantle (Fig. 5a) shows depletion in Ba, U, Th,
174	Ta, Nb, Sr, Eu and Ti. K/Rb (198–254) and Y/Ho (27–29) ratios are similar to values of
175	common igneous rocks, and the Zr/Hf ratio (33-37) is close to that of most terrestrial rocks
176	(~38). Nb/Ta ratio (7–14, with the exception of Unit-1 with values at 19 and 23) compared to
177	Zr/Hf ratio corresponds to that of barren granites (Ballouard et al., 2016). However, the K/Ba
178	(73–194), La/Nb (1.7–5.3) and La/Ta (19–123) ratios are significantly higher than those of
179	common magmatic rocks, suggesting non-CHARAC behaviour of Ba, Nb and Ta, and thus,
180	the possible effect of hydrothermal fluids (e.g. Bau, 1996; Jahn et al., 2001). Rare earth
181	elements (REE) normalized to chondrite (Fig. 5b) show moderately fractionated light REE
182	([La/Sm] <sub>N</sub> = 2.9–5.2), negative Eu anomalies (Eu/Eu* = $0.3-0.7$ ) and weakly fractionated
183	heavy REE ( $[Gd/Yb]_N = 0.6-0.8$ ). The negative slope and concave-downward shape of heavy
184	REE, along with t <sub>3</sub> parameter ([Tb/Tb* $\times$ Dy/Dy*] <sup>0.5</sup> ; Irber, 1999) around 1.01–1.13, could
185	suggest a possible weak tetrad effect. Nevertheless, the TE <sub>1,3</sub> parameter $([t_1 \times t_3]^{0.5})$ , with $t_1 =$
186	([Ce/Ce* $\times$ Pr/Pr*] <sup>0.5</sup> ; Irber, 1999) remains low (1.00–1.09) compared to the minimum value
187	(1.1) generally considered to be significant of the tetrad effect (Irber, 1999).
188	

- **4. Textures and mineral compositions**
- 190 4.1. Quartz

191 In optical microscopy quartz appears mostly as euhedral (sub-hexagonal or quadrangular) to 192 subhedral (rounded) grains. Nevertheless, more complex shapes are observed: (i) grains 193 showing serrated outline resembling incipient branching arms (Fig. 6a); (ii) crystals with 194 equilibrated grain boundaries (Fig. 6b); (iii) triangular extensions and thin films (in optical 195 continuity with a main quartz grain) pervading feldspar-feldspar or feldspar-biotite grain 196 boundaries (Fig. 6c, d); (iv) crystals with rounded re-entrants (Fig. 6e) as commonly observed 197 in high-cooling magmas (rhyolite); and (v) anhedral grains in interstitial position probably 198 pseudomorphing the residual melt-filled porosity (Fig. 6f).

199 Cathodoluminescence imaging reveals that quartz grains have an internal structure 200 consisting of a bluish core commonly surrounded by reddish overgrowth. The cores 201 correspond to more or less rounded to elliptical grains with regular to wavy outlines (Fig. 7a), 202 or to skeletal grains with jagged outlines (Fig. 7b), branching structures (Fig. 7c) and hollow 203 cores (Fig. 7a, d). Overgrowths are not systematically present and do not always form a 204 continuous rim all around cores (e.g. Fig. 7a, b); they commonly lead to the coalescence of 205 neighbouring grains, giving rise locally to equilibrated granoblastic texture (Fig. 7e). Textural 206 relationships between quartz and biotite show unambiguously that cores predate biotite 207 crystallization, whereas overgrowths formed synchronously with biotite. This is indicated by 208 (i) blue cores directly in contact with biotite without overgrowth; (ii) space between two blue 209 cores filled with quartz-2 and biotite; (iii) thin red quartz outgrowths along the cleavage of 210 biotite; and (iv) reddish quartz forming symplectitic intergrowths with biotite, for some of 211 them in continuity with the overgrowth surrounding bluish quartz grains (Fig. 7f). 212 Quartz grain size varies from one unit to another. Sizes were measured in thin section 213 considering the long axes of the whole grains, even though this undoubtedly introduces a bias 214 due the presence of overgrowths. Nevertheless, the small thickness of these overgrowths, their

215 non-systematic occurrence and their similarity in all units lead us to consider that the size

216 variation of all grains is controlled by the size variation of cores. In Unit-1, quartz grain size 217 ranges from 0.04 to 0.6 mm with a median at 0.14 mm (Fig. 8a). In Unit-3, a similar range 218 (0.05 to 0.6 mm) is observed, but the sizes appear a little more homogeneous with ca. 57 % of 219 the grains between 0.15 and 0.25 mm (Fig. 8b, c). In Unit-2, quartz population shows higher 220 dispersion, with sizes ranging from 0.05 to 1.3 mm and medians  $\geq$  0.24 mm (Fig. 8d–f). On 221 the whole, although the average grain size does not significantly differ from one unit to the 222 other, we note that the histograms of size distribution differ in each unit, the standard 223 deviation is higher in the Unit-2 attesting to its greater heterogeneity, and the proportion of 224 grains with a size higher than 0.25 mm markedly increases from Unit-1 (ca. 15 %) to Unit-3 225 (ca. 25 %) and Unit-2 (30–55 %).

226

227 4.2. Garnet

Garnet occurs as euhedral to subhedral rounded grains (Fig. 9a, b), with size ranging between 0.4 and 2.0 mm. It may occur as inclusion in feldspar or in biotite (in this case, it may show partial retrogression; Fig. 9c). Garnet corresponds dominantly to a binary solid solution of almandine (77.6  $\pm$  3.6 mol. % on average) and pyrope (15.7  $\pm$  3.8 %), with limited amounts of spessartite (4.3  $\pm$  2.0 %) and grossularite (2.4  $\pm$  0.4 %). It is slightly more Mg-rich in the Unit-3 compared to the other units (Fig. 10a), whereas garnet from Unit-1 has a slightly lower content of spessartite + grossularite.

