

1 **Revision 2**

2 **Using cathodoluminescence to identify oscillatory zoning of perthitic K-feldspar from**
3 **the equigranular Toki granite**

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13

Abstract

14

15 For the first time, cathodoluminescence (CL) was used to show oscillatory zoning in
16 perthitic K-feldspars from the equigranular Toki granite, central Japan. Based on the CL
17 patterns, two types of zoning are identified: single core oscillatory zoning (SCOZ) and
18 multiple core oscillatory zoning (MCOZ). The SCOZ is defined by oscillatory zoning
19 around a single crystal core within the K-feldspar crystal, whereas the MCOZ depicts two
20 or more such crystal cores. The crystal cores displayed in CL images reflect the nucleation
21 parts of magmatic K-feldspar. The existence of MCOZ patterns in K-feldspars indicates
22 multiple nuclei. CL patterns reveal crystal growth behavior of magmatic K-feldspar in the
23 equigranular Toki granite. CL intensities are positively correlated with titanium and barium
24 concentrations, indicating that the CL variations depend on two factors: 1) titanium
25 concentration as a CL activator and 2) density of Al-O⁻-Al structural defects. The analysis
26 of CL images revealed that albite-rich phases in microperthite and patchperthite with low
27 luminescence intensities cut across the CL bands of the oscillatory zoning, indicating that
28 the oscillatory zoning in the orthoclase-rich host phase of K-feldspar was not perturbed by
29 the formation of microperthite and patchperthite in the post-crystallization stage. The
30 luminescence intensities of albite-rich phases in patchperthite are lower than those in
31 microperthite, which is due to the differences in titanium and barium concentrations
32 between them. In the post-crystallization stage, the mass transfer of titanium and barium
33 occurred during the formation of microperthite and patchperthite. Therefore, the difference
34 in the luminescence intensities between microperthite and patchperthite lamellae reflects
35 their different formation mechanisms between exsolution coarsening and dissolution–

36 precipitation coarsening. In summary, CL analyses can be used for the evaluation of the
37 nucleation and growth not only of anhedral K-feldspar crystals in equigranular granite but
38 also of K-feldspar phenocrysts/megacrysts in porphyritic granite. It can reveal the spatial
39 extent of element partitioning between the melt and crystal, along with that of mass transfer
40 from the melt into crystals during the magma evolution. Moreover, the CL analyses can
41 also be used for the interpretation of K-feldspar textural development during the
42 post-crystallization stage.

43

44 **Keywords:** cathodoluminescence, K-feldspar, oscillatory zoning, microperthite,
45 patchperthite, granite.

46

47

Introduction

48 Oscillatory zoning is defined as multiple growth layers that are generally parallel to
49 crystallographic planes and range in thickness from tens of nanometers to several tens of
50 micrometers (Shore and Fowler, 1996). Potassium feldspar (K-feldspar) is one of the most
51 common minerals in felsic plutonic rocks. Oscillatory zoning is frequently observed in
52 K-feldspar megacrysts and phenocrysts under polarized light (Vernon and Paterson, 2008;
53 Vernon, 2010) as well as in backscattered electron images (BSE; Landi et al., 2019). For
54 example, in granitic rocks from the Sierra Nevada batholith, K-feldspar megacrysts of
55 igneous origin show elemental oscillatory zoning marked by varying concentrations of BaO,
56 which range from 0.5 wt% to 3.5 wt% (Moore and Sisson, 2008). K-feldspar phenocrysts in
57 porphyritic Half Dome and Cathedral Peak granodiorite from the Tuolumne intrusion also
58 show elemental oscillatory zoning with BaO concentrations ranging from <0.5 wt% to ~2
59 wt% (Johnson and Glazner, 2010; Glazner and Johnson, 2013).

60 Cathodoluminescence (CL) patterns of minerals in granite can be used to reveal mineral
61 genesis and growth, which also contribute to clarifying the intrusion, emplacement, and
62 cooling processes of granitic magma (Marshall, 1988; Nakano et al., 2005; Kayama et al.,
63 2010) and the magma evolution (Yuguchi et al., 2020). In previous CL studies of K-feldspar
64 in granite, Kayama et al. (2010) characterized the CL emission centers in K-feldspar from
65 the Cerro Balmaceda pluton syenite and observed CL heterogeneity in K-feldspar. Lee et al.
66 (2007) and Parsons et al. (2008) reported that albite-rich patches in perthitic K-feldspar
67 from the Klokken syenite display oscillatory zoning in CL images. Perthitic K-feldspar
68 consists of albite-rich phases (patch and lamella) in orthoclase-rich host phases. Oscillatory

69 zoning in orthoclase-rich host phases of K-feldspar megacrysts and phenocrysts was
70 reported by several previous CL studies (e.g., [Słaby et al., 2008](#); [Higgins, 2017](#); [Oppenheim](#)
71 [et al., 2021](#)). K-feldspar phenocrysts from porphyritic Half Dome and Cathedral Peak
72 granodiorites display CL oscillatory zoning in the orthoclase-rich host phase under
73 optical-CL observations (cold-cathode color cathodoluminescence stage attached to
74 stereoscope) ([Oppenheim et al., 2021](#)). [Higgins \(2017\)](#) presented cathodoluminescence
75 images of K-feldspar megacrysts from the Cathedral Peak granodiorite based on optical-CL
76 observation (cold-cathode color cathodoluminescence instrument mounted on a
77 petrographic microscope), displaying CL oscillatory zoning in the orthoclase-rich host
78 phase. CL oscillatory zoning of K-feldspar phenocrysts (without perthitic texture) in
79 microgranular magmatic enclaves in the Karkonosze granite was found based on optical-CL
80 observations (hot-cathode instrument attached to digital microscope) ([Słaby et al., 2008](#)).
81 [Słaby et al. \(2008\)](#) revealed a positive correlation between CL intensity and barium
82 concentrations, indicating that the differences in CL intensities are attributed to Al-O⁻-Al
83 structural defects due to coupled KSi–BaAl exchange.

84 However, to the best of our knowledge, CL oscillatory zoning of an orthoclase-rich host
85 phase has never been reported in anhedral K-feldspar from equigranular granitic rocks.
86 Large K-feldspar megacrysts in granites grow at super-solidus conditions in a
87 low-temperature magma chamber (e.g., [Moore and Sisson, 2008](#)) or even at sub-solidus
88 conditions (e.g., [Vernon and Paterson, 2008](#)), and large K-feldspar phenocrysts crystallize
89 in a high-temperature magma chamber ([Vernon, 1986](#)). For the formations of megacrysts
90 and phenocrysts, wide space between previously crystallized minerals is required.

91 Conversely, the anhedral magmatic K-feldspar in equigranular granite studied here grew in
92 limited space between previously crystallized minerals. Thus, the environment (and stage)
93 at which anhedral K-feldspar grows in equigranular granite differs from that of K-feldspar
94 megacrysts and phenocryst formation. In this work, we used CL imaging to observe
95 oscillatory zoning of the orthoclase-rich host phase in anhedral perthitic K-feldspar from
96 the equigranular Toki granite, central Japan. This study reports on: 1) characterization of
97 the CL patterns of anhedral magmatic K-feldspar from the Toki granite to gain insights on
98 its nucleation and growth, and 2) characterization of the CL patterns of post-crystallization
99 textures such as veins, microperthite, and patchperthite, which enhances our understanding
100 of the textural development during the post-crystallization stage.

101

102

The Toki granite

103 The Toki granite in the Tono district of central Japan is one of the Late Cretaceous plutonic
104 bodies of the Inner Zone of the Southwest Japan Arc (Fig. 1A; Ishihara and Chappell, 2007).
105 The Toki granite is a $\sim 14 \times 12 \text{ km}^2$ stock (Ishihara and Suzuki, 1969) that intruded into the
106 Jurassic sedimentary rocks of the Kamiaso Formation in the Mino Terrane (Sano et al.,
107 1992) and into the Late Cretaceous Nohi rhyolite (Sonehara and Harayama, 2007; Fig. 1B).
108 The equigranular Toki granite is a concentrically zoned pluton, which is characterized by
109 three rock facies grading from muscovite–biotite granite (MBG) at the margin through
110 hornblende–biotite granite (HBG) in the interior to biotite granite (BG) in the core (Fig.
111 1C). Studies by Shibata and Ishihara (1979) and Suzuki and Adachi (1998) revealed that the
112 Toki granite has a whole-rock Rb–Sr isochron age of $72.3 \pm 3.9 \text{ Ma}$ and a monazite
113 chemical Th–U–total Pb isochron (CHIME) age of $68.3 \pm 1.8 \text{ Ma}$. Thermochronological
114 data have been reported for samples collected at multiple sampling sites within the Toki
115 granite: zircon U–Pb ages ranging from 74.7 ± 4.2 to $70.4 \pm 1.7 \text{ Ma}$ ($N = 14$: Yuguchi et al.,
116 2016; 2019), biotite K–Ar ages from 78.5 ± 3.9 to $59.7 \pm 1.5 \text{ Ma}$ ($N = 33$: Yuguchi et al.,
117 2011c), zircon fission-track ages from 75.6 ± 3.3 to $52.8 \pm 2.6 \text{ Ma}$ ($N = 47$: Yuguchi et al.,
118 2011c; 2019), and apatite fission-track ages from 52.1 ± 2.8 to $37.1 \pm 3.6 \text{ Ma}$ ($N = 33$:
119 Yamasaki and Umeda, 2012; Yuguchi et al., 2017). Such thermochronological data allowed
120 for estimation of position-by-position cooling (time–temperature) paths within the Toki
121 granite (Yuguchi et al., 2019). The extent of microperthite and patchperthite growth can be
122 an indicator of the cooling process of the Toki granite (Yuguchi et al., 2011a, b). The
123 systematic variation in development of microperthite indicates that the Toki granite cooled

124 effectively from the paleo-roof boundary (roof boundary at the time of granitic magma
125 intrusion) during the exsolution coarsening stage (690–780 °C) (Yuguchi et al., 2011a),
126 while those of patchperthite indicate that the Toki granite cooled effectively from the
127 paleo-roof boundary and from the western margin during the deuteritic coarsening stage (<
128 500 °C) (Yuguchi et al., 2011b). The geology and petrography of the Toki granite have been
129 described in detail by Yuguchi et al. (2010; 2011a, b, and c; 2020).

