Appendix A

The separation of illite from the rock sample

The separation of illite from the rock sample was carried out using following steps (cf. Yagi 2006 and Yagi and Itaya 2011);

1) The borehole core sample (06MI03-8) was cleaved into smaller pieces (rock chips of roughly 1 cm^3), which were then washed and dried.

2) The rock chips (about 750 g) were fractured using a stamp mill, and the fractured samples were classified into three groups using sieves: granulated fractions with diameters of 150–250 μm, 100–150 μm, and 60–100 μm.

3) The granulated fractions were washed with a rabbler in tap water and then further washed using ultrasonic cleaning in deionized water to eliminate the suspension.

4) Ferromagnetic minerals were eliminated from the dried fractions using a hand magnet.

5) The fractions were again classified into three groups using sieves: 150–250 μm, 100–150 μm, and 60–100 μm fractions.

6) Ferromagnetic and quartz minerals were eliminated from the fractions using an electromagnetic separator (Isodynamic® separator). The remaining fractions consisted of K-feldspar and plagioclase: a 150–250 μm fraction of 80 g, 100–150 μm fraction of 160 g, and 60–100 μm fraction of 108 g.

7) K-feldspar was eliminated from each fraction by heavy liquid separation using a sodium polytungstate solution (2.60 g/cm^3), giving plagioclase grains because the specific gravities of orthoclase (2.56 g/cm^3), albite (2.62 g/cm^3), and anorthite (2.76 g/cm^3) in specific

gravity: 52 g of 150–250 μm, 106 g of 100–150 μm grains, and 74 g of 60–100 μm grains.

8) The separated plagioclase grains (about 15 g) were ultrasonically cleaned for 30 min (15 min × 2) in deionized water (about 400 cc) to giving a suspension including illite particles.

9) Repeating step 8 yielded about 12,000 cc of the suspension including illite particles.

10) Coarse particles more than 2.0 μm in diameter were settled and removed from the solution obtained in step 9 using a centrifugal separator (HITACHI himac CT 5L) at a speed of 1,000 rpm for 1 min 20 s (2–3 repeated runs).

11) Fine particles with diameters of 0.2–2.0 μm were settled and removed from the supernatant solution produced in step 10 using a centrifugal separator with a speed of 3,000 rpm for 14 min 40 s.

12) The obtained fine particles with 0.2–2.0 μm were dried in a constant temperature reservoir, resulting in 0.24 g of illite powder for the K-Ar dating.

Appendix B

Plagioclase alteration reactions of the Type A-2, A-3, B-1, B-2, C-1, C-2, C-3-1, C-3-2, and C-3-3 plagioclase

Type A-2 (sample No. 9-8): Overall alteration throughout the plagioclase grain (Pl → Ab + Ill + Cal + Fl). In Type A-2, plagioclase (chemical composition analysis No. 76 in Table 1) is the reactant and albite (No. J03 in Table S1), illite (No. J04), calcite (ideal $CaCO_3$ is assumed), and fluorite (ideal CaF_2) are the products. The same analysis as that used for Type A-1 gives reasonable reactions (Table 5). The reaction equations represent the albitization and production of illite, calcite, and fluorite by the consumption of plagioclase with an inflow of H_4SiO_4, CO_2, Al^{3+}, Fe^{2+}, Mg^{2+}, Na^+, K^+, and F^- from the hydrothermal fluid, accompanied by the outflow of H_2O, H^+, and Ca^{2+} into the hydrothermal fluid (Fig. S6B).

Type A-3 (sample No. 12-7): Overall alteration through the plagioclase grain (Pl → Ab + Kfs + Fl + Ep). In Type A-3, plagioclase (No. 76) the reactant and albite (No. J05), K-feldspar (J06), fluorite (ideal CaF_2), and epidote (No. 43) are the products (Table 1). The albitization, K-feldspathization, fluorite, and epidote are produced by the consumption of plagioclase with an inflow of H_4SiO_4, H^+, Fe^{2+}, Na^+, and F^- from the hydrothermal fluid, and are accompanied by the outflow of H_2O, Al^{3+}, Ca^{2+}, and K^+ into the hydrothermal fluid (Fig. S6C). The Type A-3 sample (sample No. 12-7) has the largest volume of albite among all the samples (Table 2). Therefore, the characteristics of the inflow and outflow

of components via the hydrothermal fluid in the Type A-3 represents the essential mass transfer in an albitization (Table 5): the consumption of plagioclase with an inflow of H_4SiO_4 and Na^+ and an outflow of Al^{3+} and Ca^{2+} into the hydrothermal fluid (Fig. S6C).