235

236 *4.3. Biotite* 

The common feature of biotite in all units is to be poikilitic (Fig. 11a), enclosing dominantly quartz grains but also in much smaller amount plagioclase and garnet. In Unit-1, biotite occurs as small rounded flakes to slightly larger anisometric crystals (Fig. 2a, b) with rather homogeneous sizes, mostly  $\leq 5$  mm (range 0.4–9 mm, Fig. 12a). In Unit-2, biotite occurs as

241 larger platelets parallel or at high angle to the fabric (Fig. 3d). It is more heterogeneous in size 242 (Fig. 3a), with length ranging from 0.3 to 28 mm (Fig. 12b); this heterogeneity being 243 enhanced by the presence of zones showing higher grain size (Fig. 3c). Some of the largest 244 crystals display branching bifid structure and hollow cores (Fig. 11b and 11e, respectively). In 245 Unit-3, biotite forms a network of large plates arranged at high angle to each other (Fig. 4b, 246 c), with sizes ranging from 0.7 to 850 mm (Fig. 12c). Its shape encompasses rounded or 247 slightly anisometric millimetre-sized flakes similar to those of the former units, to large 248 blades with width/length ratio around 0.4 (Fig. 4a). Radial growth patterns (Fig. 4b, c) and 249 dendritic shapes (Fig. 11c, d) are characteristic of biotite in this unit. It also shows hollow 250 cores filled with feldspathic material between thin biotite platelets in crystallographic 251 continuity with the main crystal (Fig. 11f). There is a clear difference in the histograms of 252 biotite size distribution between the three units: in Unit-1 the distribution is tightened around 253 1.4 mm, while in Unit-2 it is more heterogeneous with an average around 4.2 mm; histogram 254 of Unit-3 is given only for information, as it has very little significance due to the branching 255 nature and stronger anisotropic shape of the biotite plates. 256 Biotite is slightly magnesian (Fig. 10b), with  $X_{Mg}$  ratios ranging from 0.30 to 0.49 (0.41 ± 257 0.04 on average); TiO<sub>2</sub> contents are mostly around 3.1 % (range 1.07–4.42 wt. %); and Al<sup>VI</sup> 258 contents range from 0.54 to 1.60 atoms per formula unit (apfu). Its high total Al content (Altot

259 = 3.1-4.4 apfu) with respect to Mg (1.2–2.2 apfu) is consistent with the peraluminous

260 character of the host-rock. Secondary green biotite crystals, mainly developed at the expense

of garnet, depart from this compositional field, with higher Al<sup>VI</sup> values (0.9–1.6 compared to

262  $\leq 0.9$ ) and lower TiO<sub>2</sub> contents (mostly  $\leq 1.0$  wt. % compared to 3.1 % on average).

263

264 *4.4. Cordierite and tourmaline* 

265 Cordierite occurs as scarce centimetre-sized cockades and streaks surrounded by a biotite-266 free, quartzofeldspathic halo; cordierite includes numerous quartz grains but is free of 267 feldspars, and overall presents characteristics similar to late-magmatic cordierite reported 268 from other S-type granites (e.g. Barbey et al., 1999). It also contains a few grains of 269 tourmaline. These structures overprint the syn-magmatic fabric carried by biotite, indicating 270 that cordierite grew at its expense at the end of the crystallization sequence. 271 Cordierite is deeply retrogressed into pinite and we were not able to obtain reliable major 272 element composition. Tourmaline shows  $X_{Mg}$  values around 0.48–0.50.

273

#### 274 4.5. Feldspars

275 They correspond dominantly to K-feldspar and in lesser proportion to sodic plagioclase.

276 Plagioclase occurs as small euhedral to subhedral grains (mostly ca. 0.1–0.2 mm), scattered in

the matrix (Fig. 13a), forming locally glomerophyric aggregates (Fig. 13b), or overgrown on

quartz grains (Fig. 13f). Unaltered grains correspond to oligoclase/andesine (An $\leq 25$ ; Fig. 10a).

279 Albite is a widespread secondary mineral phase, which occurs as thin rims around

280 plagioclase, or veinlets along grain boundaries between the matrix K-feldspar grains (Fig.

13a); it also replaces primary plagioclase crystals, forming patchy zoning in association with

282 K-feldspar and muscovite (Fig. 13c, f).

283 K-feldspar, the main interstitial phase, may occur as subhedral to euhedral crystals (Fig.

13d), but in most cases, it is anhedral and forms an almost equilibrated mosaic between quartz

and plagioclase grains (Fig. 13a). It may be perthite-free, but more commonly contains

variable proportions of exsolutions, some crystals being strongly perthitic (Fig. 13e). It also

- 287 occurs as a secondary phase forming thin rims around plagioclase crystals (Fig. 13f), or
- 288 patches partly replacing them. K-feldspar has a composition ranging from Or<sub>74</sub>Ab<sub>25</sub>An<sub>01</sub> solid
- solution for the magmatic feldspar to the orthoclase end-member for the secondary one (Fig.

290 10c). Within a single grain, the composition varies very weakly, with Na<sub>2</sub>O contents ranging 291 from 0.3 to 1.2 wt.%. Analysis of a few exsolutions shows An-rich compositions (Or<sub>16-</sub> 292  $_{26}Ab_{61-68}An_{14-16}$ ).

293

#### 294 **5. Discussion**

295 Whole-rock geochemical data show that the studied alkali-feldspar granite derives from 296 parent magma generated by partial melting of an old recycled continental crust, in agreement 297 with the regional geodynamic reconstruction (Skrzypek et al., 2014). Its composition is close 298 to that of S-type felsic rocks used in crystallization experiments (e.g. Clemens and Birch, 299 2012; Scaillet et al., 1995) (Table 1), even though its rather high peraluminosity could be 300 partly related to the alteration of feldspars in sericite. However, the small compositional 301 heterogeneities (Si, Fe, REE, Th, U, Nb and Ta) observed within and in between units, and 302 their distinct textures seemingly implying distinct melt water contents (see below, section 303 5.3), suggest that the three units derive from three different magma batches with distinct water 304 content, and probably represent different stages of differentiation of a common parent magma.

305

# 306 5.1. Crystallization sequence

307 Defining the sequence of crystallization is not straightforward from textural relationships. 308 Inclusion of garnet as minute grains in plagioclase and as more or less retrogressed grains in 309 biotite suggests that it is probably one of the first phase on the liquidus. The common 310 occurrence of plagioclase as euhedral crystals overgrown on quartz grains, or in interstitial 311 position between them suggest that this phase started crystallizing after quartz. The 312 appearance of biotite postdates quartz, though their relationships are a little more complex in 313 detail (see next subsection). Timing of crystallization of biotite and plagioclase is to some 314 extent ambiguous. The presence of scarce plagioclase inclusions within biotite could suggest

that the former started to crystallize first; however, it is more likely that these two phases
followed closely each other. K-feldspar, the last magmatic phase to crystallize, mostly forms a
granoblastic mosaic of crystals embedding all the other minerals, even though it can be
locally euhedral, suggesting local crystallization without interference with neighbouring
grains. This sequence ends with the scarce growth of cordierite (± tourmaline) at the expense
of biotite, and then partial subsolidus reequilibration (albite, chlorite, etc.).