130

131 **Sampling and analytical procedures**

132 Nine samples were collected from six boreholes in all three rock facies in the Toki granite
133 for petrographic observations and analyses of CL patterns (Fig. 1C): samples No. 1
134 (DH6-28), 2 (DH10-14), and 4 (DH13-8) are from the MBG lithofacies; samples No. 5
135 (DH13-15), 6 (MIU2-13), and 9 (MIZ1-17) are from the HBG; and samples No. 3
136 (DH11-31), 7 (MIU2-34), and 8 (MIZ1-06) are from the BG lithofacies (Table S1). The
137 petrographic data used in this study were obtained from thin sections. Thin sections were
138 prepared for electron microprobe analysis by polishing with progressively finer diamond
139 pastes (particle sizes of 3 μm , 1 μm , and 0.25 μm) to remove any surface irregularities that
140 can seriously affect the CL imaging quality (Frelinger et al., 2015). Thin sections with
141 thicknesses of 30–40 μm were produced to obtain mineral chemistry using the electron
142 probe micro-analyzer (EPMA).

143 CL images were generated using a scanning electron microscope (SEM)
144 cathodoluminescence. BSE and CL images were obtained using a JEOL IT100A SEM
145 equipped with a Gatan mini-CL detector at Yamagata University, Japan, which was
146 operated at an accelerating voltage of 15 kV and a beam current of 1.0 nA. The SEM-CL
147 technique is well-suited for detailed CL imaging due to its high resolution and
148 magnification capabilities, which allow for high-quality imaging of distinct CL textures
149 compared to optical-CL imaging (Frelinger et al., 2015). The intensity of the SEM-CL
150 emission depends on the variations in the internal chemistry and structure of a K-feldspar
151 crystal. The variations in the luminescence depend on the differences in minor
152 concentrations of titanium in K-feldspar, which acts as a CL activator (Lee et al., 2007;

153 Parsons et al., 2008; Kayama et al., 2010). Slaby et al. (2008) described that the CL
154 intensity difference in K-feldspar is controlled by the density of Al-O⁻-Al structural defects
155 resulting from the coupled substitution of Ba²⁺ for K⁺ and Al³⁺ for Si⁴⁺. Therefore, the
156 intensities of the SEM-CL emissions can be used to interpret the element partitioning
157 between the melt and crystal during the mineral growth process.

158 Mineral element compositions and elemental maps were obtained using an EPMA (JEOL
159 JXA-8900) with a wavelength-dispersive X-ray spectrometer (WDS) at Yamagata
160 University. The analytical conditions for the quantitative determination of mineral
161 compositions were as follows: an acceleration voltage of 15 kV, a beam current of 10 nA, a
162 beam diameter of 3 μm, and the ZAF data correction method. The respective peak and
163 background counting times were 200 s and 100 s for Ti (PET crystals) and Fe (LIF); 100 s
164 and 50 s for Sr (TAP) and Rb (PET); 60 s and 30 s for Mn (LIF), Mg (TAP), and Ba (PET);
165 and 10 s and 5 s for Si (TAP), Al (TAP), Ca (PET), Na (TAP), and K (PET). WDS
166 elemental maps were obtained using the following operating conditions: 15 kV acceleration
167 voltage, 20 nA beam current, and 200 ms dwell time per spot. Distributions of silicon,
168 titanium, aluminum, iron, manganese, magnesium, calcium, sodium, potassium, barium,
169 lead, and strontium were analyzed during WDS elemental mapping.

170

Petrography and mineralogy

171 Petrography of the samples

172 The major mineral assemblage of the MBG consists of quartz + plagioclase + K-feldspar +
173 biotite + muscovite; that of the HBG includes quartz + plagioclase + K-feldspar + biotite +
174 hornblende ± muscovite; and that of the BG consists of quartz + plagioclase + K-feldspar +
175 biotite. The boundaries of the three lithofacies are defined by the appearance (MBG/HBG)
176 and disappearance (HBG/BG) of hornblende (Yuguchi et al., 2010). All studied samples
177 contain K-feldspar crystals. The modes of quartz, plagioclase, K-feldspar, and biotite in
178 each thin section are 20–41, 19–36, 27–51, and 0–7 vol%, respectively, corresponding to
179 monzogranite, granite, and granodiorite (see Fig. 7 in Yuguchi et al., 2010). Plagioclase
180 occurs as subhedral to euhedral 1,000–20,000 µm-long crystals, quartz as anhedral to
181 euhedral 500–25,000 µm-long crystals, and K-feldspar as anhedral 1,000–12,000 µm-long
182 crystals. Biotite has undergone variable degrees of alteration and is partially or completely
183 replaced by chlorite.

184

185 K-feldspar

186 All examined K-feldspar grains are anhedral ($N = 54$, Fig. 2A-1 and 2B-1), showing
187 microperthite and patchperthite textures (Figs. 2C-1, 2D-1, 3A-1, and 3B-1). The albite-rich
188 lamellae in microperthite are thinner and more elongate than albite-rich phases in
189 patchperthite. The interface between the albite-rich lamellae and host phase in
190 microperthite is sharp, whereas the interface in patchperthite is irregular. Albite-rich phases
191 of patchperthite occur in either bead or patch form (Fig. 3A-1 and 3B-1). Hashimoto et al.

192 (2005) suggested that an exsolution reaction in alkali feldspar is responsible for the
193 formation of microperthite with micrometer-scale albite-rich phases and cryptoperthite with
194 nanometer-scale albite-rich phases. Patchperthite was formed by dissolution and
195 reprecipitation during hydrothermal alteration (Worden et al., 1990). The formation
196 conditions of microperthite and patchperthite correspond to the exsolution coarsening stage
197 (690–780 °C) and deuteric dissolution–precipitation coarsening stage (<500 °C),
198 respectively (Yuguchi et al., 2011A; 2011B). Patchperthite formed later than microperthite
199 during sub-solidus cooling.

200

201

Results and discussion

202 **Characterization of CL patterns of magmatic K-feldspar crystals**

203 In this study, CL images of fifty-four K-feldspar crystals were obtained. The CL patterns of
204 the K-feldspar crystals can be broadly divided into two categories based on the presence or
205 absence of oscillatory zoning. Oscillatory zoning was observed in forty-two K-feldspar
206 crystals as the cyclic alternation of dark and bright bands of luminescence (Fig. 2).
207 Oscillatory zoning can be further categorized into two types: single crystal core oscillatory
208 zoning (SCOZ; Fig. 2) and multiple crystal core oscillatory zoning (MCOZ; Fig. 3).

209

210 **Single core oscillatory zoning.** The SCOZ (Fig. 2) is defined as oscillatory zoning
211 defining a single crystal core within K-feldspar crystals in the CL image. Thirty-seven
212 K-feldspar crystals with major axes longer than $\sim 1,000$ μm show SCOZ patterns (Fig. 2).
213 The SCOZ exhibits linear and curved boundaries between luminescence bands (Fig. 2). The
214 SCOZ pattern is the most abundant in K-feldspars from the BG lithofacies: fifteen grains in
215 the observed K-feldspars from the BG (total K-feldspars are $N = 19$), nine grains from the
216 HBG (total $N = 16$), and nine grains from the MBG (total $N = 19$).

