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1 **Microstructural records of multiple retrograde local H<sub>2</sub>O supplement in the pelitic**  
2 **gneiss, Lützow-Holm Complex at Akarui Point, East Antarctica**

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**ABSTRACT**

The alkali-feldspar and biotite in the sillimanite-biotite-garnet gneiss from East Antarctica preserves characteristic microstructural evidence of multi-stage H<sub>2</sub>O supplement during the retrograde metamorphism. The first microstructural evidence is the ‘zoned feldspar’, in which the mesoperthitic zone, the anti-perthitic zone and lamella-free plagioclase zone coexist within a single crystal. They are occasionally found next to biotite, and are always depleted in orthoclase (Or) component toward the biotite. The formation process of this microstructure could be explained by the diffusion that oversteps the solvus. The second microstructural evidence is the serrate boundary between alkali-feldspar and biotite. The projections of biotite are selectively developed next to Or lamellae of alkali-feldspar every 3-5 μm. These two microstructures would have formed as the biotite grew by consuming potash in alkali-feldspar when H<sub>2</sub>O-bearing fluid locally passed through the grain boundaries. The former microstructure was formed at 825-900 °C before lamella formation, and the latter microstructure was formed after the lamella formation. These microstructures are the indicators of fluid pathways formed under two different temperature conditions. The common coexistence of these microstructures implies that the fluid used similar pathways during the retrograde metamorphism.

**Keywords:** diffusion, ternary feldspar, fluid, solvus, serrate grain boundary

## INTRODUCTION

Fluid plays an important role in geologic processes through mass and heat transfer. It also defines mechanical strength of rocks, density, and volatile content, thus elastic properties of rocks (Jamtveit and Austrheim, 2010). Fluid infiltration can trigger the metamorphic reactions (Austrheim, 1987) or partially affect the isotopic composition of geochronologically important minerals such as zircon and monazite (Geisler et al., 2007; Kawakami and Suzuki, 2011; Williams et al., 2011) or assist new growth of such minerals (Kirkland et al., 2009). Therefore, recognition of the fluid activity and determination of its timing during metamorphism is crucial for the correct interpretation of the results from dating as well as the pressure-temperature-time (*P-T-t*) path.

Fluid activity in metamorphic rocks is commonly preserved as microstructures involving hydrous minerals, such as garnet partly replaced by the biotite due to retrograde hydration reactions. This kind of retrograde hydration is a common phenomenon in most metamorphic rocks. Although multiple fluid activity with different fluid compositions is likely (e.g., Higashino et al., 2013), determination of the timing at which fluid infiltration took place has been difficult to constrain especially for high-temperature (upper-amphibolite to granulite facies conditions, including ultrahigh-temperature condition) where most of the mineral compositions are commonly altered by the later diffusion processes.

Ternary feldspars, however, can preserve chemical evidence of high-temperature conditions if the original single-phase composition can be calculated from the lamella microstructure, even for ultrahigh-temperature metamorphic rocks (e.g., Hokada, 2001). Utilizing this method for ternary feldspars with zonal, lamellar microstructures potentially serve as a record of chemical zoning formed at high temperatures. In this study, we utilize unique microstructures of ternary feldspars and biotite as a recorder of fluid activity in the high-temperature metamorphic rocks and show how these can be interpreted.

## ANALYTICAL METHOD

Chemical analysis of constituent minerals and the X-ray elemental mapping were performed with the JEOL JXA8105 at Kyoto University. The analytical conditions for the quantitative analyses were 15 kV accelerating voltage, 10 nA probe current, 3  $\mu$ m beam diameter and counting time of 10 and 5 seconds for peaks and backgrounds

101 respectively for most elements, and 30 and 15 seconds, respectively, for F and Cl.  
102 Natural and synthesized oxides were used as standards, and the ZAF correction was  
103 employed for processing the X-ray intensity data. Aluminosilicates were identified by  
104 Raman spectroscopy (JASCO NRS 3100) at Kyoto University. Several back scattered  
105 electron images were obtained using the field emission-scanning electron microscope  
106 (JEOL JMS-7001) at Osaka University.

107 In order to calculate the pre-exsolution composition of feldspars, chemical analyses  
108 of the host feldspar and lamella, coupled with their relative proportions, were utilized  
109 based on the methodology described in Kroll et al. (1993) and Hokada (2001). The areal  
110 proportions of host feldspar and lamella were estimated utilizing back scattered electron  
111 (BSE) images and an image processing software 'Image J' (Rasband, 2007). Areal  
112 proportion was assumed to be identical to the volume proportion, and the weight  
113 proportion was calculated utilizing densities of 2.67 g/cm<sup>3</sup> for albite (Ab) and 2.57  
114 g/cm<sup>3</sup> for orthoclase (Or). The celcian component (< 0.6 mol%) and ferric iron (< 0.1  
115 mol%) was neglected because of low concentrations.

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#### SAMPLE DESCRIPTION

118 The sample utilized in this study is a sillimanite-biotite-garnet gneiss (sample  
119 TK2002122304) collected from the Lützow-Holm Complex at Akarui Point during the  
120 44th Japan Antarctic Research Expedition (JARE). It occurs intercalated with the  
121 biotite-hornblende gneiss. The peak metamorphic condition of 7.7-9.8 kbar and 770-790  
122 °C was previously estimated from the same sample (Kawakami et al., 2008). For further  
123 details of the geological setting and sample locality, see Kawakami et al. (2008).

124 The studied sample mainly consists of garnet porphyroblasts, alkali-feldspar,  
125 plagioclase, quartz, biotite and sillimanite (Fig. 1a-c), with minor amounts of ilmenite,  
126 spinel, rutile, apatite, monazite and zircon. The sample can be divided into two parts. In  
127 the pinkish 'leucosome' lenses and layers, alkali-feldspar, quartz and garnet are  
128 dominant. In the residuum white-colored part with melanocratic patches, garnet, biotite  
129 and alkali-feldspar are dominant (Fig. 1a). The leucosome is concordant with the  
130 gneissosity, which is mainly defined by biotite ± sillimanite (Fig. 1a, c).

131 Garnet (Alm<sub>62-75</sub>Prp<sub>19-26</sub>Grs<sub>1-16</sub>Sps<sub>1-2</sub>, X<sub>Mg</sub> [= Mg/(Mg + Fe<sub>total</sub>)] = 0.20-0.28)  
132 occurs as porphyroblast of up to ca. 10 mm in diameter (Fig. 1a-b). It commonly  
133 accompanies pressure shadows of the leucosome (Fig. 1a), suggesting that at least part

134 of the garnet rim is a peritectic product of partial melting and the leucosome originally  
135 represents the melt (Vernon and Clarke, 2008). The garnet is chemically zoned, and the  
136  $X_{Mg}$  value increases from the core to the rim, suggesting its prograde origin. The garnet  
137 core includes some rare inclusions of anorthite(An)-rich plagioclase ( $An_{30-50}$ ) compared  
138 to that in the matrix. The rim includes biotite, quartz, plagioclase ( $An_{12-16}$ ), and kyanite.  
139 A kyanite inclusion in the rim supports a prograde origin of garnet because the kyanite  
140 is considered to be the prograde relic (e.g., Hiroi et al., 1983).

141 Feldspars in the matrix can be divided into three types; ternary feldspar with  
142 Or-rich alkali-feldspar rim (Figs. 2a-c, 3); plagioclase ( $An_{2-15}$ ; Fig. 2a); and ‘zoned  
143 feldspar’ (Fig. 2d). The distribution of these three types of feldspars can best be  
144 recognized by X-ray element maps (Fig. 3). Although Fig. 3 mainly covers the residuum  
145 part, thin-section-scale element mapping showed that three types of feldspars are  
146 present in both the leucosome and the residuum.