321 From previous works (Latouche et al., 1992; Rey et al., 1989; Skrzypek et al., 2012) and 322 U-Pb zircon data indicating that the alkali-feldspar granite predates the Central Vosges 323 granite association (Supplementary material SM.1), a poorly constrained 200–500 MPa range 324 could be envisaged for the conditions of emplacement. Temperatures estimated from zircon 325 saturation thermometry (740–800°C) are too low to be considered those of the magma at the 326 time of its emplacement, especially since part of the zircon is inherited. Although the high 327 variance of the phase assemblage does not allow a precise reconstruction of the P-T 328 conditions of emplacement, we attempted a comparison with the experimental results 329 obtained by Clemens and Birch (2012) on the Rubicon Ignimbrite, compositionally close to 330 our granite. From their T–X<sup>Fl</sup>H<sub>2</sub>O phase diagrams it appears that the early crystallization of 331 garnet in the absence of orthopyroxene, cordierite and biotite implies crystallization pressure 332 around 500 MPa from hydrated magma, and this seems to occur in a restricted field (800 < T 333  $< 880^{\circ}$ C,  $0.4 < X^{Fl}$ H<sub>2</sub>O < 0.8). Their results also indicate that (i) the stability field of 334 cordierite expands widely with decreasing pressures, and (ii) feldspars are the late phases to 335 appear. Comparing our sequence of crystallization with these experimental data could suggest 336 that crystallization started at pressure around 500 MPa from a magma with significant melt 337 water content, but took place mainly at lower pressure to account for the early appearance of 338 quartz and the late crystallization of cordierite. In fact, we should expect cordierite to appear 339 earlier in the sequence, which is not the case; it cannot be excluded that its absence in the first

steps could be due to inhibited nucleation as observed in experiments (Clemens and Birch,
2012). A last point is that the onset of plagioclase crystallization occurred before biotite and
K-feldspar, which is at odds with the experimental results showing that plagioclase is late in
the crystallization sequence. Nevertheless, in experiments on Himalayan leucogranite
(Scaillet et al., 1995) also compositionally close to our granite, plagioclase appears early,
suggesting that minor compositional differences may lead to inversion in the crystallization
order.

347

# 348 5.2. Significance of the textures of quartz and biotite

349 From the cathodoluminescence properties of quartz we consider that the bluish cores and 350 reddish overgrowths of quartz grains represent two successive growth stages. Indeed, studies 351 of quartz from igneous rocks (Götze et al., 2001; and references therein) suggest that Al (and 352 possibly Ti) could play a role in the blue emission component, whereas the red component 353 (often observed in rims and cracks of crystals) would be related to lattice defects. Quartz-1 354 (bluish cores) predates biotite and corresponds to both skeletal crystals and rounded to 355 subhedral grains with a regular to wavy outline. Quartz-2 (reddish overgrowths) and biotite 356 crystallized synchronously as indicated by their symplectitic relationships (Fig. 7f). Growth of 357 quartz-2 resulted in the formation of rounded to euhedral grains from the skeletal cores, as 358 well as in grain coalescence and equilibrated textures by grain feeding along boundaries from 359 the melt. On the whole, the sharp contact between the two generations of quartz shown by CL 360 and their distinct textural position mark an abrupt change in growth conditions. This is 361 distinct from what is observed in olivine wherein P enrichment appears to reflect a continuous 362 decay in growth rate (e.g. Welsch et al., 2014).

363 The skeletal shapes of quartz-1 can be interpreted in two ways, either dissolution or364 disequilibrium growth. Dissolution appears highly unlikely even though adiabatic

365 decompression can lead to remelting of early crystallized assemblages (Johannes and Holtz, 366 1996), and this for two main reasons. First, quartz dissolution experiments in rhyolitic melts 367 (Donaldson and Henderson, 1988) show that this process leads to round corners and edges, 368 and overall to more compact morphologies, but not to sharp crystal edges as we observe (see 369 also Swanson and Fenn, 1986). Secondly, if we rely on the phase relationships of the Rubicon 370 ignimbrite, it appears that for a magma (500 MPa, 850°C,  $X^{Fl}H_2O = 0.5$ ) ascending 371 adiabatically, quartz starts to crystallize only down to 200 MPa, i.e. close to the level of 372 emplacement. We rather consider that the skeletal shapes result from diffusion-controlled 373 growth as observed in crystallization experiments at high undercooling (Swanson and Fenn, 374 1986; MacLellan and Trembath, 1991). In addition, variation of the degree of undercooling 375 during emplacement could account for the occurrence of both skeletal habits (early high 376 supersaturation) and rounded grains with smooth outlines (subsequent decreasing 377 undercooling). In any case, the skeletal shape of quartz-1, whatsoever resulting from 378 dissolution or disequilibrium crystallization, can be considered as primary disequilibrium 379 textures preserved behind the apparent "coarse-grained" texture. 380 It is worth of note that the CL image of Fig. 8b contains a quartz grain showing a hollow 381 core with arms radiating towards its centre. This grain presents a sub-hexagonal section (faces 382 are easily recognizable), thus not far from being orthogonal to the c axis. This disposition 383 seems therefore to indicate that the arms are radial with respect to this axis, which is in 384 agreement with the experimental results of Swanson and Fenn (1986). Looking at the quartz grain size in the three units (Fig. 8), it appears that its distribution in 385 386 Unit-1 and Unit-2 are quite comparable, differing mainly by the average grain size and the 387 proportion of larger grains (~ 15 % in Unit-1 vs. 30–55 % in Unit-2). This may simply result 388 from distinct cooling, due to the thermal state of the host rock. Compared to Unit-2, the 389 histogram of Unit-3 differs by a limited population of small grains ( $\leq 0.1$  mm) and a very

390 restricted population of larger grains (~ 25  $\% \ge 0.3$  mm) which is closer to that of Unit-1. 391 Although we cannot fully discard cutting effects, we consider that two processes may account 392 for this peculiar distribution. Firstly, the limited amount of small grains suggests coarsening 393 by Ostwald ripening probably mainly in favour of medium sizes since this process seems to 394 be only effective for small sizes (Cabane et al., 2001). This also implies that there was no 395 creation of new grains and, thus, that the nucleation of quartz has not been continuous. 396 Secondly, the restricted population of larger grains implies that coarsening was rapidly 397 impeded.