217 Based on the analysis of the CL pattern, the nucleation and crystal growth behavior of
218 K-feldspar can be determined. The crystal cores reflect the nucleus of the K-feldspar. For
219 example, the crystal cores of K-feldspar grains No. 3 (DH11-31)-4, 9 (MIZ1-17)-7, and 3
220 (DH11-31)-5 are shown in Fig. 2A-3, B-3, and C-3, respectively. The luminescence
221 intensities of the crystal cores of the K-feldspar crystals vary. Cores with high-intensity
222 luminescence are observed in grains No. 3 (DH11-31)-4, 9 (MIZ1-17)-7, and 3

223 (DH11-31)-5 (Fig. 2A-2, B-2, and C-2) and low-luminescence cores are identified in grain
224 No. 3 (DH11-31)-7 (Fig. 2D-2). The difference in luminescence intensities of crystal cores
225 may be due to different sectioning locations of each K-feldspar crystal during the
226 preparation of thin sections. As oscillatory zoning consists of the multiple growth shells in
227 three dimensions (Shore and Fowler, 1996), such sectioning effects can influence the
228 evaluation of the oscillatory zoning within the mineral. While the geometric core of
229 K-feldspar grain No. 9 (MIZ1-17)-7 corresponds to the crystal core (Fig. 2B), the geometric
230 cores of grains No. 3 (DH11-31)-4, 3 (DH11-31)-5, and 3 (DH11-31)-7 do not correspond
231 to the crystal cores (Fig. 2A, C, and D). The crystals grew within the melt until they
232 intercepted previously crystallized minerals. For K-feldspar grain No. 3 (DH11-31)-7, the
233 crystal grew along the X and Y directions on the observation surface of the thin section (Fig.
234 2D-3). The growth of the crystal along the Y-direction was stopped by a euhedral quartz,
235 while the growth along the X-direction continued. The outer rim of the oscillatory zoning
236 away from the crystal core is not well defined. The outside region around the K-feldspar
237 nucleus in grain No. 3 (DH11-31)-7, e.g., α region in Fig. 2D-3, does not show any
238 oscillatory zoning.

239

240 **Multiple core oscillatory zoning.** The MCOZ (Fig. 3) is defined as a pattern in which two
241 or more crystal cores are present in the K-feldspar grains. The K-feldspar crystal of grain
242 No. 4-1 is a coarse-grained crystal with a size of $3,210 \times 2,594 \mu\text{m}$, which shows an MCOZ
243 pattern with five cores in the CL image (Fig. 3A-2 and A-3). All five crystal cores visible in
244 CL show the same extinction angle under polarized light, indicating the same crystal

245 orientation; such characteristics cannot be explained by a simple assemblage of several
246 SCOZs. The MCOZ patterns are observed in five grains, all of which are relatively
247 coarse-grained crystals up to $\sim 3,000$ μm in length. The crystals with MCOZs exhibit
248 rounded boundaries between luminescence zonings (Fig. 3A-2 and B-2) and their patterns
249 contain two to five crystal cores within a grain (Fig. 3A-3 and B-3). The crystal cores in the
250 MCOZ exhibit low luminescence (Fig. 3A-2 and B-2). The existence of multiple crystal
251 cores in K-feldspar grains reveals multiple nuclei.

252

253 **Unzoned CL pattern.** The K-feldspar grains without zoning are characterized by
254 homogeneous luminescence across the grain (Fig. 4). These patterns do not have crystal
255 cores, that is, they cannot be used to determine any nuclei. Among studied samples, twelve
256 relatively small (less than ~ 700 μm long) grains lack any zoning. The K-feldspars with
257 unzoned CL patterns are the most abundant in the MBG among the three lithofacies in the
258 Toki granite: BG ($N = 2$), HBG ($N = 2$), and MBG ($N = 8$).

259

260 **CL intensity and chemistry of K-feldspar.**

261 In previous studies, variations in luminescence were explained by differences in either: 1)
262 minor concentrations of titanium in K-feldspar (Lee et al., 2007; Parsons et al., 2008;
263 Kayama et al., 2010), or 2) the density of Al-O⁻-Al structural defects (Słaby et al., 2008).
264 Figures 5 and S1 show the WDS elemental maps of SCOZ K-feldspar grains No. 3
265 (DH11-31)-4 and No. 9 (MIZ1-17)-7, respectively, which are illustrated by elemental
266 distributions of silicon, titanium, aluminum, calcium, sodium, potassium, and barium.

267 Comparing the WDS elemental maps and CL images, there are weak correlations between
268 titanium concentrations and CL intensity (Fig. 5A-1 and 5C). High barium concentration
269 areas correspond to high CL intensity areas and low barium areas correspond to low CL
270 intensity areas (Fig. 5A-1 and 5H). Figure 6 shows chemical variations in SiO₂, TiO₂, Al₂O₃,
271 Na₂O, K₂O, and BaO along the transect line from 0 μm (the core) to 1164 μm (the rim) in
272 the SCOZ K-feldspar grain No. 3 (DH11-31)-4. Chemical variations from cores to rims in
273 K-feldspars display high TiO₂ concentrations (> 0.01 wt%; Fig. 6C) and high BaO
274 concentrations (> 0.10 wt%; Fig. 6F) in high CL intensity areas, confirming a causal
275 relationship. The positive correlation between CL intensity and titanium concentration
276 confirms that titanium acts as a CL activator. Barium incorporation causes local structural
277 distortions due to coupled KSi–BaAl exchange (Viswanathan and Brandt, 1980;
278 Viswanathan and Kielhorn, 1983). The KSi–BaAl exchange causes aluminum
279 rearrangement over the tetrahedral sites in K-feldspar, and this rearrangement increases
280 structural disorder and the density of Al-O⁻-Al structural defects (Viswanathan and
281 Kielhorn, 1983; Słaby et al., 2008). In the K-feldspar grain No. 3 (DH11-31)-4, chemical
282 variations in high CL intensity areas show a gradual decrease in Al₂O₃ and BaO and a
283 gradual increase in SiO₂ and K₂O along the transect line from 167 μm to 470 μm (Fig. 6A,
284 C, E, and F), which supports the assumption of the KSi–BaAl exchange in K-feldspar. Thus,
285 CL variations are related to variations in the density of the Al-O⁻-Al structural defects. In
286 summary, the CL intensity differences in K-feldspar can be attributed to the following
287 factors: 1) variations in concentrations of titanium that acts as a CL activator, and 2)
288 variations in the density of Al-O⁻-Al structural defects.

289

290 **CL patterns of post-crystallization textures of K-feldspar.**

291 In grain No. 8 (MIZ1-06)-2, a fine vein composed of K-feldspar and biotite cuts through a
292 large K-feldspar grain with the SCOZ pattern (parts between arrows in [Fig. 7A](#)). The fine
293 vein within the K-feldspar is barely identifiable in the BSE image because the vein and its
294 host K-feldspar have almost identical chemical compositions ([Fig. 7A](#)). In contrast, in the
295 CL image, the vein characteristics, e.g., width, direction, and length, are readily visible,
296 with the vein displaying lower luminescence compared to the host K-feldspar grain ([Fig.](#)
297 [7B](#)).

298 The luminescence intensities of most albite-rich phases in microperthite and
299 patchperthite are lower than those of the orthoclase-rich host phases ([Fig. 8A-2, B-2, and](#)
300 [C-2](#)). WDS elemental maps of the SCOZ K-feldspar grain No. 2 (DH10-14)-5 ([Fig. 9](#)) and
301 chemical compositions of albite-rich and orthoclase-rich phases in patchperthite ([Table S2](#))
302 indicate that the titanium and barium concentrations of the albite-rich phases are lower than
303 those of the orthoclase-rich host phases. [Figure 8A-2, B-2, and C-2](#) (white arrows) show
304 that the low-luminescence albite-rich phases cut across the CL bands in the regions with
305 oscillatory zoning. This indicates that the oscillatory zoning in the orthoclase-rich host
306 phase was not perturbed by the formation of microperthite and patchperthite in the
307 post-crystallization stage. In grain No. 2 (DH10-14)-5 ([Fig. 8C-2](#)), the contrast between the
308 albite-rich phases and the orthoclase-rich host phase in patchperthite (β region) is stronger
309 than that in microperthite (γ region). This means that the luminescence intensity of the
310 albite-rich phase in patchperthite is lower than that in microperthite. WDS elemental maps

311 presented in [Figure 9](#) and chemical compositions of the albite-rich phases in microperthite
312 and patchperthite ([Table S2](#)) show that the titanium and barium concentrations of albite-rich
313 phases in patchperthite are lower than those in microperthite. An exception is that the
314 albite-rich phase in patchperthite in the region without oscillatory zoning shows higher
315 luminescence intensity than the host phase (e.g., α region of grain No. 3 (DH11-31)-7; [Fig.](#)
316 [2D-3](#)). WDS elemental maps of the α region in patchperthite display: 1) high calcium
317 concentrations of the albite-rich phases relative to the orthoclase-rich host phases, and 2)
318 homogeneous distribution of titanium, aluminium, and barium in the albite-rich and
319 orthoclase-rich host phases ([Fig. S2](#)). Although the albite-rich phases with low
320 luminescence are accompanied with high concentrations of calcium and aluminium ([Fig. 9](#)),
321 the albite-rich phase with high luminescence in the α region is characterized by constant
322 aluminium and increased calcium concentrations ([Fig. S2](#)). Thus, the high CL intensity of
323 the albite-rich phase may be derived from Al-O⁻-Al structural defects resulting from
324 calcium incorporation. Mass transfer, that is, titanium and barium transfers occurred during
325 the formation of microperthite and patchperthite. Therefore, the difference in the titanium
326 and barium concentrations in the albite-rich phase between microperthite and patchperthite
327 indicates the difference in their mass transfers between exsolution coarsening and deuteric
328 dissolution–precipitation coarsening. In the exsolution coarsening stage (690–780 °C:
329 [Yuguchi et al., 2011a](#)) when the microperthite was produced, interdiffusion between sodium
330 and potassium essentially occurred within magmatic K-feldspar, resulting in a small
331 amount of titanium and barium that diffused from the original distribution within the
332 K-feldspar. Conversely, in the deuteric dissolution–precipitation coarsening stage (<

333 500 °C: [Yuguchi et al., 2011b](#)), titanium and barium were released from K-feldspar into the
334 fluid, and subsequent precipitation with low titanium and barium resulted in low
335 luminescence.