147 Alkali-feldspar is coarse-grained (up to ~3 mm) and characterized by the strong  
148 development of exsolution lamellae (Fig. 2a-c). In general, each grain consists of an  
149 inner region with a mesoperthitic microstructure and an outer region with a perthitic  
150 microstructure (Fig. 3). Some grains of alkali-feldspar consist of a region with only  
151 perthitic microstructure (Figs. 2a-b, 3). Since they are smaller than those with  
152 mesoperthitic and perthitic regions, their appearance can be explained by preferential  
153 cutting during the fabrication of the thin section. They also could be small grains of  
154 mesoperthite re-equilibrated to form perthite and matrix plagioclase during retrograde  
155 metamorphism (see below). The change from mesoperthitic domain to perthitic domain  
156 is microstructurally gradual (Fig. 2b), and thus the integrated feldspar composition  
157 varies gradually as well. Reintegration of the exsolution microstructures shows that the  
158 inner region of the alkali-feldspar has the intermediate composition of Ab and Or with  
159 1-2 wt% CaO, and that the outer regions has an Or-richer and An-poorer composition  
160 compared to the inner region (Table 1). The former is termed ‘ternary feldspar’ and the  
161 latter ‘Or-rich alkali-feldspar’ hereafter for simplicity. The width of the lamellae in the  
162 ternary feldspar (mesoperthitic regions) varies from 3 to 10  $\mu m$ . Commonly, both fine  
163 (~3  $\mu m$ ) and coarse (~10  $\mu m$ ) lamellae coexist in a single alkali-feldspar core (Fig. 2b),  
164 although domains with only fine lamellae or coarse lamellae are also present. The  
165 proportion of Ab lamellae between these two domains is very similar, suggesting that  
166 pre-exsolution single-phase composition of these domains were almost the same. Fine

167 lamellae, especially, are not perfectly straight although they are almost uniform in width  
168 (Fig. 2b), and they occasionally bifurcate. Therefore, this mesoperthitic microstructure  
169 is considered to have formed through spinodal decomposition.

170 Plagioclase ( $An_{2-15}$ ) is commonly present in the matrix along the grain boundaries  
171 between the alkali-feldspar (Figs. 2a, 3), or surrounding biotite aggregate (Fig. 3c).  
172 Plagioclase contains more An content ( $\sim An_{15}$ ) when surrounding a biotite aggregate,  
173 whereas those present between alkali-feldspar grains as films are more albitic and vary  
174 in composition ( $An_{2-10}$ ) (Fig. 3c). Very albitic plagioclase ( $An_{2-3}$ ) tends to be  
175 concentrated at the rim of the plagioclase films developed between alkali-feldspar  
176 grains (Fig. 3c). Where Or-rich alkali feldspar is finer grained and abundant, the  
177 plagioclase is abundant along the grain boundaries, suggesting a genetic relationship  
178 (Fig. 3).

179 Zoned feldspar is commonly observed next to biotite at the biotite-rich portion of  
180 the matrix. The occurrence of this type of feldspar is rare, and shows a localized  
181 distribution on the thin section scale. The following three zones coexist in each zoned  
182 feldspar grain: (i) zone 1 with mesoperthitic microstructure, (ii) zone 2 with  
183 anti-perthitic microstructure, and (iii) zone 3 consisting of albitic plagioclase without  
184 lamellae (Fig. 2d). Zoned feldspar lacking zone 3 is rarely present. The microstructure  
185 and pre-exsolution composition of zone 1 are, in most cases, quite similar to that of the  
186 ternary feldspar. Therefore, this zone remained almost unaffected by the formation of  
187 the zonal structure, retaining the original, high-temperature composition. The lamellae  
188 in the zone 1 are considered to have formed as a result of spinodal decomposition. Zone  
189 2 has microstructural features interpreted to have formed as a result of spinodal  
190 decomposition, such as lamellae uniform in width but incompletely straight (Figs. 2d,  
191 4a).

192

### 193 **Textural correlation between zoned feldspar and biotite**

194 Zoned feldspar is commonly developed next to matrix biotite. Towards the biotite,  
195 the feldspar changes from zone 1 through zone 2 to zone 3. In order to detect the  
196 original chemical zoning that was formed before the lamellar microstructures, an  
197 integrated composition for the long rectangular areas in Fig. 4a were estimated (termed  
198 'line traverse analysis' hereafter) and the result is plotted in Fig. 4b. The overall trend in  
199 the recalculated single-phase feldspar composition is to become depleted in the Or

200 component as biotite is approached. The change in composition from zone 1 to zone 2 is  
201 discontinuous, while in many cases, the lamellae are continuous at the boundary  
202 between zones 1 and 2 (Fig. 4a). Within each zone, the recalculated pre-exsolution  
203 composition varies as a function of distance from the adjacent biotite (Fig. 4b). The  
204 pre-exsolution composition of the reintegrated areas in the zone 1 is occasionally  
205 enriched in the Or component near the boundary between zones 1 and 2. In zone 2, the  
206 Or component decreases near the boundary between zones 1 and 2 (Fig. 4b).

207 When the adjoining biotite is at a high angle with respect to the Or lamella in  
208 alkali-feldspar, the boundary between the alkali-feldspar and the biotite is often serrated.  
209 Each of the biotite projections are selectively developed next to the Or lamella every  
210 3-5  $\mu\text{m}$ . In addition the thickness of the Or lamellae often gradually decreases towards  
211 the biotite (Fig. 5). These biotite projections are not developed next to the Ab lamella.  
212 When the lamellae in alkali-feldspar are at a low angle with the adjoining biotite, this  
213 kind of projection also does not occur. It should be noted that the development of this  
214 serrate boundary is a phenomenon which requires a smaller scale of mass transfer than  
215 the zoned feldspar formation as discussed below.

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## DISCUSSION

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### Prograde to peak metamorphism

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Since the garnet in this study was formed during the prograde metamorphism, the  
minerals found as inclusions in the garnet rim, biotite, kyanite, garnet, quartz,  
alkali-feldspar and plagioclase ( $\text{An}_{12-16}$ ), represent the mineral assemblage of the  
prograde stage. The peak  $P$ - $T$  conditions estimated using the same sample as this study  
(7.7-9.8 kbar and 770-790  $^{\circ}\text{C}$ ; Kawakami et al., 2008) uses the garnet grains that  
include sillimanite in the rim. Therefore, garnet porphyroblast growth continued from  
the kyanite stability field into the sillimanite stability field in the sample used in this  
study.

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Mesoscopic microstructure, such as the development of leucosome (Fig. 1a), both  
as lenses and pressure shadows of garnet, indicates that the sample experienced partial  
melting, and that the garnet is the peritectic product of the partial melting reaction.  
Replacement of the garnet rim by the intergrowth of biotite and plagioclase (Fig. 1b) is  
a characteristic of a microstructure representing the back reaction between garnet and  
melt during cooling (Holness et al., 2011a). Therefore, it is highly likely that melt was



233 present at the peak of metamorphism, and the melt forming reaction is likely to be

234  $Bt + Als (Ky/Sil) + Qtz \rightarrow Grt + Akfs + melt.$

235 Since the studied sample is very rich in the Or component, the Akfs component was not  
236 only incorporated into the melt but also existed as a solid phase during the peak  
237 metamorphism.

238

### 239 **Formation of ternary feldspar and Or-rich alkali-feldspar rim**

240 The pre-exsolution compositions of the alkali-feldspars are plotted on the  
241 An-Ab-Or ternary diagram of Fuhrman and Lindsley (1988) (Figs. 4 and 6). The ternary  
242 feldspar (Fig. 6) and zone 1 of the zoned feldspar (Fig. 4) preserves the highest  
243 temperature among the feldspars of the sample at the isotherm for 750-900 °C, with the  
244 majority around 825 °C, suggesting that the ternary feldspar was formed at 825-900 °C  
245 (Figs. 4 and 6). This temperature range is slightly higher than the peak metamorphic  
246 temperature of Akarui Point previously estimated as 770-790 °C (Kawakami et al. 2008).  
247 This suggests that the ternary feldspar was stable at the peak of metamorphism.

248 Orthoclase-rich alkali-feldspar developed at the rim of the ternary feldspar can be  
249 interpreted to have coexisted with plagioclase ( $\sim An_{10}$ ) present at the grain boundaries.  
250 This mineral assemblage is the same as that consisting of the host and exsolution  
251 lamella in the ternary feldspar. The An content of the matrix plagioclase is almost the  
252 same as that of the plagioclase lamella (Fig. 6; Table 1), except for the case of extremely  
253 albitic plagioclase in the matrix ( $An_{2-3}$ ). In addition, the composition of the Or-rich  
254 alkali-feldspar mostly plots on the 750 °C isotherm near the Ab-Or join (Fig. 6). These  
255 observations indicate that the Or-rich alkali-feldspar was formed at lower temperature  
256 than the ternary feldspar.

257 As described above, the development of the Or-rich alkali-feldspar rim on the  
258 ternary feldspar is not concentric, indicating a formation mechanism other than the  
259 overgrowth. Furthermore, the ternary feldspar compositions are on the tie line between  
260 the Or-rich alkali-feldspar composition and most of the grain boundary film plagioclase  
261 (Fig. 6). These observations suggest that the Or-rich alkali-feldspar and the matrix  
262 plagioclase (except for extremely albitic ones) formed as a result of ternary feldspar  
263 decomposition during cooling. That is, the core/rim relationship between the ternary and  
264 Or-rich alkali-feldspars in a single grain can be explained by the diffusion of the Ab and  
265 An components toward matrix plagioclase nucleated at the grain boundaries.