398 Textural relationships indicate that biotite postdates quartz-1 and possibly plagioclase, 399 while it is coeval with quartz-2. Furthermore, variation of its size and morphology from one 400 unit to the other suggests distinct nucleation and growth rates, and therefore distinct magma 401 supersaturation in each unit. More particularly, the large size of biotite crystals in Unit-3 and 402 their tendency to form radiating clusters (nucleation of several grains at a single site) implies 403 very limited nucleation. This precludes in situ differentiation of the three units from a unique 404 magma batch by fractional crystallization. We note that the morphology of biotite from Unit-3 405 resembles those of biotite from stockscheider pegmatites (see for example Haapala and 406 Lukkari, 2005; their Fig. 3c). However, the absence, in this unit and in the whole granite body 407 as well, of rhythmic mineralogical laminations, feldspar and quartz segregation, inward 408 gradual increase of crystal size and unidirectional growth texture from walls indicates that the 409 studied granite cannot be equated to pegmatite (for pegmatite features, see London and 410 Morgan, 2017), even though similar process may have been at work.

411

412 5.3. A possible crystallization scenario

413 Some experimental results may help us in building an appropriate scenario. (1) As pointed out

414 by Swanson (1977) and Swanson and Fenn (1986), the nucleation of quartz is an abrupt

415 process at high rates in granitic melts. (2) Ostwald ripening of quartz grains seems to be an 416 active process only at the end of nucleation event, becoming negligible when grains reach 417 millimetre size (Cabane et al., 2001). (3) At high undercooling (i.e. high supersaturation 418 conditions), crystal habit evolves towards skeletal and dendritic shapes, with radial growth 419 patterns (e.g. London, 2005; and references therein, for silicic rocks). More specifically, 420 London and Morgan (2017) show in their experiments that limited undercooling leads to fine-421 grained disseminated crystals, whereas at high undercooling crystal evolves towards skeletal 422 and radiating morphologies.

Considering that the three units correspond to three distinct batches from a common source, emplaced at the same level in the crust, the only two parameters that are likely to vary to account for distinct supersaturation in each unit at the site of emplacement, are (i) the temperature of the surrounding rocks, owing to the fact that the Unit-1 was emplaced in colder host-rocks while the Unit-2 and Unit-3 are nested in the preceding ones; and (ii) the volatile content of the melt and extent of degassing that may differ from one magma batch to the other. We therefore propose a crystallization scenario in three main steps.

(1) *The first step* common to all units corresponds to the onset of crystallization of garnet
from a hydrated silicic peraluminous melt under pressures of ca. 500 MPa to avoid early
crystallization of orthopyroxene or cordierite. It is not unlikely that this happened in an
evolving intermediate magma chamber that fed the successive injections (Fig. 14a).

(2) *The second step* corresponds to magma emplacement and the onset of crystallization of
quartz, followed by plagioclase, biotite and K-feldspar. It is this step that imprints the textural
characteristics of the three units. Decompression and cooling during magma ascent may have
led to undercooled melt, resulting in low nucleation and rapid growth of skeletal quartz-1
when magma reached its site of emplacement (Fig. 14b). The subsequent evolution has to be
discussed for each unit, separately (Fig. 14c, d).

The first batch of magma (Unit-1) was emplaced in a colder environment. This induced rapid cooling and significant undercooling, which led to high nucleation of quartz followed by limited but to some extent protracted growth resulting in a certain heterogeneity towards larger grain sizes. The crystallization of quartz led in turn to significant supersaturation with respect to plagioclase and biotite. An important peak of nucleation followed by limited growth gave a homogeneous population of small-sized biotite crystals.

446 The next magma injection (Unit-2) intruded Unit-1, i.e. an environment hotter than for the 447 first batch. This led to moderate cooling rate and undercooling, lowered nucleation but 448 increased growth of quartz, with limited Ostwald ripening; the result is a more heterogeneous 449 population of larger quartz grains. The subsequent crystallization of biotite followed the same 450 evolution as quartz, also resulting in a heterogeneous population of larger crystals. The 451 heterogeneity and larger size of both quartz and biotite along with the higher amount of 452 deformation recorded in this unit seem to suggest a longer duration of solidification. Overall, 453 Unit-1 and Unit-2 show crystallization process no so much different from common igneous 454 rocks.

455 The third batch of magma (Unit-3) is distinguished by a more homogeneous population of 456 quartz compared to Unit-2, suggesting that both nucleation and coarsening were to some 457 extent hindered, and therefore that crystallization process has been limited in time. This needs 458 conditions distinct from those operating in the former units, i.e. a process other than cooling 459 rate; this is also supported by the absence of unidirectional growth pattern, thus precluding a 460 significant role of thermal contrast between Unit-2 and Unit-3. The best way to explain these 461 features is to invoke a sudden drop of water pressure by degassing of a magma with higher 462 melt water content, triggering high degree of undercooling by shifting the liquidus of the 463 system, which makes growth and nucleation more rapid. However, this is counteracted by 464 increase in the viscosity of the melt, slowing down diffusion. This may also account for

reducing nucleation of biotite and thus for its high growth rate and dendritic habit. The low
diffusivity may also be the cause of biotite crystals forming radiating clusters (nucleation of
several grains at a single site).

The end of this step corresponds to the onset of crystallization of K-feldspar (even though it is not possible to determine precisely when this phase started crystallizing), forming mostly a mosaic of equilibrated grains. Note that crystallization of biotite and feldspars maintained in all units the supersaturation of the residual melt in silica and led concomitantly to the crystallization of quartz-2 overgrowing former quartz grains and filling pore spaces.

473 (3) *The last step* common to all three units corresponds to the local growth of cordierite (±
474 tourmaline); and, finally, to subsolidus fluid-assisted reequilibration (albite, chlorite, etc.).

We are aware that this scenario is very qualitative, but linking it to quantitative assessment
of nucleation and growth rates as functions of undercooling is not really possible considering
the present state of available experimental studies.

478

## 479 **6.** Conclusions

(1) The studied S-type alkali-feldspar granite was emplaced as three nested units. The
crystallization sequence started with garnet, then proceeded with quartz, plagioclase, biotite
and K-feldspar, and ended with cordierite. In Unit-1, quartz and biotite are small-sized; in
Unit-2, they are coarser-grained forming more a heterogeneous population; and in Unit-3,
quartz forms a homogeneous population of smaller grains than in Unit-2, whereas biotite is
significantly larger in size (up to 8.5 cm).

486 (2) Quartz occurs mostly as subhedral to euhedral grains, even though more complex
487 shapes are observed. Cathodoluminescence imaging reveals that quartz grains consist of
488 bluish rounded or skeletal core filled and/or surrounded by reddish overgrowth. Biotite is

489 poikilitic in all units including mostly quartz grains but also plagioclase and garnet; in unit 3,
490 biotite crystals are arranged radially, have dendritic habits and also skeletal cores.

491 (3) The variability in size and morphology of quartz and biotite is interpreted in terms of
492 variable undercooling related to variable temperature of the host-rocks and to more or less
493 intense degassing due to magma batches characterized by variable melt water content.