336

337

Implications

338 The CL analyses of perthitic anhedral K-feldspars in the equigranular Toki granite provide
339 new insights into the nucleation and growth of magmatic K-feldspar crystals and their
340 textural development of K-feldspar during the post-crystallization stage. Previous CL
341 studies presented oscillatory zoning in K-feldspar megacrysts and phenocrysts in granitic
342 rocks. It is known that K-feldspar megacrysts crystallize at super-solidus conditions in a
343 low-temperature magma chamber or at sub-solidus conditions in granite and that K-feldspar
344 phenocrysts crystallize in a high-temperature magma chamber. They grow in wide space
345 between previously crystallized minerals. Anhedral K-feldspars crystallize at super-solidus
346 conditions in a low-temperature magma chamber and grow in limited space between
347 previously crystallized minerals. Oscillatory zoning is common in anhedral K-feldspar of
348 equigranular granite but it may have been overlooked in previous studies. In the CL
349 analyses, the features of nucleation and crystal growth in the limited space (anhedral
350 K-feldspar) enable us to compare with those in the wide space (K-feldspar megacryst and
351 phenocryst). For example, new insights from oscillatory zoning in the anhedral K-feldspar
352 include 1) the discordance between geometric cores and crystal cores in the CL image and
353 2) multiple nuclei deduced from the existence of multiple crystal cores, which are
354 uncommon in the megacryst and phenocryst studies. The detailed CL analyses, not only for
355 anhedral K-feldspar crystals in the equigranular granite but also for K-feldspar
356 phenocryst/megacrysts in porphyritic granite, provide insights into the nucleation and
357 crystal growth processes of K-feldspar through the several genetic environments and
358 thermal stages. Furthermore, the relationship between growth texture (oscillatory zoning)

359 and chemistry reveals the spatial extent of the element partitioning between the melt and
360 crystal and of the mass transfer from the melt into crystal during the magma evolution. The
361 observed CL patterns can also be used to reveal the textural development in K-feldspar
362 during the post-crystallization stage.
363

364

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487 **Figure captions**

488 **Figure 1.** The Toki granite. (A) Map of Southwest Japan showing the location of the Toki
489 granite (square symbol) in central Japan and the distribution of the San-in, Sanyo, and
490 Ryoke belts in the inner zone of Southwest Japan (modified after [Ishihara and Chappell,](#)
491 [2007](#)). (B) Geologic map of the Toki granite showing the borehole sites (modified after
492 [Itoigawa, 1980](#)). The topographic contours in the Tono district are based on topographic
493 maps of the Geographical Survey Institute (1:25,000) titled “Mitake,” “Takenami,”
494 “Toki,” and “Mizunami”. Borehole investigations of the Toki granite were performed by
495 the Japan Atomic Energy Agency ([Japan Nuclear Cycle Development Institute 2000,](#)
496 [2002](#)). (C) Rock facies cross-section of the Toki granite along the X to X’ transect of the
497 geologic map ([Fig. 1B](#)). MBG: muscovite–biotite granite, HBG: hornblende–biotite
498 granite, and BG: biotite granite ([Yuguchi et al. 2010](#)).

499
500 **Figure 2.** Backscattered electron (BSE: A-1, B-1, C-1, and D-1) and cathodoluminescence
501 (CL: A-2, B-2, C-2, and D-2) images of K-feldspar crystals showing SCOZ patterns (A:
502 grain No. 3 (DH11-31)-4, B: grain No. 9 (MIZ1-17)-7, C: grain No. 3 (DH11-31)-5, and
503 D: grain No. 3 (DH11-31)-7). The CL images reveal the crystal core parts displayed in
504 CL images (crystal core: A-3, B-3, and C-3) and crystal growth direction (D-3). If the
505 nucleus of K-feldspar crystallization in grain No. 3 (DH11-31)-7 is defined as the origin
506 of growth, “0 μm ”, the regions beyond 480 μm in the X-direction and 270 μm in the
507 Y-direction do not show oscillatory zoning (α region) (2D-3).

508

509 **Figure 3.** Backscattered electron (BSE: A-1 and B-1) and cathodoluminescence (CL: A-2
510 and B-2) images of K-feldspar crystals showing MCOZ patterns (A: grain No. 4
511 (DH13-8)-1, and B: grain No. 4 (DH13-8)-3). The CL images reveal the nuclei displayed
512 in CL images (crystal core: A-3 and B-3).

513

514 **Figure 4.** Backscattered electron (BSE: A-1 and B-1) and cathodoluminescence (CL: A-2
515 and B-2) images of K-feldspar crystals showing unzoned CL patterns (A: grain No. 1
516 (DH6-28)-1, and B: grain No. 1 (DH6-28)-7).

517

518 **Figure 5.** Backscattered electron image (A-1), cathodoluminescence image (A-2) and WDS
519 elemental maps showing silicon (B), titanium (C), aluminum (D), calcium (E), sodium
520 (F), potassium (G), and barium (H) of a SCOZ K-feldspar grain No. 3 (DH11-31)-4.
521 WDS elemental maps were obtained with 3 μm step size at a resolution of up to 300 \times
522 300 pixels.

523

524 **Figure 6.** WDS linescans of SiO_2 (A), TiO_2 (B), Al_2O_3 (C), Na_2O (D), K_2O (E), and BaO
525 (F) along the scanning lines in BSE and CL images. Scanning line from 0 μm to 1164 μm
526 in the SCOZ K-feldspar grain No. 3 (DH11-31)-4.

527

528 **Figure 7.** Backscattered electron (A) and cathodoluminescence (B) images of K-feldspar
529 crystals (grain No. 8 (MIZ1-06)-2) with veins in the SCOZ (between arrows).

530

531 **Figure 8.** Backscattered electron (BSE: A-1, B-1, and C-1) and cathodoluminescence (CL:
532 A-2, B-2, and C-2) images of K-feldspar crystals showing the distribution of albite-rich
533 lamellae in microperthite and patchperthite (A: grain No. 6 (MIU2-13)-3, B: grain No. 5
534 (DH13-15)-8, and C: grain No. 2 (DH10-14)-5).