266 Another possible mechanism to account for the formation of the Or-rich  
267 alkali-feldspars may be an interface-coupled dissolution-precipitation mechanism (e.g.,  
268 Lobotka et al., 2004; Putnis and Putnis, 2007, Niedermeier et al., 2009), because we  
269 assume the presence of low H<sub>2</sub>O activity [a(H<sub>2</sub>O)] fluid around the Or-rich  
270 alkali-feldspars as discussed in the next section. Lobotka et al. (2004) shows an example  
271 of Ab replaced by Or as a result of this mechanism. In the present case, the change from  
272 mesoperthitic domain to perthitic domain is microstructurally gradual (Fig. 2b), which  
273 is inconsistent with the sharp boundary expected to form by the interface-coupled  
274 dissolution-precipitation mechanism. Extremely albitic plagioclase in the matrix could  
275 be the product of the final stage fluid infiltration.

276 After formation of the Or-rich alkali-feldspar during cooling, the ternary feldspar  
277 that survived from the depletion of the An and Or components must have experienced  
278 spinodal decomposition to form the current lamellar microstructure. After further  
279 cooling, Or-rich alkali-feldspar areas must have exsolved. Differences in the thickness  
280 and periodicity of these lamellar microstructures support this order.

281

## 282 **Formation of the zoned feldspar**

283 Zoned feldspar (Fig. 2d) shows two features different from the other alkali-feldspar  
284 grains; (i) It is found only in grains adjacent to biotite, and (ii) the Or component of the  
285 re-integrated grain composition decreases at those feldspar rims in contact with biotite,  
286 from zone 1 to zone 3 (Fig. 4). The re-integrated compositions from each zone of the  
287 feldspar (Table 2) plot approximately on a straight line corresponding to X-Or<sub>75</sub>Ab<sub>25</sub> in  
288 the An-Ab-Or ternary diagram (Fig. 4c), where X is a composition at which a gray  
289 broken line crosses the Ab-An apex..

290 Ternary feldspar could have formed originally at a temperature of 825-900 °C as  
291 discussed above. At 825-900 °C, an H<sub>2</sub>O-bearing fluid infiltrated along the grain  
292 boundaries between ternary feldspar and biotite that was already present in the matrix  
293 (Fig. 4a). New biotite was then formed by consuming H<sub>2</sub>O component in the fluid and  
294 Or component from the ternary feldspar. This reaction was the driving force responsible  
295 for the diffusion profile shown in Fig. 4b (roughly from point 20 to point 5, except for  
296 the points 15-10 formed after the formation of zones 1 and 2). The diffusion profile only  
297 slightly oversteps the solvus at 825 °C, such the diffusion profile is smooth (a thick gray  
298 line of Fig. 4b). As shown by the similar mean diffusion distance during the zoned

299 feldspar formation, reaction (1) took place at the right-hand side of Fig. 4a (corresponds  
300 to point 4 to point 1 of Fig. 4b) under similar temperature conditions, and produced  
301 another diffusion profile.

302 If the infiltrating H<sub>2</sub>O-bearing fluid was near pure H<sub>2</sub>O, it will trigger partial  
303 melting. This is because the temperature estimated is higher than 825 °C, well above the  
304 water-saturated solidus for the pelitic rocks (e.g. Spear et al., 1999). However, the  
305 preservation of such a high-temperature microstructure is consistent with the infiltration  
306 of a low a(H<sub>2</sub>O) fluid such as CO<sub>2</sub>-rich fluid or the F- and/or Cl-bearing fluid (e.g.,  
307 Higashino et al., 2013) that prevented partial melting, but acted as a source of H<sub>2</sub>O. It is  
308 likely that biotite formed where the a(H<sub>2</sub>O) in the fluid was relatively high. The fluid  
309 only promoted the material transport at the grain boundaries to form the plagioclase film  
310 along the grain boundaries where a(H<sub>2</sub>O) in the fluid was low.

311 The observation that compositional variation in the zoned feldspar is not aligned  
312 on the X-Or(Bt) tie line but shifted to the more albitic side (X-Or<sub>75</sub>Ab<sub>25</sub>) implies that the  
313 Ab component was lost from the ternary feldspar into the fluid phase at the same time  
314 the Or component went to form biotite (Fig. 4c). This release of the Ab component  
315 might have been driven by a reaction between An-rich plagioclase that was present in  
316 the matrix during near-peak metamorphic conditions as indicated by the plagioclase  
317 inclusions in the garnet rim (An<sub>12-16</sub>). Another possible source for an Ab component in  
318 the fluid is the plagioclase included in the prograde garnet core (An<sub>30-50</sub>; Fig. 6) that was  
319 released to the matrix via partial breakdown of the garnet (Fig. 1b).

320 With further temperature decrease, the compositional trend formed by the diffusion  
321 processes oversteps the solvus; for example, the trend X-Or<sub>75</sub>Ab<sub>25</sub> in Fig. 4c crosses the  
322 solvus at 825 °C. It is at this stage that the large compositional gap between zones 1 and  
323 2, as shown by the broken lines in Fig. 4b, was created.

324 The fact that compositions of the zoned feldspar (zones 1, 2 and 3), matrix  
325 plagioclase (~An<sub>10</sub>), and Or-rich alkali-feldspar are aligned on a single trend, and the  
326 fact that the composition of the Or-rich alkali-feldspar preserves the composition  
327 characteristic of the 750 °C isotherm (Figs. 4 and 6) suggest that formation of these  
328 major microstructures was complete by 750 °C. The composition near the solvus for the  
329 Ab-Or join at ~700 °C (Ab<sub>27.5</sub>Or<sub>71.8</sub>An<sub>0.8</sub>; Table 1) is preserved as the Or phase of the  
330 Or-rich alkali-feldspar. Preservation of such a composition, without changing to more  
331 Or-rich composition is rare (cf. Hokada, 2001), and requires a rapid cooling process

332 after their formation.

333 Further decrease in temperature diminished the mean diffusion distance for the  
334 biotite-forming reaction at the boundary between biotite and the zone 3. This resulted in  
335 the elimination of zone 1 such that zone 1 no longer consumed Or component. However,  
336 consumption of the Or component by zone 2 continued. Finally at relatively lower  
337 temperatures, the local equilibrium at the zone 1/ zone 2 boundary caused the zone 1  
338 side to become Or-richer and the zone 2 side to become Ab-richer. Further consumption  
339 of the Or component probably resulted in zone 3.

340 Lamella formation in zones 1 and 2 by spinodal decomposition probably started to  
341 take place at the minimal and maximal of the Or component in zones 1 and 2,  
342 respectively (point 9 of zone 1 and point 16 of zone 2), because the fast cooling forces  
343 these compositions to hit the spinodal curve first. The lamella formation probably  
344 propagated to the surrounding compositions as a temperature decrease.

345 The interface-coupled dissolution-precipitation mechanism may potentially be  
346 applicable to the formation of the zoned feldspar. The important microstructural  
347 constraint in applying this mechanism is that the zoned feldspar is only found adjacent  
348 to biotite. Therefore, biotite and zoned feldspar should have a genetic link, and probably  
349 formed simultaneously when a fluid infiltrated between them. However, since biotite is  
350 not included in the zoned feldspar, and the zoned feldspar is not porous, we consider  
351 that microstructural observation does not necessarily support that the interface-coupled  
352 dissolution-precipitation occurred in the present case.

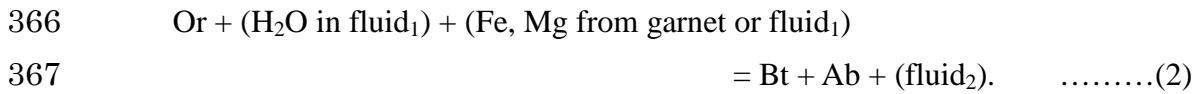
353 As a conclusion, the zoned feldspar described in this study is a rare case where the  
354 formation process could be explained by the diffusion that oversteps the solvus (e.g.,  
355 Sekerka and Wang, 1999).

356

### 357 **Formation of serrate boundary between biotite and feldspar**

358 The boundary between biotite and feldspar is rarely serrated. Where it occurs the  
359 projections of biotite are selectively developed next to the Or phase (Fig. 5). This  
360 microstructural feature implies that serrated boundary formation postdated the lamella  
361 formation, and that Or lamella and the projections of biotite have some genetic  
362 relationship. Because the Or lamellae in contact with the biotite projections are thinner  
363 near the contact with biotite (Fig. 5), this boundary likely formed by the consumption of  
364 Or to form biotite (Fig. 4), or in other words, dissolution of Or to recrystallize biotite

365 through the reaction



368 Although biotite projections at the serrate boundary are too small to perform  
369 quantitative analyses, their textural features indicate a very short diffusion distance and  
370 suggest that they developed during the final stage of biotite formation during retrograde  
371 metamorphism.