(4) Overall, this study illustrates that behind the common "coarse-grained" texture of
quartz in granite, subsist primary disequilibrium crystallization textures. This probably results
from a favourable situation in which the thermal evolution has not totally erased the initial
crystallisation textures. However, it is not impossible that majority of granites kept relics that
will not be visible without examination under cathodoluminescence. Finally, the claim of
Welsch et al. (2013) that all euhedral olivine crystals in basaltic rocks are initially dendritic or
skeletal could also be applicable to other minerals and rock types.

501

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509

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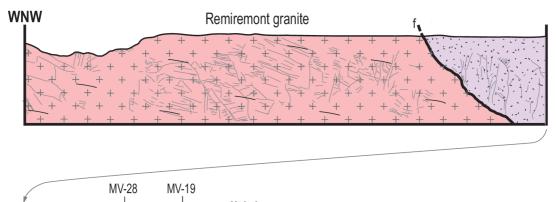
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687	
688	Figure captions
689	
690	Fig. 1. Cross-section of the alkali-feldspar granite body, with sample location. Sample
691	numbers refer to whole-rock and mineral analyses of Table 1 and Supplementary material
692	SM.3.
693	
694	Fig. 2. Unit-1. Size and morphology of biotite and quartzofeldspathic matrix. (a) Polished
695	slab; (b) and (c) thin sections in plane polarized light and crossed nicols, respectively. ( $S_m =$
696	magmatic fabric; qz = quartz; fsp = feldspar; Kfs = K-feldspar; pl = plagioclase; bt = biotite;
697	grt = garnet.)
698	
699	Fig. 3. Unit-2. (a) Polished slab showing the size of biotite and its rough orientation along the
700	syn-magmatic foliation ( $S_m$ ). (b) Detail of biotite (blades) and garnet (dots) in a weakly
701	foliated part of the unit (rough surface). (c) Polished slab showing an autolithic enclave
702	wrapped by the foliation; note also the larger grain size of the lower part of the slab. (d) Thin
703	section in plane polarized light showing the size and morphology of biotite, and texture of the
704	quartzofeldspathic matrix.
705	

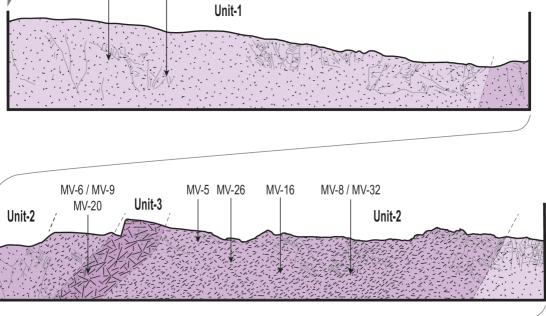
706	<b>Fig. 4.</b> Unit-3. (a) and (b) Polished blocs showing the 3D organization of biotite plates. (c)
707	Polished slab showing the disposition of biotite and small rounded garnet grains. (c) Thin
708	section in plane polarized light showing the size and morphology of biotite and quartz.
709	
710	Fig. 5. (a) Multi-element plot normalized to Primitive Mantle (Sun and McDonough, 1989).
711	(b) REE patterns normalized to chondrite (Evensen et al., 1978).
712	
713	Fig. 6. Optical microphotographs of quartz. (a) Quartz grain with serrated edges. (b)
714	Equilibrated grain boundaries between quartz grains (see also Fig.7e). (c) Quartz film
715	(arrowed) along grain boundaries, in crystallographic continuity with the rounded quartz
716	grain. (d) Triangular extensions (arrowed) of quartz along feldspar-feldspar or feldspar-quartz
717	grain boundaries. (e) Re-entrants with tips (arrowed) tending to meet and form an inclusion.
718	(f) Interstitial quartz (arrowed) along grain boundaries between feldspar grains. (b), (c), (d)
719	and (e): Unit-3; (a) and (f): Unit-2.
720	
721	Fig. 7. Cathodoluminescence images of quartz. (a) Elliptical cores (qz-1) with wavy or jagged
722	outlines, overgrown and welded by a second generation of quartz (qz-2); note the hollow core
723	of qz-1 to the right. (b) and (c) Cores showing branching arms. (d) Hollow crystal with
724	roughly radiating branches. (e) Coalescence of quartz grains welded by reddish quartz
725	overgrowth leading to an equilibrated grain assemblage. (f) Symplectitic quartz intergrowth
726	with biotite (arrowed); note that the symplectitic quartz overgrows a bluish quartz grain.
727	Other mineral phases are K-feldspar (blue and green), plagioclase (light brown), albite
728	(yellow) and biotite (dark). Scale bar: 100µm. (a), (b) and (e): Unit-3; (c) and (d): Unit-2.
729	

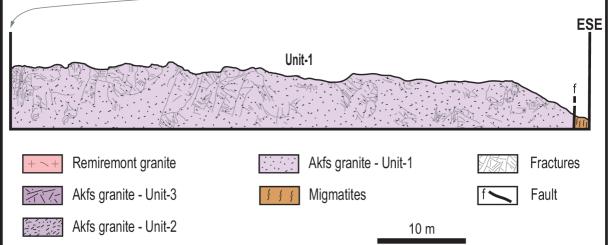
730	Fig. 8. Size distribution of whole quartz grains in the three units. Note the greater size
731	heterogeneity in Unit-2. (aver. = average and standard deviation; n= number of grains
732	measured; diam. = diameter.)
733	
734	Fig. 9. Microphotographs of garnet. (a) Euhedral garnet crystal (Unit-3). (b) Rounded garnet
735	grain included in plagioclase (Unit-2). (c) Retrogressed garnet crystal embedded in a biotite-
736	plagioclase groundmass (Unit-2).
737	
738	Fig. 10. Mineral compositions: (a) Garnet in the Almandine–Pyrope–Spessartite+Grossularite
739	triangle; (b) Biotite in the Al <sup>VI</sup> vs. (Fe + Mg) diagram; and (c) Feldspars in the Ab–Or–An
740	triangle (p = perthite). Compositions presented in these diagrams are given in the
741	Supplementary material SM.3.
742	
743	Fig. 11. Morphology of biotite. (a) Strongly poikilitic aspect of biotite including numerous
744	quartz grains (Unit-3). (b) Branching, bifid, biotite crystal from Unit-2. (c) and (d) Dendritic
745	crystals from Unit-3. (e) Biotite showing hollow core (Unit-2). (f) Biotite with hollow core
746	filled with feldspar mainly (Unit-3).
747	
748	<b>Fig.12.</b> Size distribution of biotite crystals in the three units (aver. = average and standard
749	deviation; n= number of grains measured; diam. = diameter).
750	
751	Fig. 13. Morphology of feldspars. (a) Back-scattered electron microscopy (BSEM) image
752	showing a K-feldspar groundmass containing euhedral plagioclase crystals and rounded
753	quartz grains; note the presence of albite veinlets along K-feldspar grain boundaries and
754	around the euhedral plagioclase crystal. (b) BSEM image of glomerophyric texture of