535

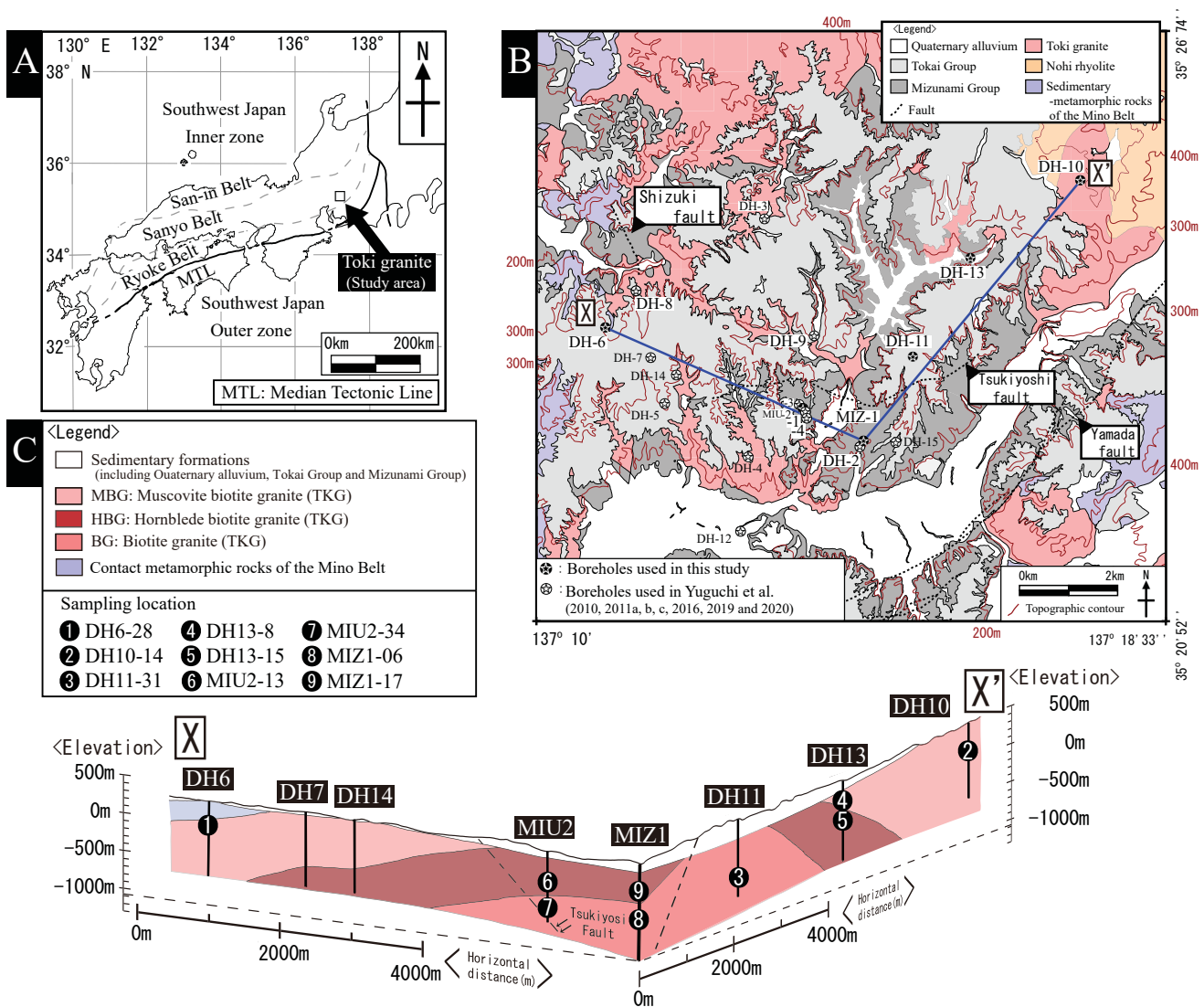
536 **Figure 9.** Backscattered electron image (A-1), cathodoluminescence image (A-2), and
537 WDS elemental maps showing silicon (B), titanium (C), aluminum (D), calcium (E),
538 sodium (F), potassium (G), and barium (H) of a SCOZ K-feldspar grain No. 2
539 (DH10-14)-5. WDS elemental maps were obtained with 4 μm step size and a resolution
540 up to 260×260 pixels.

541

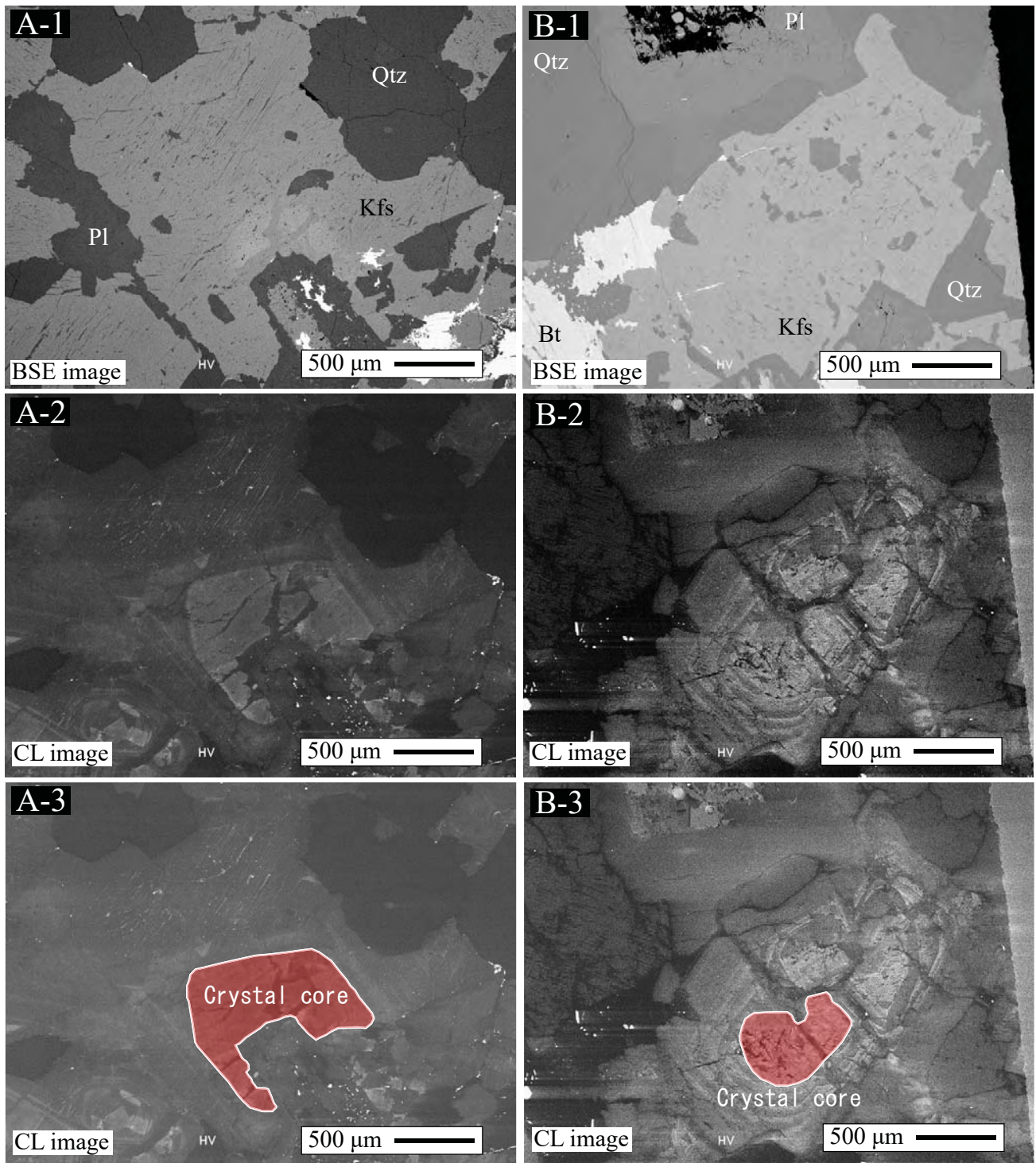
542 **Figure S1.** Backscattered electron image (A-1), cathodoluminescence image (A-2), and
543 WDS elemental maps showing silicon (B), titanium (C), aluminum (D), calcium (E),
544 sodium (F), potassium (G), and barium (H) of a SCOZ K-feldspar grain No. 9
545 (MIZ1-17)-7. WDS elemental maps were obtained with a 5 μm step size and a resolution
546 up to 200×200 pixels.

547

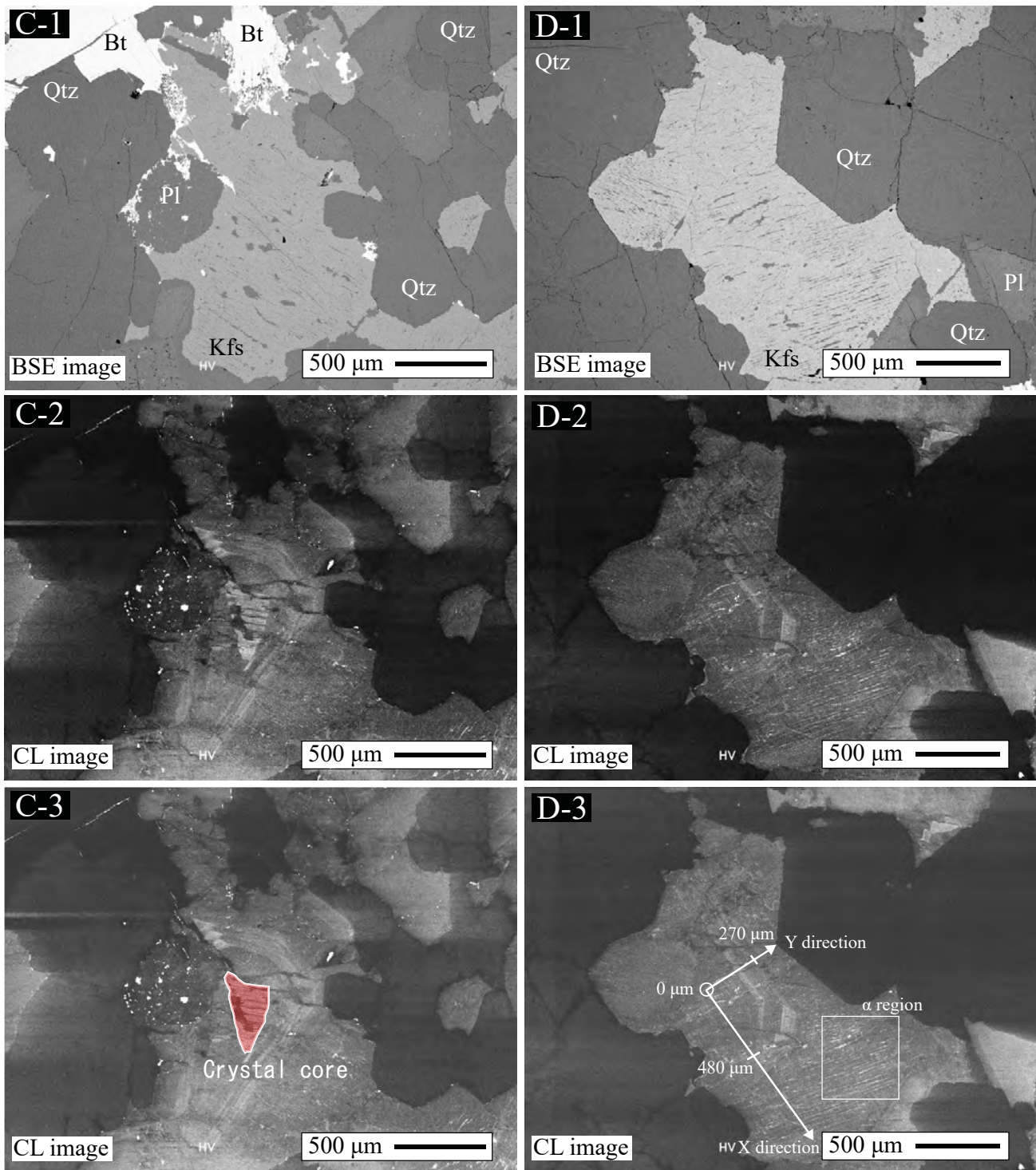
548 **Figure S2.** Backscattered electron image (A-1), cathodoluminescence image (A-2), and
549 WDS elemental maps showing silicon (B), titanium (C), aluminum (D), calcium (E),
550 sodium (F), potassium (G), and barium (H) of a SCOZ K-feldspar grain No.
551 3(DH11-31)-7. WDS elemental maps were obtained with a 3 μm step size and a
552 resolution up to 150×150 pixels.