372 Abart et al. (2009) reports similar microstructure as above, which is the Ab/Or  
373 interface within the mesoperthite in an anatexite from Tanzania, corrugated on the  
374 sub-micron scale; lobes of Or extend into more An-rich lamellae within the Ab host.  
375 This is interpreted as growth phenomenon associated with coarsening of the Or lamellae,  
376 that took place after Ab lamella exsolved about the peristerite gap (Abart et al., 2009).  
377 Different from the serrate boundary between biotite and feldspar, this microstructure  
378 lacks apparent evidence for the presence of fluid during its formation, such as the  
379 presence of hydrous mineral.

380 Holness et al. (2007; 2011b) also reports similar microstructure ('stepped grain  
381 boundary') from the Skaergaard Intrusion, East Greenland, where  
382 clinopyroxene/plagioclase grain boundaries have serrations associated with the  
383 dissolution of exsolution lamellae from Ca-poor pyroxene, and the corresponding  
384 growth of Ca-rich plagioclase is observed. Non-ubiquity of the serrated boundaries on  
385 the thin-section scale, and their association with larger, non-isochemical, symplectic  
386 structures, suggests that they formed by the dissolution of metastable Ca-poor pyroxene  
387 and recrystallization of adjacent plagioclase and augite consequent to the introduction of  
388 a metasomatizing fluid along grain boundaries (Holness et al., 2007). Therefore, similar  
389 fluid-related process observed in the solidified layered intrusion is likely to have taken  
390 place in the course of solidification of partially molten high-temperature metamorphic  
391 rocks as well. By understanding the mechanism of these microstructure formation  
392 discussed above, these microstructures will serve as a tool to detect the fluid related  
393 processes in high-temperature rocks.

394

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402

403

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480

## 481 **FIGURE CAPTIONS**

482

483 **FIGURE 1. (a)** A slab photo of the sample TK2002122304. Dark gray spots are the  
484 garnet porphyroblasts and melanocratic patches are mainly biotite and sillimanite.  
485 Leucosome is developed as layer-parallel lenses or as the pressure shadows of garnet.  
486 **(b)** A back scattered electron (BSE) image showing the biotite replacing garnet with  
487 plagioclase, and biotite included in the garnet rim. **(c)** A BSE image showing the mode  
488 of occurrence of coarse-grained, biotite and prismatic sillimanite enclosed in a  
489 plagioclase moat. Finer grained biotite occurs as a matrix mineral in the alkali-feldspar-  
490 and quartz-dominant portion. **(d)** Photomicrograph (crossed polarized light) of the  
491 biotite and zoned feldspars surrounding it. **(e)** Enlargement of the boxed area in **(d)**.  
492 Cross polarized light. Note that the zoned feldspar (contoured with a solid line) shows  
493 the same grey retardation except for white lamellae, suggesting that the zoned feldspar,  
494 as a whole, is a single crystal. The gray and white parts in the zoned feldspar correspond  
495 to the Ab phase and Or phase, respectively. The zoned feldspar crystal changes its  
496 composition from an Or-rich one to almost pure albitic plagioclase at the contact with



497 biotite. Mineral abbreviations are after Kretz (1983) except for the followings: Ilm =  
498 ilmenite, Afs = alkali-feldspar, Pl = plagioclase and Z.F. = zoned feldspar.

499

500 **FIGURE 2. (a)-(d)** Feldspar BSE images. **(a)** Ternary feldspar and Or-rich alkali-feldspar  
501 in the matrix. Dark films at the grain boundaries between alkali-feldspar grains are  
502 albitic plagioclase. Note that Or-rich alkali-feldspar is finer grained than the ternary  
503 feldspars, and is locally developed at the rim of the ternary feldspar as well. They are  
504 both accompanied by the development of a plagioclase film at the grain boundaries. **(b)**  
505 Ternary feldspar has domains with fine-grained lamellae (indicated by 'fine') and  
506 coarse-grained lamellae (indicated by 'coarse') in a single grain. **(c)** Or-rich  
507 alkali-feldspar coexisting with biotite in the matrix. Its lamellae vary in width, but  
508 exceedingly fine-grained lamellae are uniformly distributed at the core of the grain. At  
509 the rim, the lamellae are partly absent under BSE image. Note the finer-grain size of the  
510 Or-rich alkali-feldspar compared to the ternary feldspar. **(d)** Mode of occurrence of the  
511 zoned feldspar. The zones 1, 2, and 3 are labeled as 1, 2, and 3, respectively. Zones with  
512 mesoperthitic microstructure (zone 1), anti-perthitic microstructure (zone 2) and  
513 lamella-free albitic plagioclase (zone 3) can be recognized. Toward the grains of biotite,  
514 each alkali-feldspar grain becomes depleted in the Or component.

515

516 **FIGURE 3.** Elemental mapping of the matrix (principally residuum). Warm colors  
517 represent high concentrations of each element. Note the mode of occurrence of ternary  
518 feldspar and the Or-rich alkali-feldspar rim. A plagioclase film is developed at the grain  
519 boundaries of the alkali-feldspars. **(a)** Elemental map of Na. **(b)** Elemental map of K. **(c)**  
520 Elemental map of Ca. **(d)** Back scattered electron image.

521

522 **FIGURE 4. (a)** A BSE image of the zoned feldspar showing the rectangular areas used to  
523 calculate the change in the modal proportion of Or phase (light gray) as a function of  
524 distance from the adjacent biotite. The modal proportion of the Or phase is determined  
525 in each rectangular area and the result is plotted in **(b)**. The white broken line is a  
526 baseline for the measurement of distance from the biotite. Note that the thickness of the  
527 Or phase increases at the boundary between zones 2 and 1. Quartz at the bottom right is  
528 the matrix phase. **(b)** Change of the modal proportion of the Or component as a function  
529 of distance from biotite to zone 3. Gray thick line represents the diffusion profile

530 originally present in the pre-exsolution ternary feldspar. Note the sharp increase in the  
531 Or at the zone 1 /zone 2 boundary, which is interpreted to postdate zones 1 and 2  
532 formation. (c) The An-Ab-Or ternary diagram for the pre-exsolution, re-integrated  
533 compositions for each of the three zones of the zoned feldspar, with isotherms at 750 °C,  
534 825 °C, and 900 °C as shown by solid lines after Fuhrman and Lindsley (1988). Gray,  
535 broken line represents the tie line between the compositions of zone 3 and the Or-rich  
536 part of the zone 1.

537

538 **FIGURE 5.** (a) The BSE image of the serrated boundary between biotite and  
539 alkali-feldspar. The projections of biotite are selectively developed next to the Or  
540 lamellae (light gray) and not developed next to the Ab lamellae (dark gray). (b)  
541 Enlargement of the boxed area in (a). (c) An illustration showing the formation  
542 mechanism of serrated boundary between biotite and feldspar. See text for detailed  
543 explanation.