Type B-1 (sample No. 12-2): Alteration in the core of the plagioclase grain (Pl → Ab + Kfs + Ill + Cal + Fl). The Type B-1 reaction involves plagioclase (No. J09) as the reactant and albite (No. J10), K-feldspar (No. J12), illite (No. J11), calcite (ideal $CaCO_3$), and fluorite (ideal CaF_2) as the products (Table 1). The overall reactions are characterized by the consumption of plagioclase with an inflow of H_4SiO_4, CO_2, Al^{3+}, Fe^{2+}, Mg^{2+}, K^+, and F^-, accompanied by an outflow of H^+, Ca^{2+} and Na^+ into the hydrothermal fluid (Fig. S6D).

Type B-2 (sample No. 7-8): Alteration in the core of the plagioclase grain (Pl → Ab + Kfs + Ill + Fl). The reactant is plagioclase (No. J07 in Table 1) and the product minerals consist of an albite area (No. O04), K-feldspar (No. O07), illite (No. J08), and fluorite (ideal CaF_2). The obtained reactions indicate that the albitization, K-feldspathization, and production of illite and fluorite occurred through the consumption of plagioclase with an inflow of H_4SiO_4, H^+, Al^{3+}, Fe^{2+}, Mg^{2+}, K^+, and F^- from the hydrothermal fluid, accompanied by an outflow of H_2O, Ca^{2+}, and Na^+ into the hydrothermal fluid (Fig. S6E).

Type C-1 (sample No. 6-6): Alteration of the rim of the plagioclase grain (Pl → Ab + Kfs + Cal + Fl). The reactant mineral in Type C-1 is plagioclase (No. J13 of Table 1), and the products are albite (No. 54), K-feldspar (No. 55), calcite (ideal $CaCO_3$), and fluorite

(ideal CaF_2). The reactions show that the albitization, K-feldspathization, and production of calcite and fluorite occurred through the consumption of plagioclase with an influx of H_4SiO_4, CO_2, H^+, Na^+, K^+ and F^-, accompanied by the outflow of H_2O, Al^{3+}, and Ca^{2+} into the hydrothermal fluid (Fig. S6F).

Type C-2 (sample No. 6-9): Alteration of the rim of the plagioclase grain (Pl → Ab + Kfs + Ill + Fl). Type C-2 plagioclase (No. 62 in Table S1) is the reactant and albite (No. 66), K-feldspar (No. 67), illite (No. J18), and fluorite (ideal CaF_2) are products. The reactions represent the albitization, K-feldspathization, and production of illite and fluorite though the consumption of plagioclase with an inflow of H_4SiO_4, Al^{3+}, Fe^{2+}, Mg^{2+}, K^+, and F^- from the hydrothermal fluid, accompanied by an outflow of H_2O, H^+, Ca^{2+}, and Na^+ into the hydrothermal fluid (Fig. S6G).

Type C-3-1 (sample No. 7-6): Alteration of the rim of the plagioclase grain (Pl → Ab + Kfs + Ill + Cal + Fl). The Type C-3-1 is characterized by plagioclase (No. 76 of Table 1) as the reactant and albite (No. J21), K-feldspar (No. J24), illite (No. J23), calcite (ideal $CaCO_3$), and fluorite (ideal CaF_2) as the products. The obtained reactions represent the albitization, K-feldspathization, and production of illite and fluorite by the consumption of plagioclase with an inflow of H_4SiO_4, CO_2, Al^{3+}, Fe^{2+}, Mg^{2+}, K^+, and F^- from the hydrothermal fluid, accompanied by an outflow of H_2O, H^+, Ca^{2+}, and Na^+ into the hydrothermal fluid (Fig. S6H).

Type C-3-2 (sample No. 9-4): Alteration of the rim of the plagioclase grain (Pl → Ab + Kfs + Ill + Cal + Fl). The reactant mineral of Type C-3-2 is plagioclase (No. J27 of Table S1) and the products are albite (No. J28), K-feldspar (No. J30), illite (No. J29), calcite (ideal $CaCO_3$), and fluorite (ideal CaF_2). The reactions show that the albitization, K-feldspathization and production of illite, calcite, and fluorite proceeded by consumption of plagioclase with an influx of H_4SiO_4, CO_2, H^+, Fe^{2+}, Mg^{2+}, (Na^+: volume constant), K^+, and F^- accompanied by the removal of H_2O, Al^{3+}, Ca^{2+}, and (Na^+: volume decrease of 4.4 vol%) into the hydrothermal fluid (Fig. S6I).

Type C-3-3 (sample No. 9-9): Alteration of the rim of the plagioclase grain (Pl → Ab + Kfs + Ill + Cal + Fl). The Type C-3-3 reactions involve the reactant plagioclase (No. 96 of Table S1) and the products albite (No. J32), K-feldspar (No. 99), illite (No. J33), calcite (ideal $CaCO_3$), and fluorite (ideal CaF_2). The overall reactions are characterized by the consumption of plagioclase with an inflow of H_4SiO_4, CO_2, Al^{3+}, Fe^{2+}, Mn^+, Mg^{2+}, Na^+, K^+ and F^- accompanied by an outflow of H_2O, H^+, and Ca^{2+} into the hydrothermal fluid (Fig. S6J).