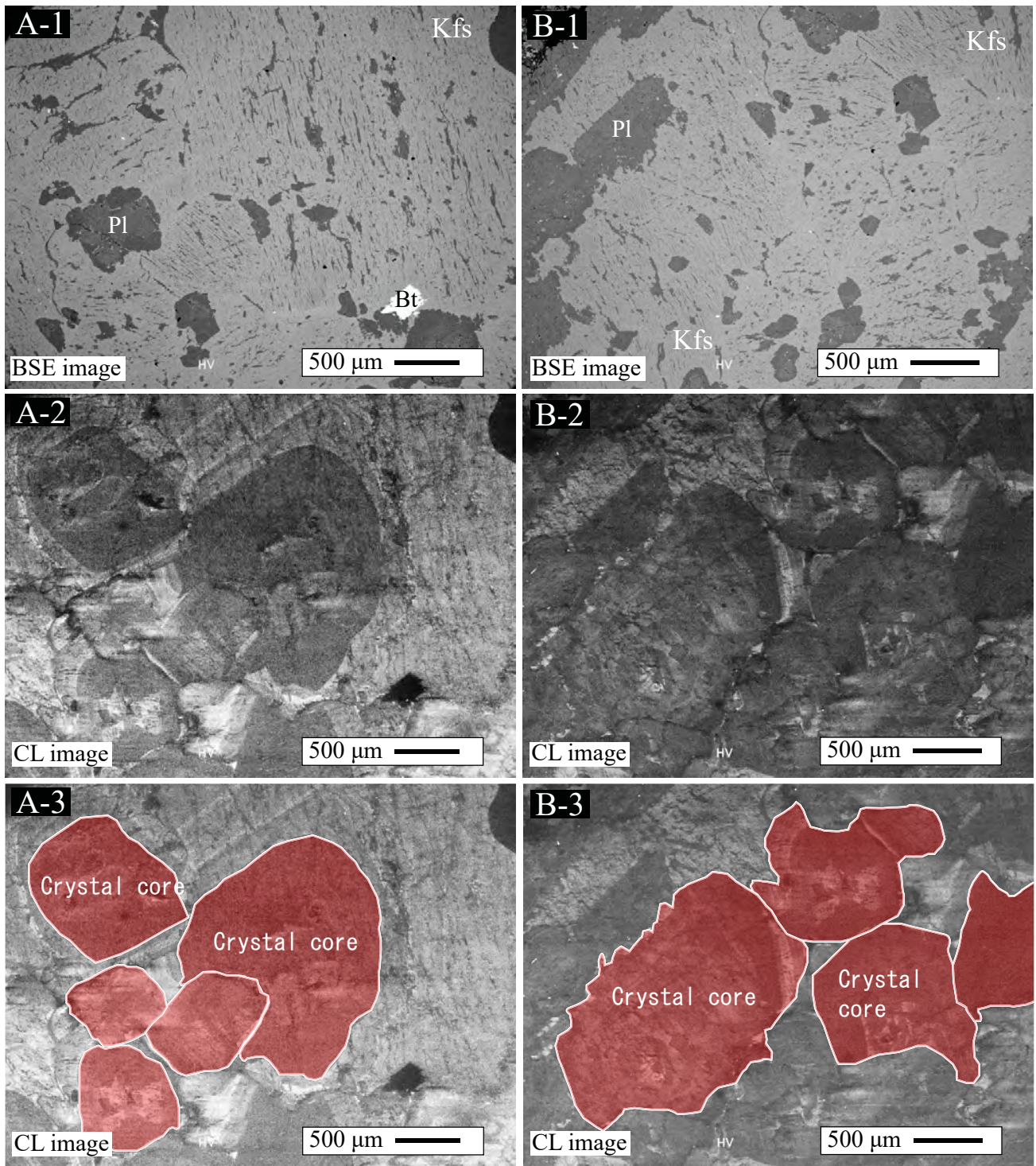
Yuguchi et al. Fig. 1



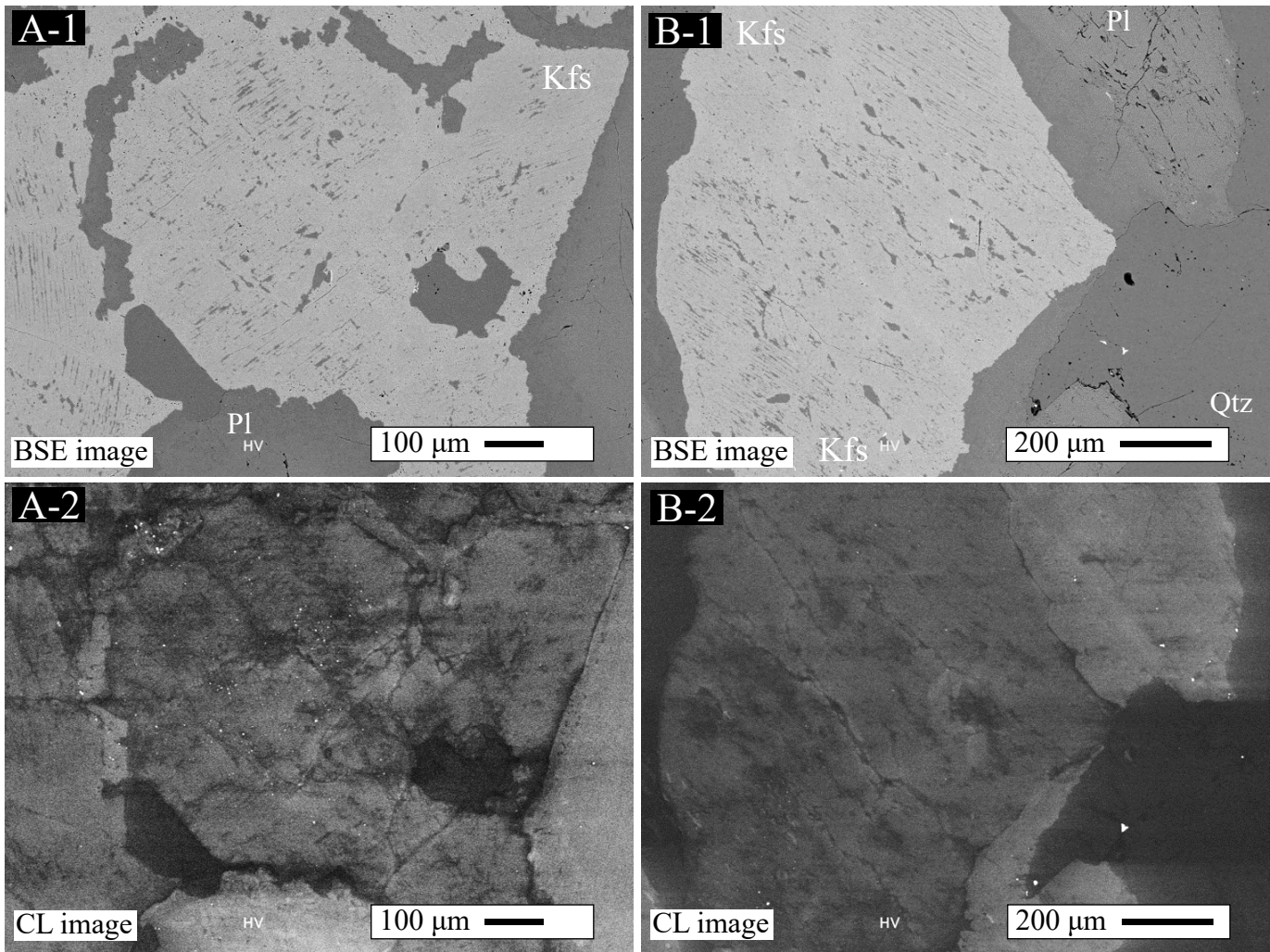
Yuguchi et al. Fig. 2



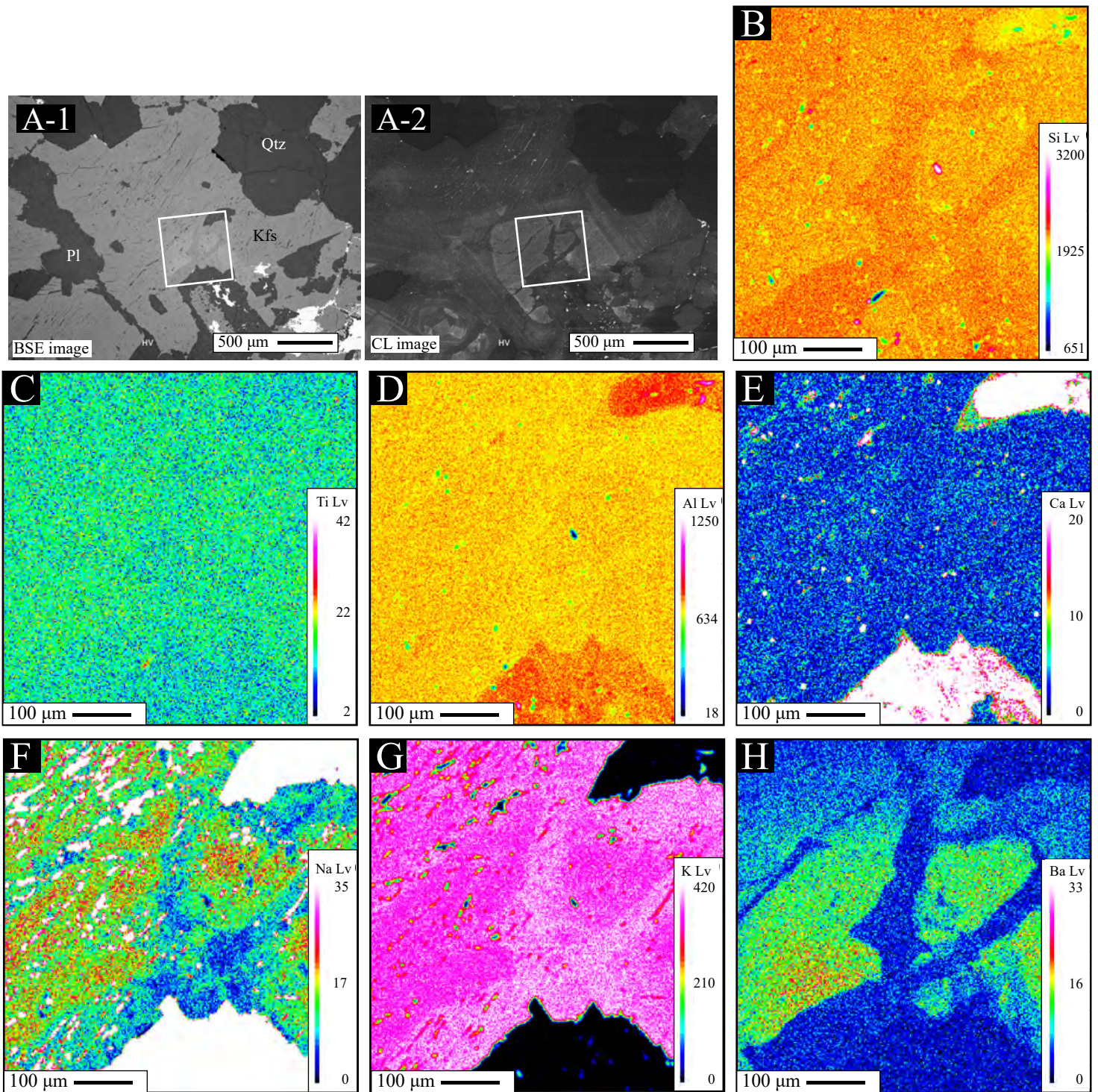