544

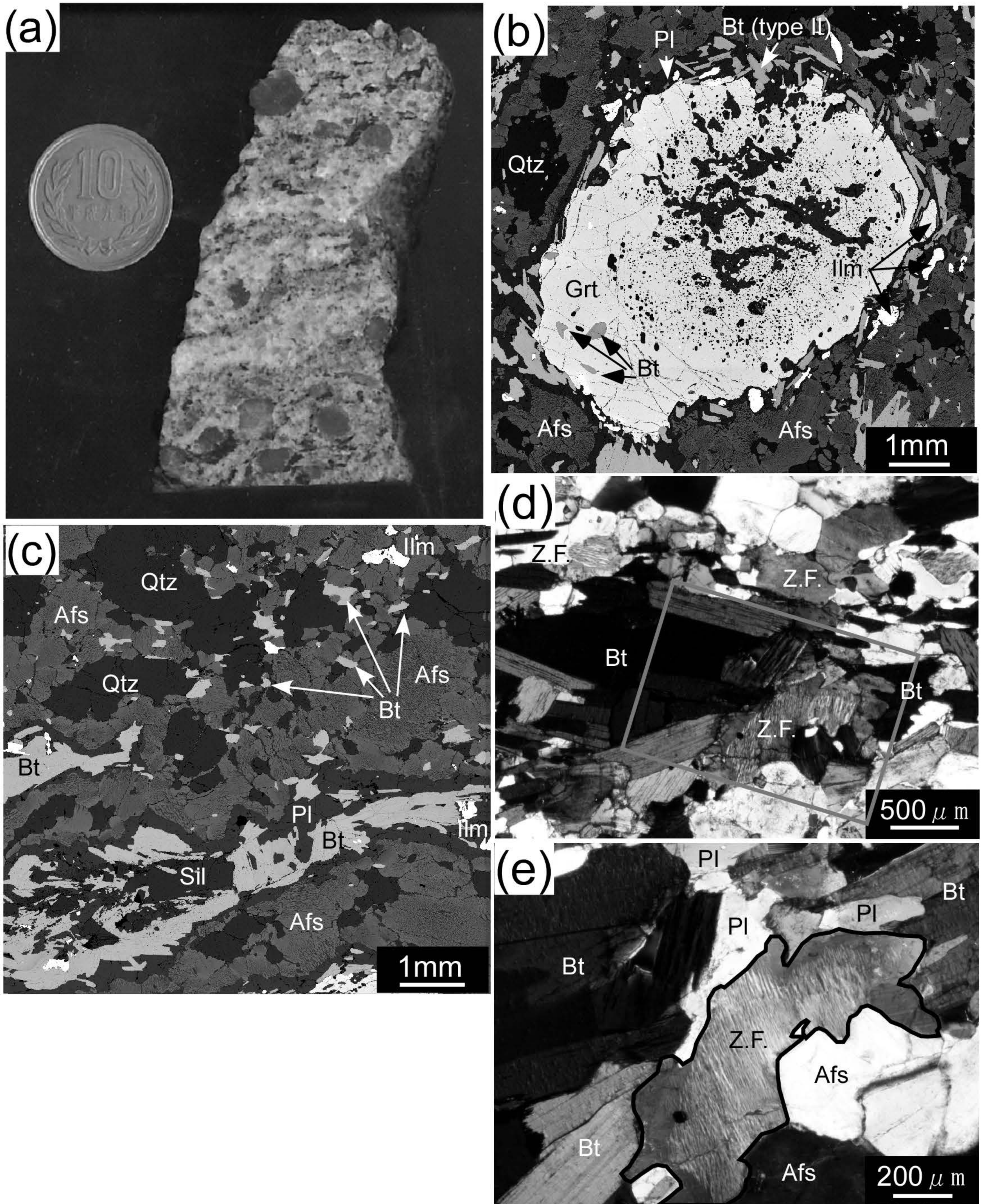
545 **FIGURE 6.** An-Ab-Or ternary diagram for recalculated pre-exsolution alkali-feldspar  
546 compositions, with the isotherms at 750 °C, 825 °C, and 900 °C are shown by the solid  
547 lines (after Fuhrman and Lindsley, 1988). Compositions of prograde plagioclase  
548 included in garnet and retrograde plagioclase in the matrix are also shown.

549

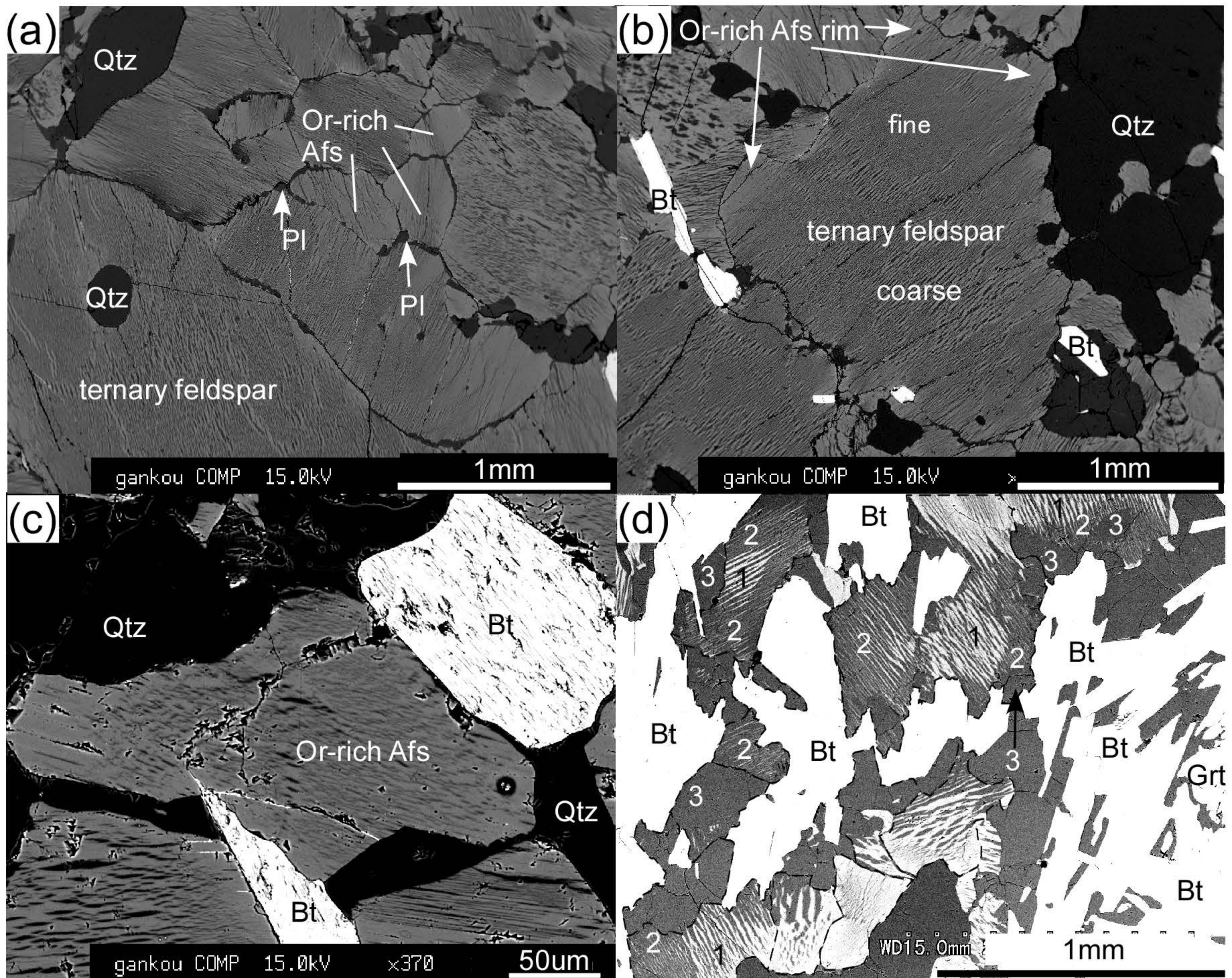
550 **TABLE 1.** Representative compositions of Or phase and Ab phase of ternary feldspar,  
551 Or-rich alkali feldspar and the zoned feldspar (zone 1, zone 2 and zone 3). Or; Or phase,  
552 Ab; Ab phase, S.D.; standard deviation.