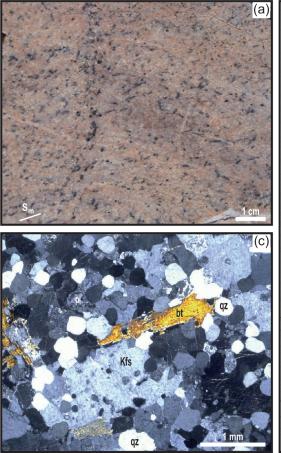
plagioclase in a groundmass consisting of quartz (dark grey) and K-feldspar (light grey). (c)
Optical microscopy image of plagioclase crystal showing a partly retrogressed core rimmed
by albite and K-feldspar. (d) CL image showing the matrix forming euhedral to subhedral Kfeldspar crystals (green to grey). (e) Optical microscopy image of a perthitic K-feldspar
crystal. (f) CL image showing partial replacement of plagioclase crystals into albite or
plagioclase rimmed with K-feldspar and albite (same grain as in c). (a), (b), (c), (e) and (f):
Unit-3; (d): Unit-2.

763	<b>Fig. 14.</b> Interpretative sketch illustrating the main step of solidification of the alkali-feldspar
764	granite (melt = light pink, garnet = red, quartz-1 = grey, quartz-2 = light grey, plagioclase =
765	brown, biotite = black, primary K-feldspar = green, secondary albite = yellow, secondary K-
766	feldspar = blue).