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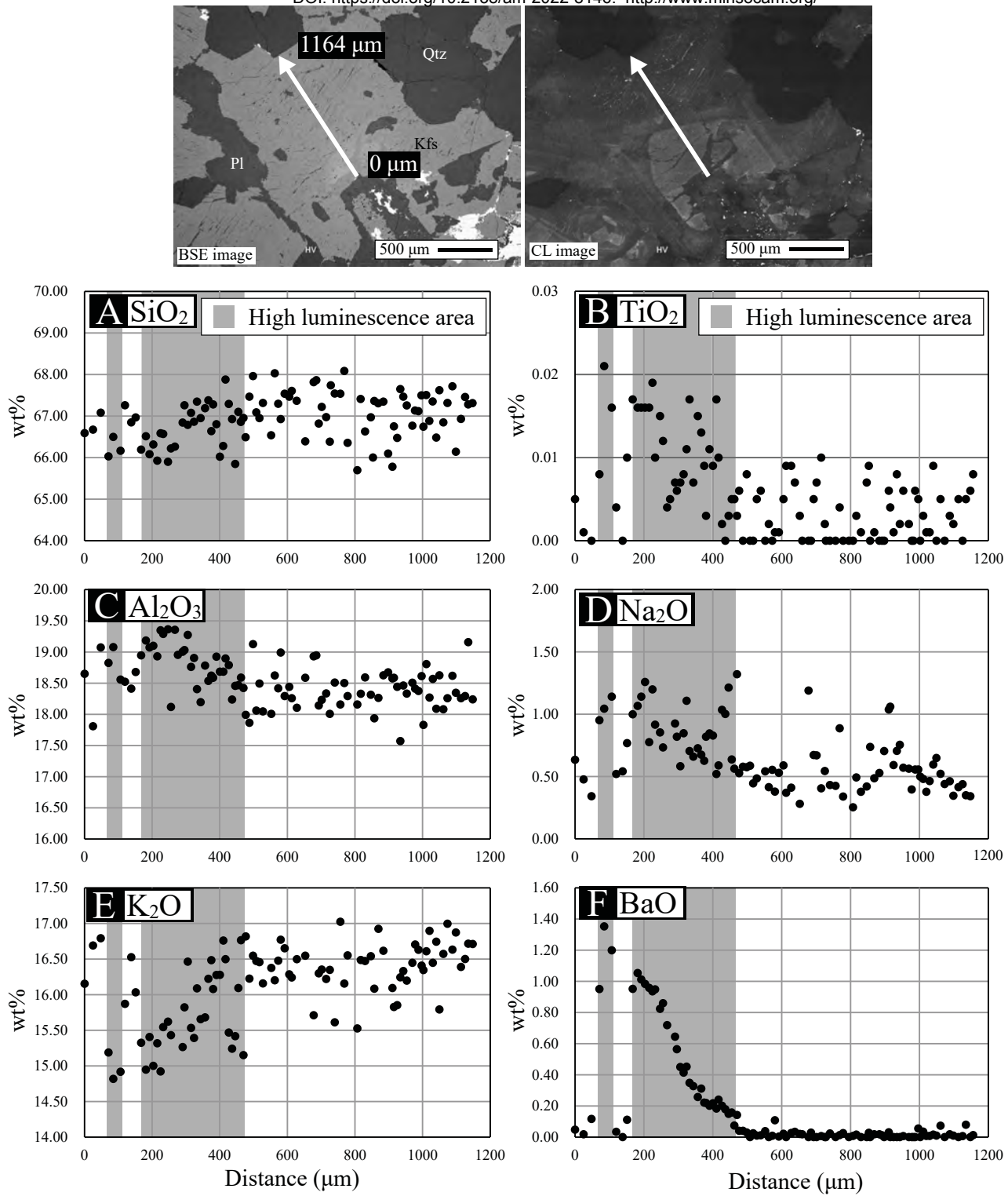
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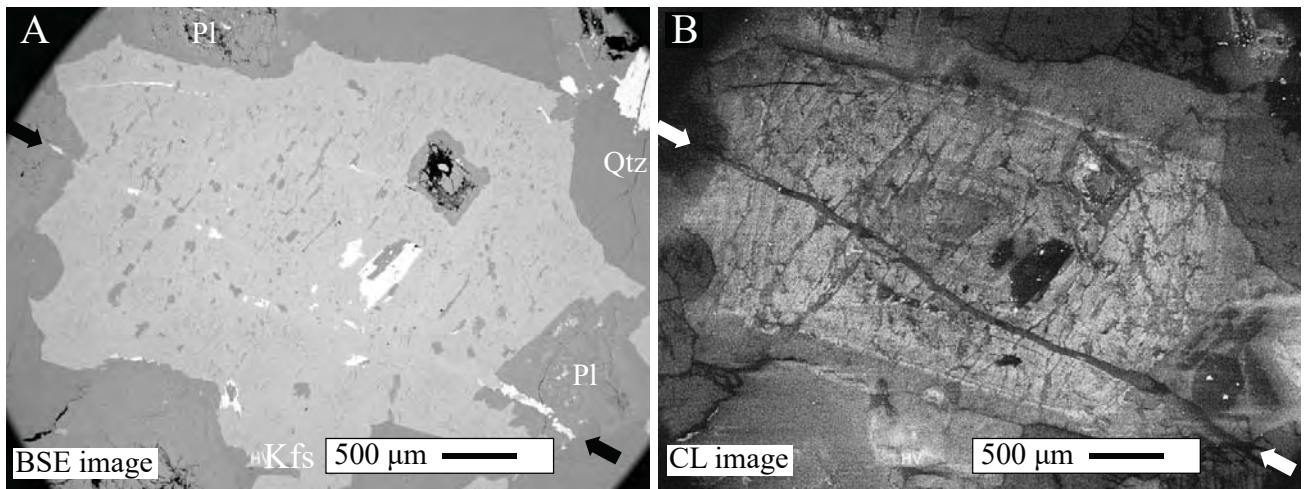