553

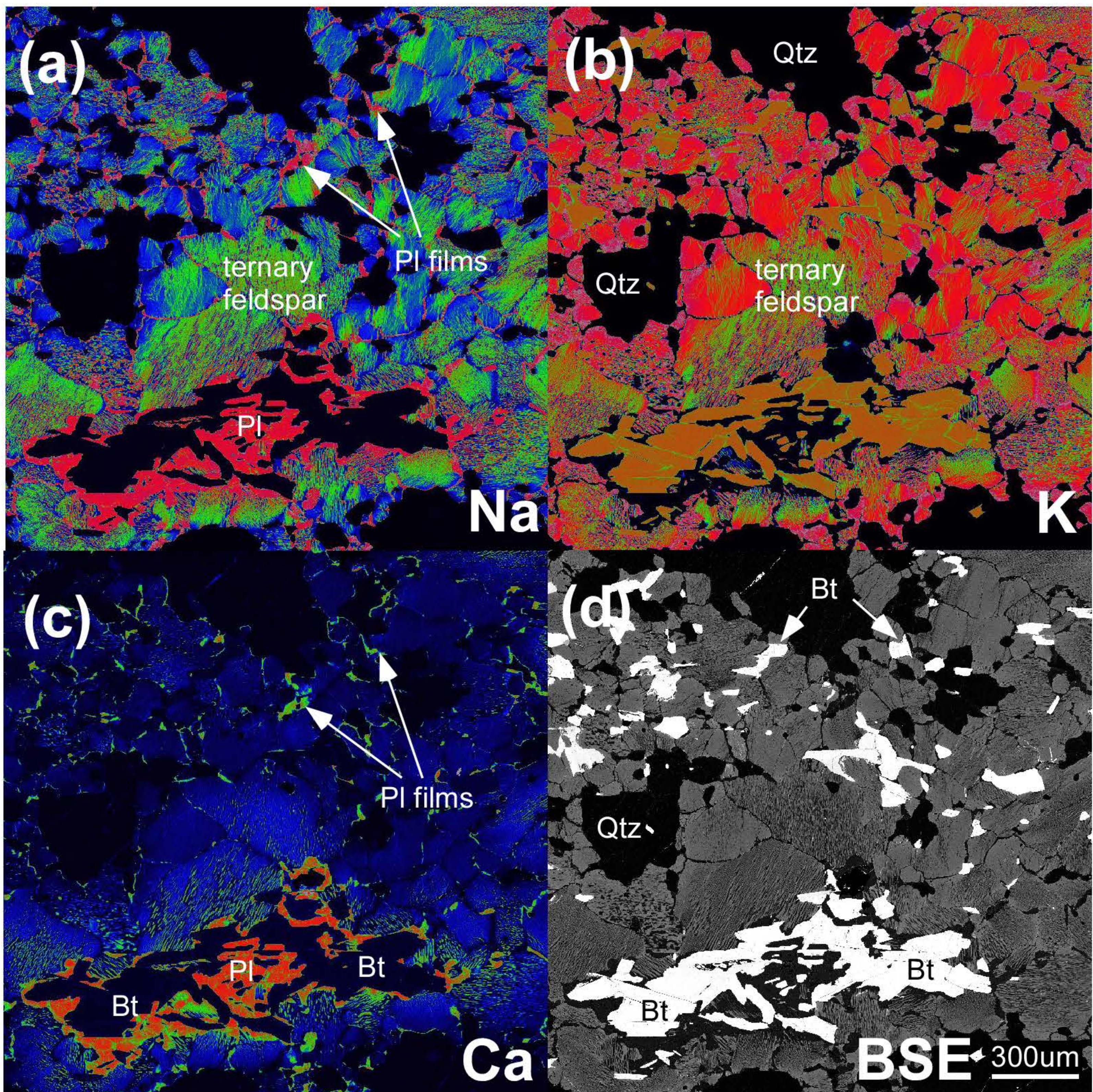
554 **TABLE 2.** Summary of the reintegrated composition of zones 1 and 2 of the zoned  
555 feldspar and the result of temperature estimate utilizing the ternary feldspar  
556 thermometry by Fuhrman and Lindsley (1988) (FL) and Elkins and Grove (1990) (EG).  