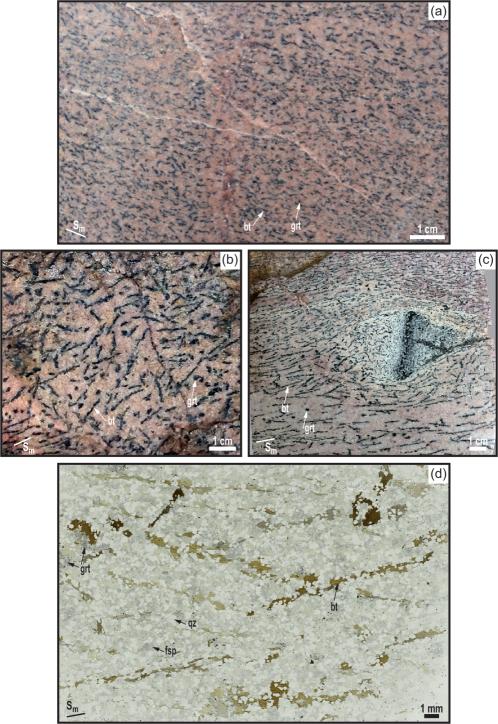


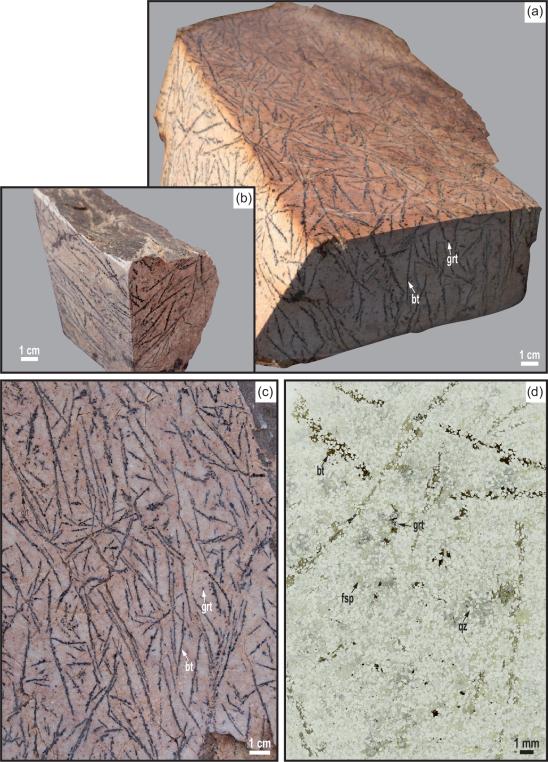


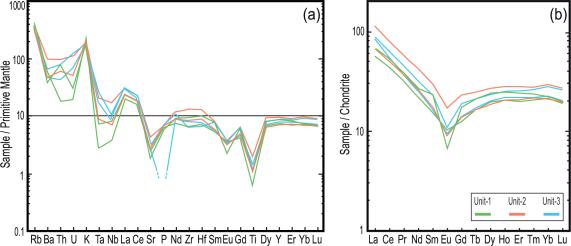


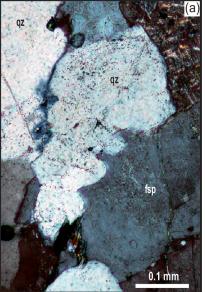


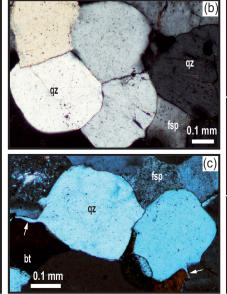


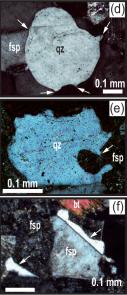


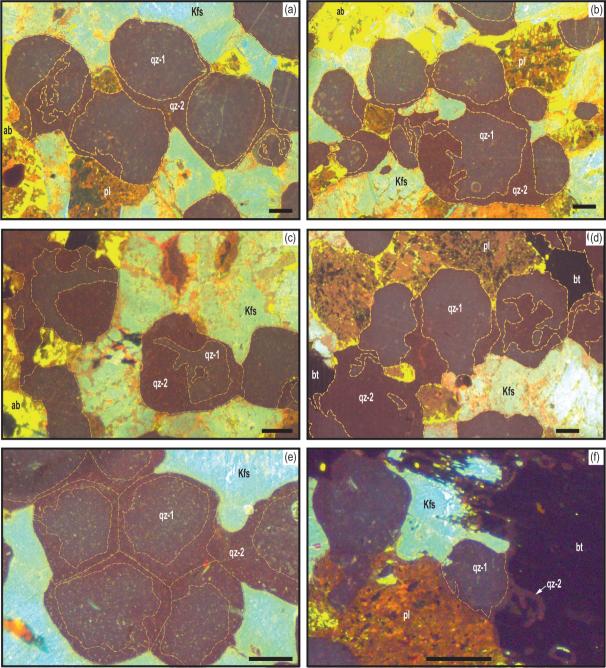


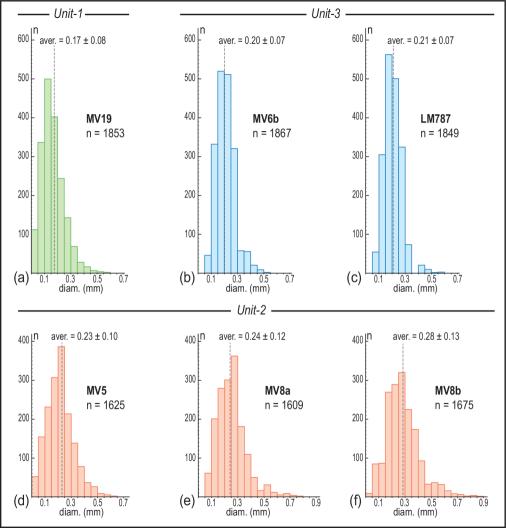


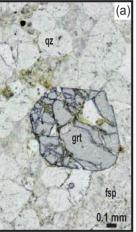


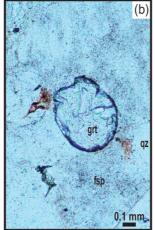


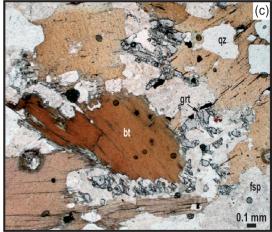


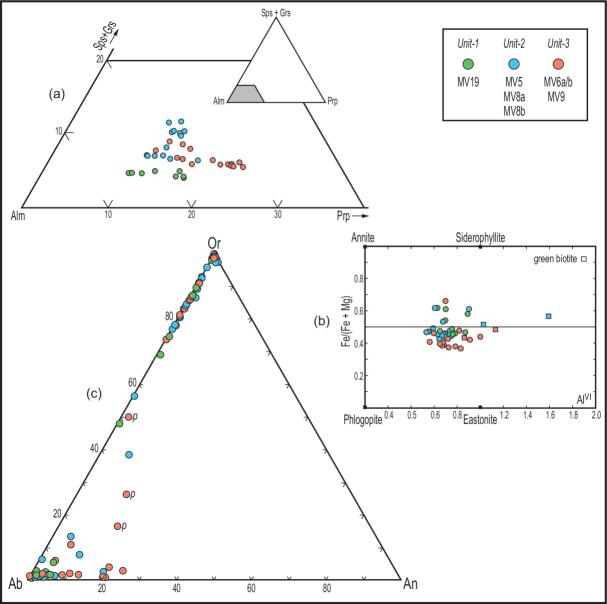


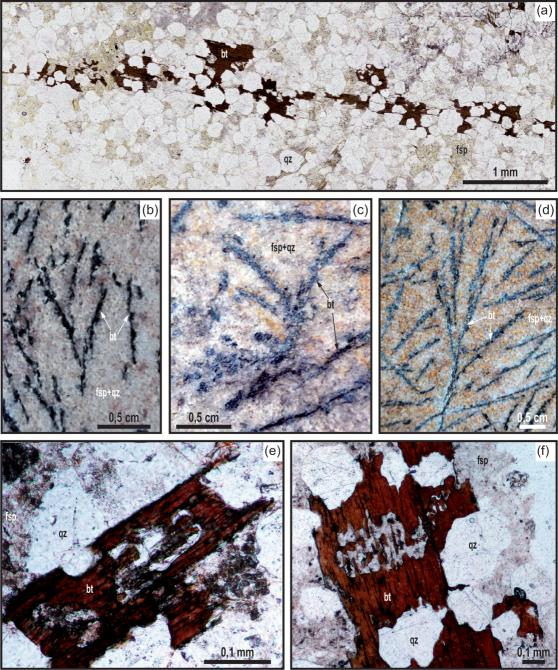


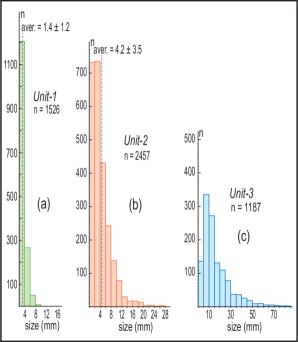


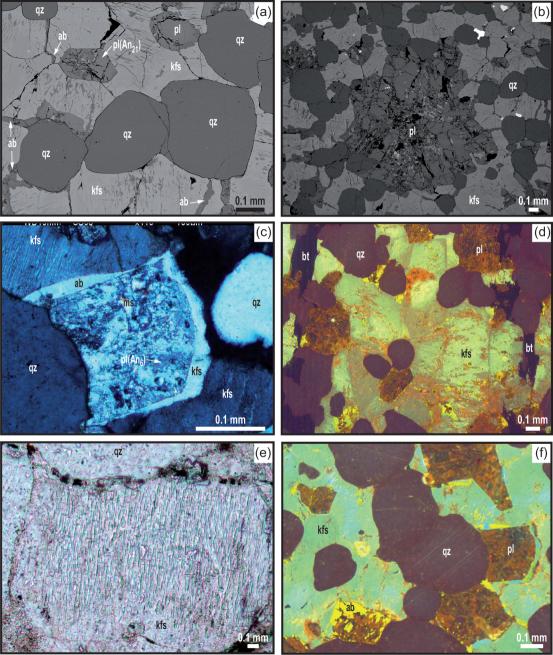


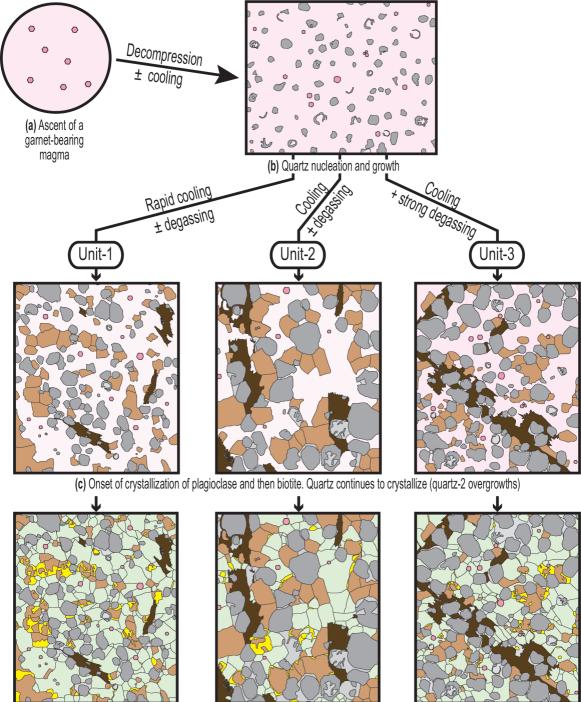












(d) Onset of K-feldspar crystallization. Quartz continues to crystallize, filling residual pore spaces. Late crystallization of cordierite (not shown), followed by subsolidus fluid-assisted alteration (albite, chlorite, etc.)