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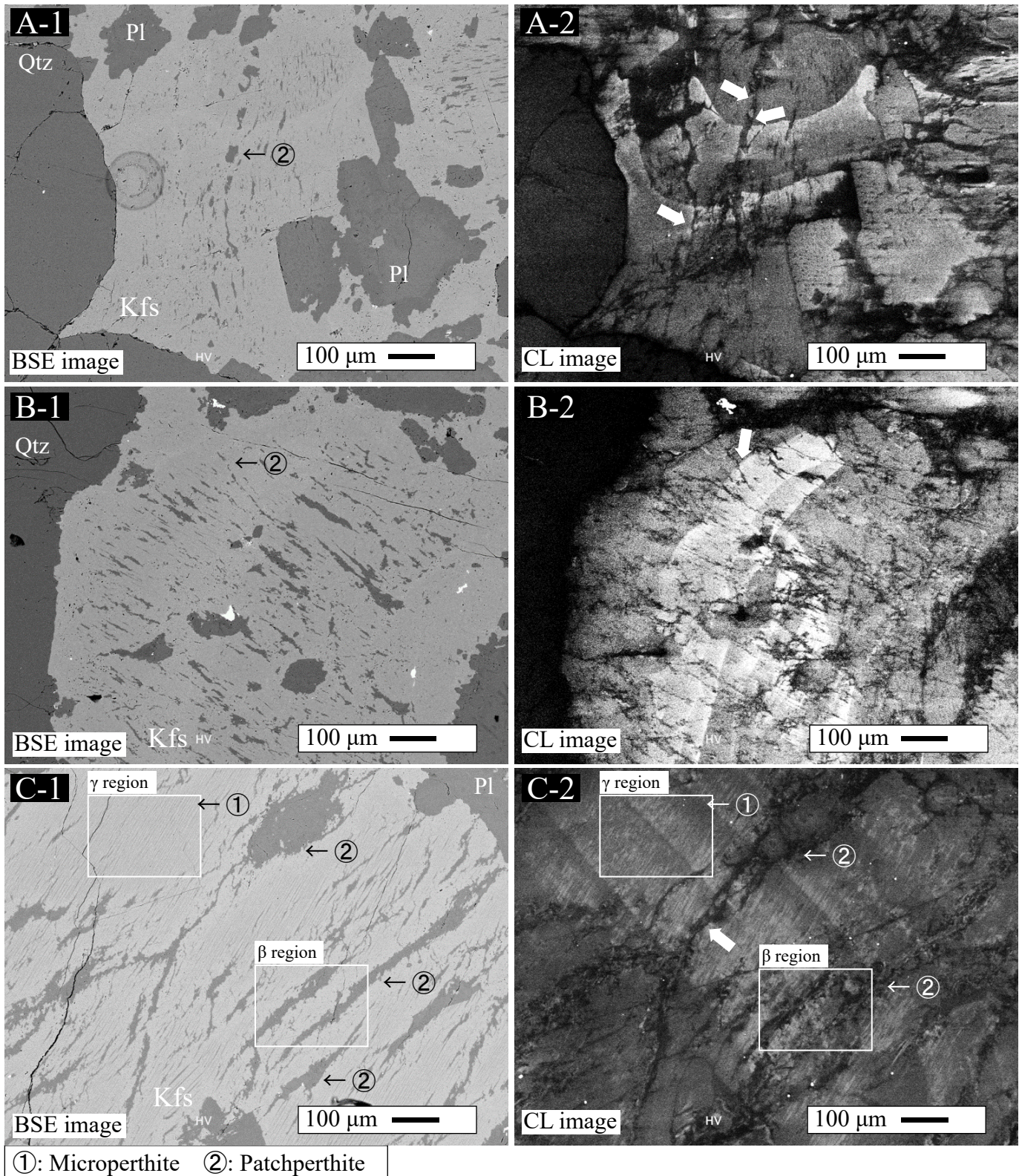


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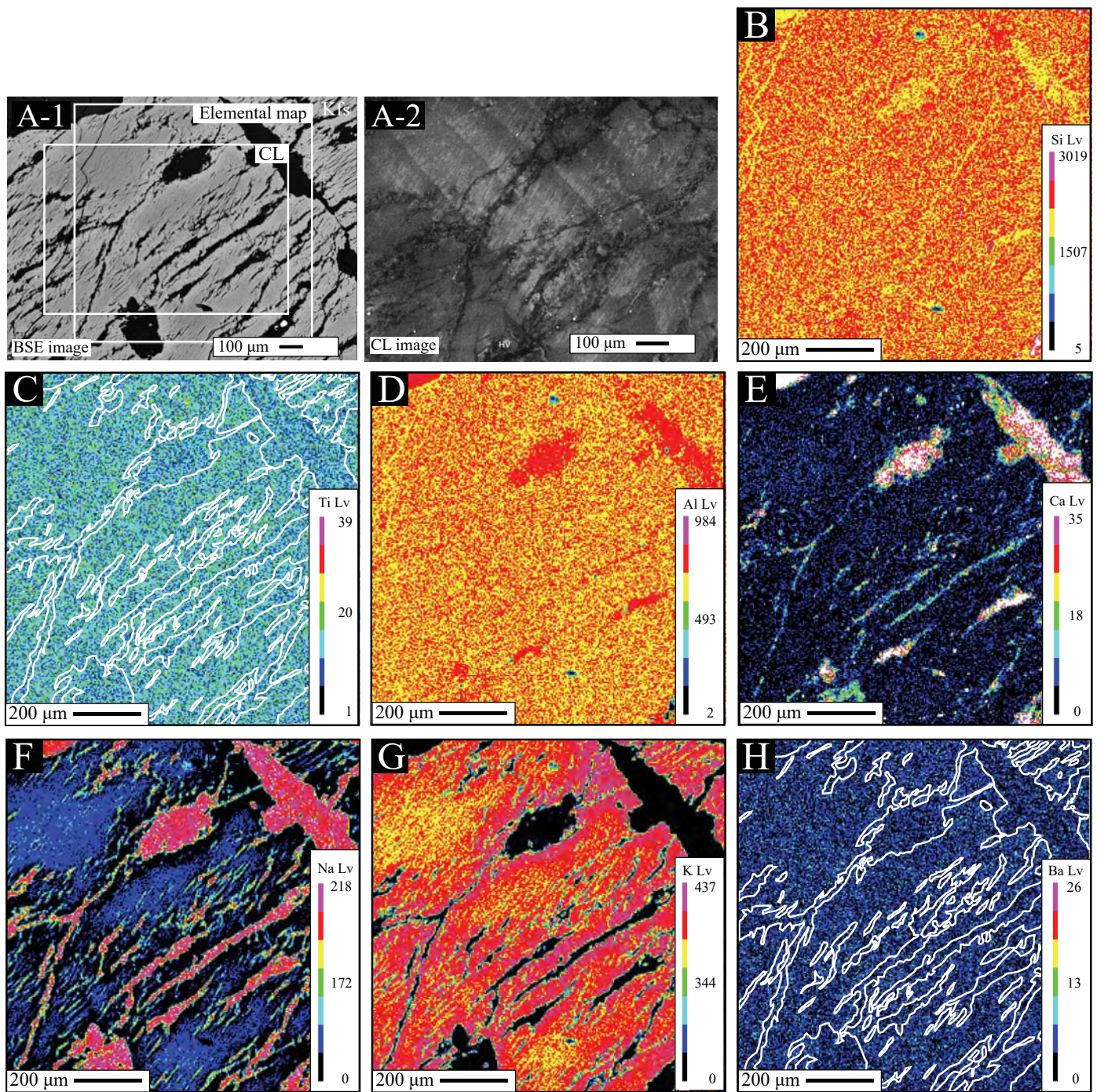




Yuguchi et al. Fig. 7



Yuguchi et al. Fig. 8



Yuguchi et al. Fig. 9