557 The different characters in the 'grain' line represent the grain names of feldspar  
558 analyzed. The 'Zone' column shows the zone number of the zoned feldspar.



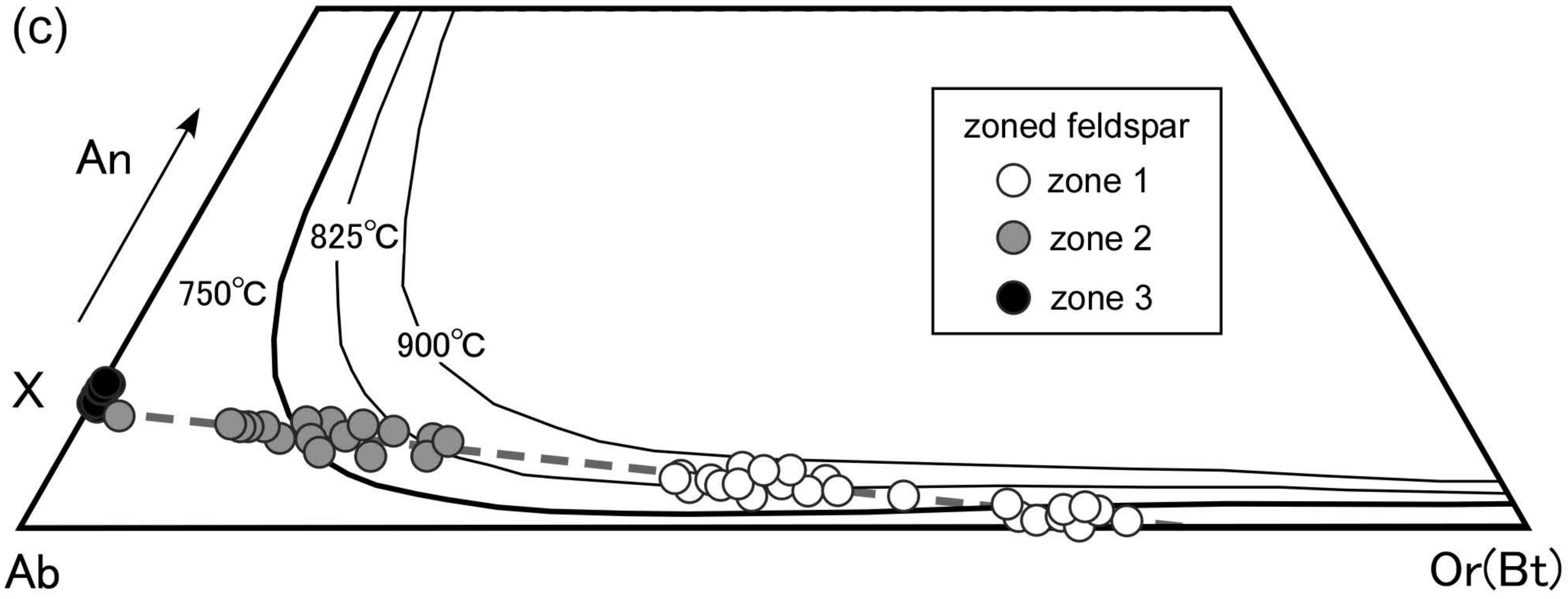
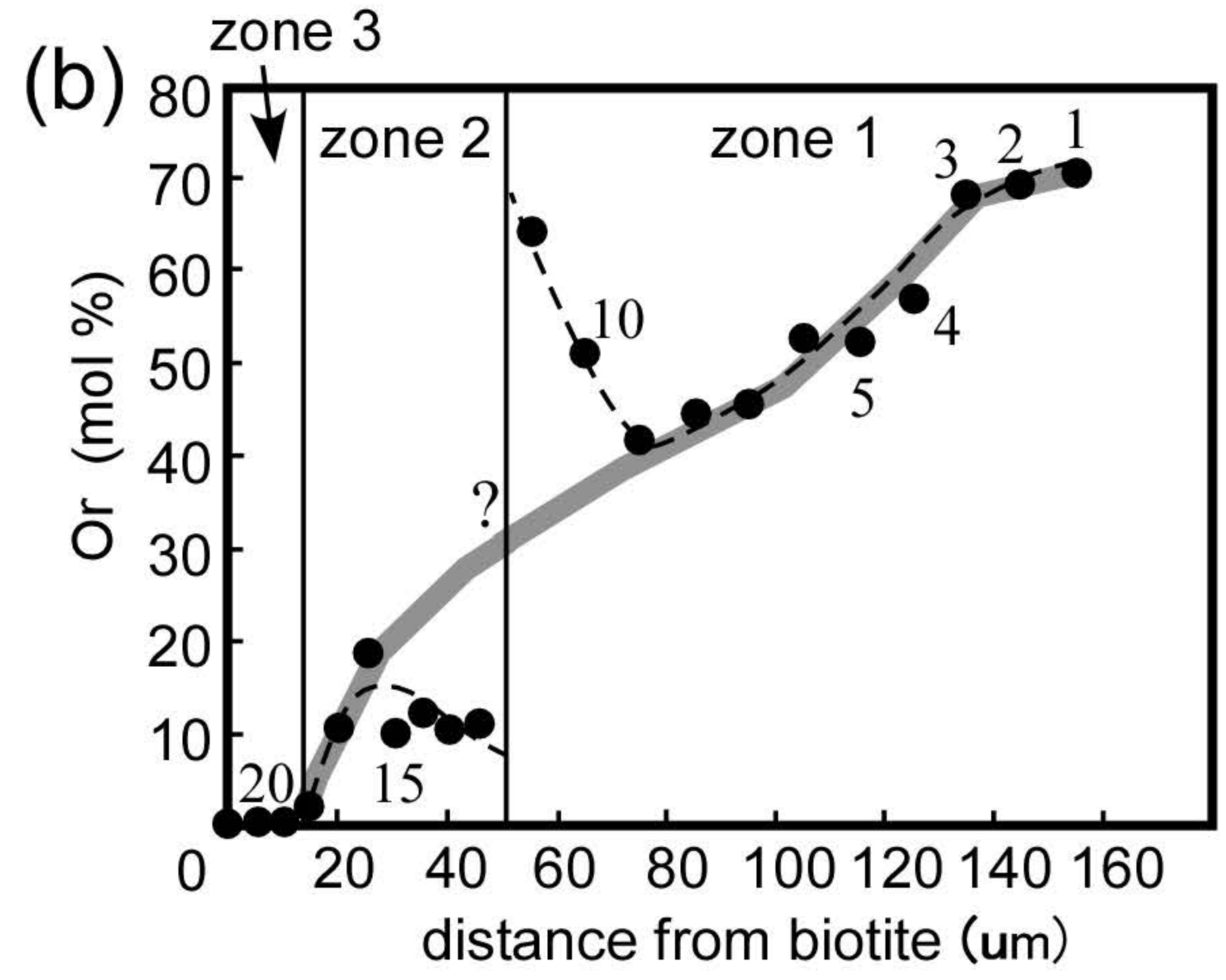
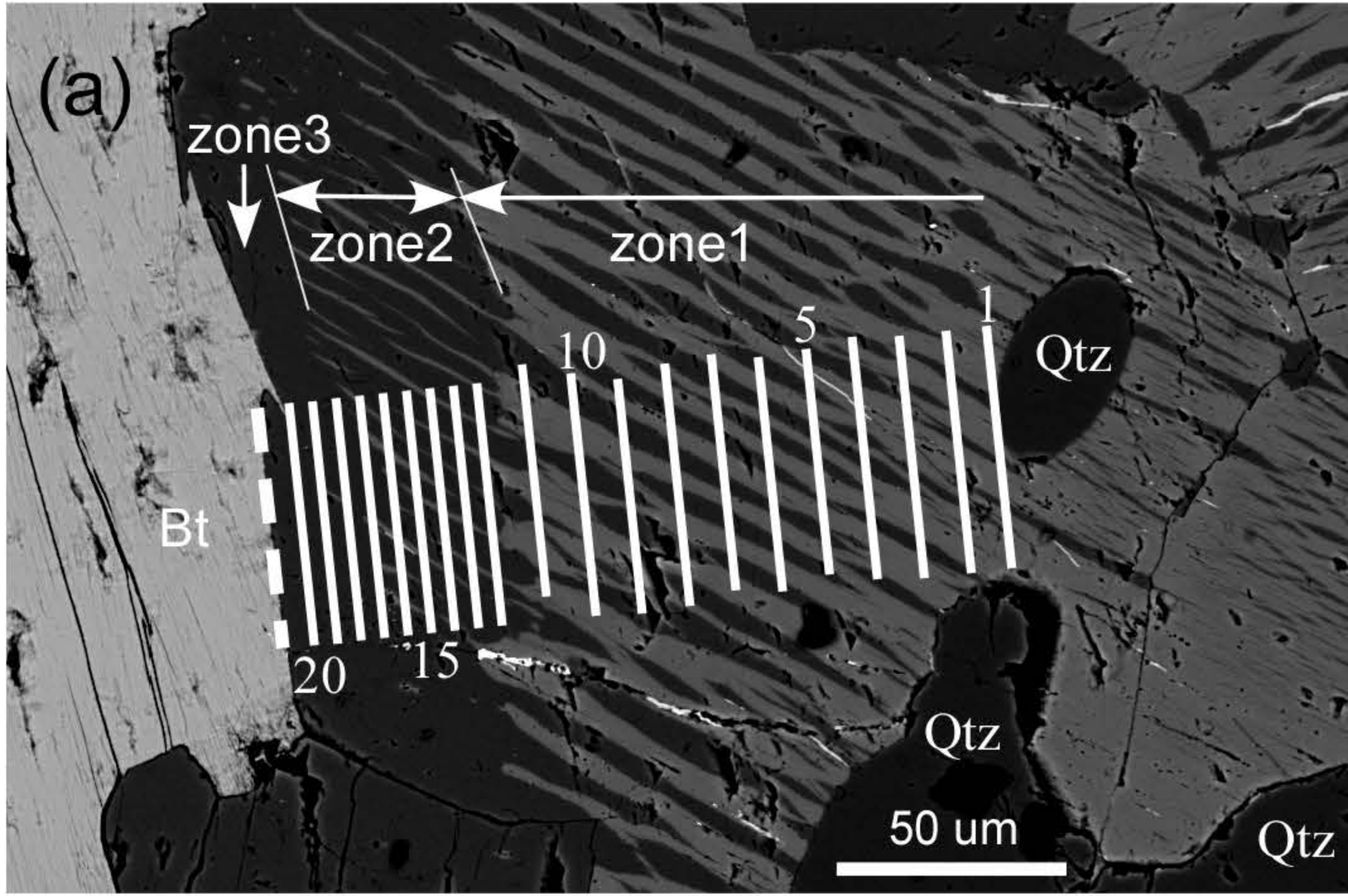
Nakamura et al. FIGURE 1