Table 1: Whole-rock major (wt.%) and trace element ( $\mu g/g$ ) composition of the three units of the alkali-feldspar							
granite. Shown for comparison: whole-rock major element composition of Himalayan leucogranite (HL;							
Scaillet et al., 1995) and Rubicon ignimbrite (RI; Clemens et al., 2012).							

Unit	Unit-1		Unit-2		Unit-3		HL	RI
Sample	MV19	MV28	MV8	MV16	MV9	MV20	DK89	9397
SiO <sub>2</sub>	74.18	74.03	69.70	74.10	72.86	75.22	73.04	75.77
$Al_2O_3$	14.19	12.70	14.09	13.01	13.70	13.04	15.32	13.54
FeOtot	1.10	1.89	3.53	1.87	2.41	1.56	0.74	1.69
MnO	_	0.03	0.06	0.03	0.04	0.02	0.01	0.01
MgO	0.32	0.52	1.30	0.60	0.94	0.57	0.20	0.31
CaO	0.24	0.21	0.36	0.30	0.36	0.29	0.85	0.65
Na <sub>2</sub> O	2.24	1.88	2.02	2.23	2.61	2.59	3.85	2.64
K <sub>2</sub> O	7.08	5.97	6.02	6.27	5.31	5.40	4.96	5.27
TiO <sub>2</sub>	0.13	0.23	0.44	0.24	0.29	0.23	0.13	0.11
$P_2O_5$	0.15	0.12	0.14	0.14	_	0.15	0.14	
LOI	1.29	1.23	1.32	1.04	1.48	1.34	0.79	
Total	100.93	98.80	98.97	99.82	100.00	100.40	100.03	100.00
As	3.7	3.1	4.0	3.0	9.4	7.2		
Ba	429	255	4.0 689	333	331	448		
Be	429 0.7	0.9	1.0	0.9	1.0	1.2		
Co	1.6	2.2	7.2	3.0	4.7	3.1		
Cr	4.8	12	40	13	25	18		
Cs	4.8	7.2	12.7	9.4	11.9	9.4		
Cu	4.3	2.7	11.5	7.1	7.4	6.7		
Ga	1.5	16	18	16	17	16		
Ge	1.2	1.2	1.5	1.2	1.4	1.4		
Hf	2.3	3.1	4.1	2.8	2.5	2.1		
Nb	2.3	5.8	12	5.1	7.4	6.0		
Ni	43	4.6	80	6.9	60	33		
Pb	11	11	17	23	14	14		
Rb	264	234	236	205	222	211		
Sc	2.3	0.1	12	0.1	7.2	4.9		
Sn	2.4	1.8	3.6	2.7	6.3	5.6		
Sr	51	38	91	56	61	64		
Ta	0.1	0.3	0.9	0.4	1.1	0.7		
Th	1.6	6.9	8.5	5.1	3.7	6.7		
U	0.4	0.6	2.4	1.1	1.5	2.7		
V	4.8	13	49	16	29	21		
W	-	_	1.1	-	1.5	1.5		
Y	33	39	44	32	36	39		
Zn	16	25	55	27	33	28		
Zr	75	106	149	96	93	72		
La	14.02	16.74	27.55	16.57	20.65	21.50		
Ce	28.36	34.07	50.95	32.16	20.03 35.57	40.86		
Pr	3.06	3.65	5.66	3.53	3.58	40.80		
Nd	10.34	12.83	20.10	12.36	12.12	15.05		
Sm	2.36	3.57	4.59	2.65	2.51	3.57		
Eu	0.59	0.38	0.99	0.53	0.54	0.61		
Gd	2.60	3.61	4.77	2.83	2.87	3.89		
Tb	0.62	0.79	0.94	0.62	0.65	0.81		
Dy	5.01	6.09	6.80	4.78	5.10	5.92		
Ho	1.16	1.41	1.59	1.16	1.24	1.42		
Er	3.35	3.97	4.66	3.47	3.66	4.25		
Tm	0.53	0.60	4.00 0.71	0.56	0.56	0.67		
Yb	3.53	3.66	4.86	3.48	0.50 3.70	4.67		
Lu	0.49	0.50	4.80 0.69	0.50	0.52	4.07		
Eu	total iron as		0.09	0.50	0.32	0.07		

 $FeO^{tot} = total iron as Fe^{2+}$ ; LOI = loss on ignition; - = below detection limit.

Table 2: Whole-rock Sr and Nd isotopic composition of the "spinifex" unit (sample MV9).

Rb (ppm)	222.02	Sm (ppm)	2.51
Sr (ppm)	61.08	Nd (ppm)	12.12
<sup>87</sup> Rb (µmol)	0.72305	<sup>147</sup> Sm/ <sup>144</sup> Nd	0.1253
<sup>86</sup> Sr (µmol)	0.06842	143Nd/144Nd	0.512066
<sup>87</sup> Rb/ <sup>86</sup> Sr	10.5681	2σ	0.000008
<sup>87</sup> Sr/ <sup>86</sup> Sr	0.776134	( <sup>143</sup> Nd/ <sup>144</sup> Nd)t	0.511797
2σ	0.000014	εNd <sub>(t)</sub>	-8.2
( <sup>87</sup> Sr/ <sup>86</sup> Sr)t	0.7265	T <sub>DM</sub> (Ma)	1865

 $\frac{(-517-517)t}{CHUR \text{ composition used to calculate } \epsilon_{Nd} \text{ values is } ^{143}\text{Nd}/^{144}\text{Nd}} = 0.512638 \text{ and } ^{147}\text{Sm}/^{144}\text{Nd} = 0.1967; \text{ the decay constant is } 6.54x10^{-12}; \text{ Nd model ages are calculated using } ^{143}\text{Nd}/^{144}\text{Nd} = 0.511580 \text{ for the depleted mantle; } (^{87}\text{Sr}/^{86}\text{Sr})_t, (^{143}\text{Nd}/^{144}\text{Nd})_t \text{ and } \epsilon\text{Nd}_{(t)} \text{ calculated at } 330 \text{ Ma.}$