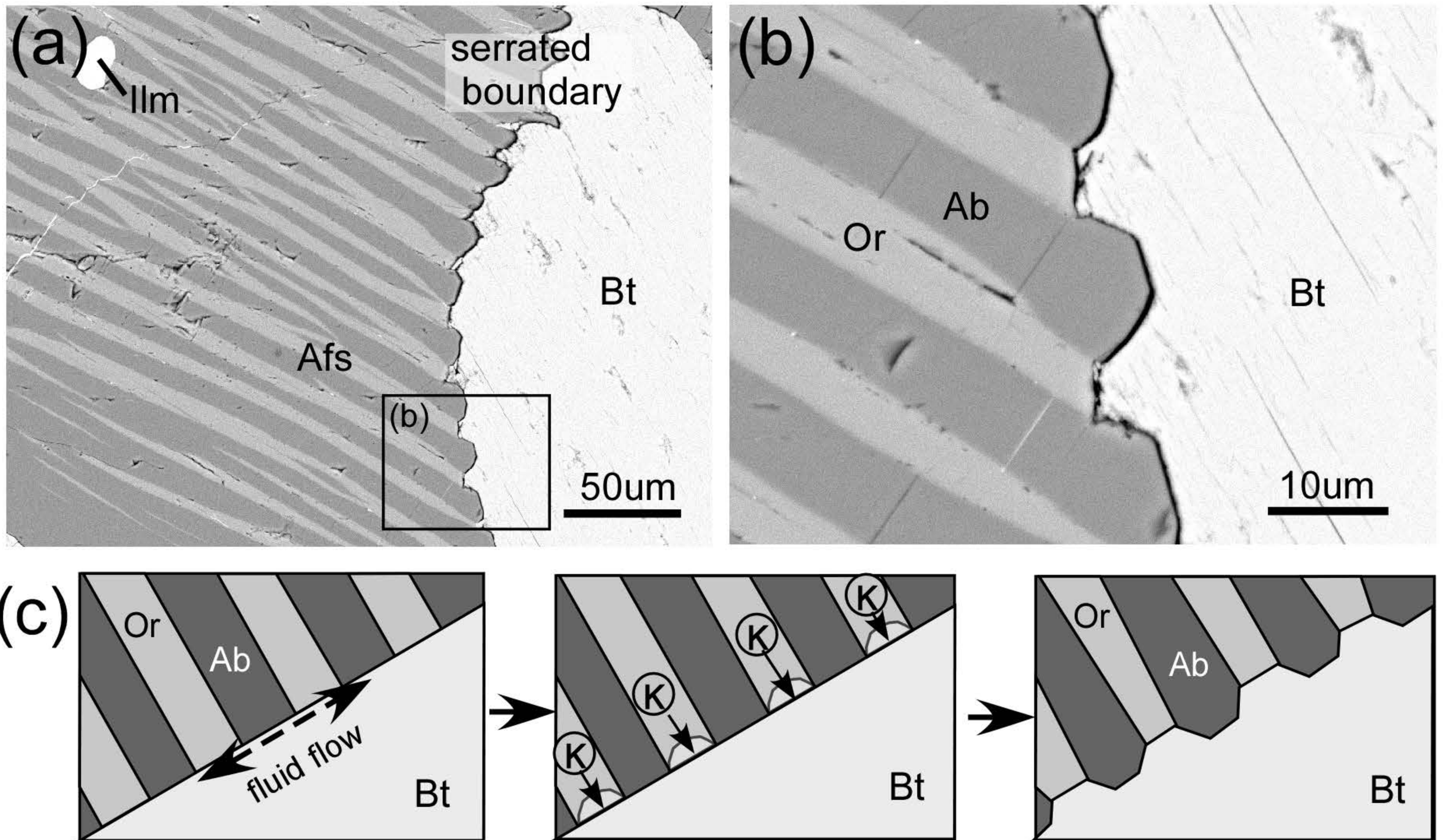
Nakamura et al FIGURE 2



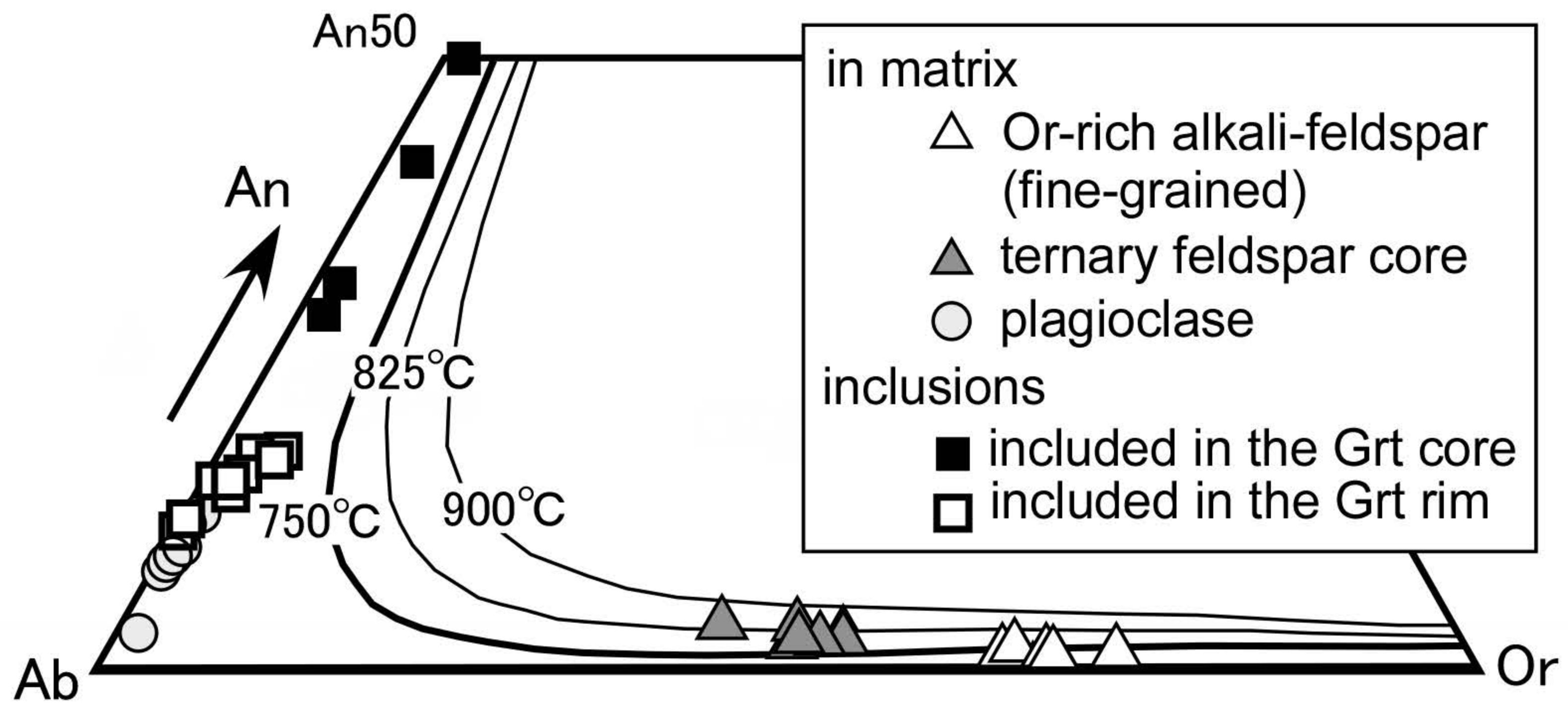
Nakamura et al Fig. 3



Nakamura et al. FIGURE 4



Nakamura et al. FIGURE 5



Nakamura et al. FIGURE 6



texture	ternary feldspar		Or-rich Afs rim			zoned feldspar			
zone					zone 1		zone 2	zone 3	
phase	Or	Ab	Or	Ab	Or	Ab	Ab		
			average	1 S.D.					
wt%									
SiO <sub>2</sub>	63.87	66.00	64.43	0.46	67.97	64.85	66.45	66.29	66.02
Al <sub>2</sub> O <sub>3</sub>	18.55	21.46	18.87	0.19	20.27	18.05	20.56	20.97	21.13
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.02	0.02	0.05	0.02	0.07	0.04	0.00
BaO	0.61	0.09	0.32	0.08	0.00	0.56	0.00	0.08	0.00
CaO	0.01	1.61	0.16	0.04	0.40	0.01	1.90	2.06	2.20
Na <sub>2</sub> O	1.35	10.82	3.03	0.42	11.48	0.89	10.44	10.34	10.12
K <sub>2</sub> O	14.58	0.14	12.05	0.62	0.20	15.64	0.09	0.12	0.12
P <sub>2</sub> O <sub>5</sub>	0.21	0.10	0.14	0.06	0.00	0.10	0.06	0.05	0.08
Total	99.18	100.20	99.02	0.39	100.53	100.12	99.57	99.94	99.66
	formula (O = 8)								
Si	2.97	2.89	2.97		2.96	3.00	2.93	2.91	2.91
Al	1.02	1.11	1.03		1.04	0.98	1.07	1.09	1.10
Fe <sup>3+</sup>	0.00	0.00	0.00		0.00	0.00	0.00	0.00	0.00
Ba	0.01	0.00	0.01		0.00	0.01	0.00	0.00	0.00
Ca	0.00	0.08	0.01		0.02	0.00	0.09	0.10	0.10
Na	0.12	0.92	0.27		0.97	0.08	0.89	0.88	0.86
K	0.87	0.01	0.71		0.01	0.92	0.01	0.01	0.01
P	0.01	0.00	0.01		0.01	0.00	0.00	0.00	0.00
Total	5.00	5.01	5.00		5.00	5.00	4.98	4.99	4.98
An	0.0	8.0	0.8		1.9	0.1	9.1	9.8	10.7
Ab	12.0	92.0	27.5		97.0	8.0	90.4	89.5	88.7
Or	88.0	1.0	71.8		1.1	92.0	0.5	0.7	0.7

Table 1 Nakamura et al.

grain name of Z.F.	zone number	Areal proportions (%)		Host and lamellae compositions						Re-integrated composition (mol%)			Equilibrium temperature (°C)	
		Ab phase	Or phase	Ab phase (mol%)			Or phase (mol%)			An	Ab	Or	T (FL)	T (EG)
				An	Ab	Or	An	Ab	Or					
A	1	46.9	53.1	7.2	92.2	0.6	0.4	8.6	91.1	3.6	48.6	47.8	741	747
	2	85.1	14.9	8.2	91.1	0.7	0.4	8.6	91.1	7.0	79.2	13.8		
B	1	43.0	57.0	8.5	90.9	0.6	0.3	8.3	91.5	3.9	44.6	51.6	752	756
	2	81.2	18.8	7.4	92.4	0.2	0.3	8.3	91.5	6.1	77.1	16.8		
C	1	47.0	53.0	9.1	90.4	0.5	0.1	8.0	92.0	4.4	47.5	48.1	780	785
	2	82.1	17.9	9.8	89.5	0.7	0.1	8.0	92.0	8.1	75.4	16.5		
E	1	45.4	54.6	9.8	89.5	0.1	0.1	9.3	90.6	4.6	46.5	48.6	794	815
	2	79.1	20.9	9.8	89.6	0.7	0.1	9.3	90.6	7.8	73.3	18.9		
1	1	47.6	52.4	7.6	91.7	0.7	0.0	11.2	88.8	3.7	50.3	46.0	795	799
	2	78.9	21.1	9.1	90.6	0.3	0.0	11.2	88.8	7.2	74.3	18.4		
2	1	47.0	53.0	9.6	89.9	0.5	0.1	13.7	86.2	4.7	50.2	45.1	797	801
	2	76.9	23.1	9.6	90.1	0.4	0.1	13.7	86.2	7.4	72.9	19.6		
3	1	45.2	54.8	8.5	90.9	0.6	0.0	11.9	88.1	3.9	48.4	47.7	819	823
	2	71.3	28.7	9.3	90.2	0.5	0.0	11.9	88.1	6.7	68.3	25.0		
4	1	49.5	50.5	9.3	90.2	0.6	0.0	8.5	91.4	4.7	49.7	45.6	806	811
	2	76.7	23.3	9.6	89.9	0.5	0.0	8.5	91.4	7.5	71.5	21.0		
5	1	45.7	54.3	8.6	90.9	0.5	0.1	8.8	91.1	4.0	47.1	48.8	760	764
	2	82.4	17.6	8.4	91.1	0.4	0.1	8.8	91.1	7.0	77.1	15.9		
7	1	48.9	51.1	7.3	92.2	0.5	0.0	7.7	92.3	3.6	49.8	46.6	822	827
	2	73.6	26.4	9.0	90.7	0.4	0.0	7.7	92.3	6.7	69.4	24.0		

Table 2 Nakamura et al.