

## Granitic Pegmatite of the Umanotani-Shiroyama Quartz-Feldspar Mine, Shimane Prefecture, Southwest Japan

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**Abstract:** The pegmatite deposits at the Umanotani-Shiroyama mine, the largest producer of K-feldspar and quartz in Japan and characterized by a simple mineralogy, dominated by quartz and K-feldspar with minor muscovite and plagioclase, are hosted in the ilmenite-series Masago Granite. The ore zones are classified into the following three zones based on the distance from the host granite inwards: (1) marginal; (2) transitional; and (3) central ore zones. Crystal grain size increases in this order with the resultant occurrence of gigantically grown crystals of quartz in the central ore zone. The evidences presented here reveal that the pegmatite deposits were formed during the latest stage of crystallization and differentiation of the granitic magma being responsible for the Masago Granite. They include: (1) close temporal and spatial association of the granite and the pegmatite deposits (about 95 to 90 Ma), (2) common occurrence of macroscopic “graphic intergrowth” displayed by quartz and K-feldspar in orebodies, (3) common occurrence of melt inclusions trapped in “ore quartz” and “ore K-feldspar”, (4) common occurrence of perthites observed in “ore K-feldspar” and (5) inheritance of oxygen isotopic signature, especially of quartz, from the surrounding biotite granite. The  $\delta^{18}\text{O}$  values of quartz, whether in the related igneous rocks or in ores, are almost the same, around +12 ‰, while those of K-feldspar in the igneous rocks is around +11‰ and those in ores are significantly depleted in  $^{18}\text{O}$ , about +7 to +8 ‰. It might be due to the difference in the exchange reaction rate of oxygen between quartz and K-feldspar. Abundant two-phase (liquid+gas) fluid inclusions are trapped in “ore quartz” and “ore K-feldspar”, as well as in the related igneous rocks. Their presence strongly indicates that hydrothermal fluids, possibly admixture of magmatic fluids released from the Masago Granite and circulating meteoric water, entered to the magmatic-hydrothermal system and circulated through the orebodies mainly during the later phase of the pegmatite formation. Measured homogenization temperatures of two-phase inclusions trapped in “ore quartz” are in the following ranges: 250 to 400 °C in the marginal ore zone; 230 to 370 °C in the transitional ore zone; 240 to 340 °C in the central ore zone. Also suggested is that late circulation of hydrothermal fluids lasted for not so long time as to affect the  $\delta^{18}\text{O}$  values of quartz within the magmatic-hydrothermal system, resulting in escape of the fluids from the system through ubiquitously observed quartz±muscovite veins and fissures.

**Keywords:** Umanotani-Shiroyama, pegmatite, melt inclusions, fluid inclusions, “perthites”, oxygen isotopes, Shimane Prefecture, Southwest Japan

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## 1. Introduction

The pegmatite deposits at the Umanotani-Shiroyama mine are located in Masuda City, Shimane Prefecture, Southwest Japan. This mine is the largest producer of quartz and K-feldspar in Japan with reserves of more than 1 million tonnes of ore (unpublished mine's data). The pegmatites are hosted in ilmenite-series biotite granite, called Masago Granite, occurring as a discordant pluton. They are typically characterized by the following features: (1) very simple mineral assemblage, mainly K-feldspar + quartz; (2) absence of internal mineral zonation; (3) absence of biotite, fluorite, gem minerals, including beryl, garnet and tourmaline, and strategic materials, including lithium, tantalum and beryllium; and (4) very low contents of fluxing components, such as boron, phosphorus, and fluorine, as well as REE or other rare elements.

Although granitic pegmatites have widely been studied in the world (e.g., Cerny, 1982; Brown and Ewing, 1986; Jahns, 1982; London, 1992, 1996), there have been very few studies in Japan. Recently, granitic pegmatites at the Kanamaru mine, Niigata Prefecture, Central Japan, were discussed based on mineralogy, petrochemistry, oxygen isotope composition, and K-Ar age determination (Nanbu *et al.*, 1997; Ishihara *et al.*, 1998). More recently, the present authors have established a temporal association of a series of felsic magmatic events with pegmatite formation at the Umanotani-Shiroyama mine by using K-Ar datings (Kihara *et al.*, 2005). No other important mineralogical and geochemical data, however, have so far been published.

In order to reveal the genesis of the Umanotani-Shiroyama pegmatite deposits, we here document their geological, petrographical, and geochemical characteristics, based on field observation, mineralogy, chemistry, fluid inclusion microthermometry, and oxygen isotope analysis.

## 2. Geological setting

### 2.1 Regional geology

The geology of the Masuda district (Fig. 1), which includes the Umanotani-Shiroyama mine and its surrounding area, is composed of the oldest rock units of the Sangun metamorphic rocks, called the Misumi Group, including Permian-Triassic phyllite, sandstone, chert and green rock, which have undergone high pressure/low temperature-type regional metamorphism, resulting in the formation of crystalline schists. The Early Mesozoic sedimentary rocks, called the Kanashi Group, characterized by a *mélange* facies, including Triassic to Jurassic chert and normal clastic rocks with upper Paleozoic limestone, have fault contact with the Sangun metamorphic rocks. These rocks are unconformably overlain by Late Cretaceous subaerial pyroclastic rocks of rhyolitic composition. The Late

Cretaceous granitic rocks intruded into these rocks and produced thermal metamorphism. Small-scale volcanism and plutonism occurred also in Tertiary time. The metamorphic and sedimentary rocks have been deformed with east-west fold axes. The granitic intrusions may have been controlled by the fold structures. Northeasterly faulting and fracturing became predominant from the Late Cretaceous onward.

Throughout the Masuda district, the largest pluton with east-west elongated ovoid shape, called Masago Granite has E-W and N-S dimensions of 8 km and 4 km, occurring as a stock. The pluton is of ilmenite-series biotite granite of various grain sizes with often porphyritic texture. In the west-central part, it hosts the large pegmatite deposits of the Umanotani-Shiroyama mine. In the marginal facies of the pluton, there occur small wolframite-quartz veins of the Masago mine (Tsuboya and Ishihara, 1961) and molybdenite-wolframite-scheelite-quartz vein deposits of the Kiyomizu mine (Soeda *et al.*, 1979; Watanabe and Soeda, 1981). A number of skarn and vein-type base metal deposits also occur around the Masago Granite (e.g., Hirowatari *et al.*, 1981).

### 2.2 Umanotani-Shiroyama Pegmatite Deposits

Geological plan and section of the Umanotani-Shiroyama mine are shown in Figs. 2 and 3. Igneous rocks exposed in the mine are the Late Cretaceous biotite granite, granite porphyry (coarse- and fine-grained) and aplite, as well as the small-scaled Tertiary breccia dike. The biotite granite occurs surrounding the pegmatite deposits as their host rock. Modal analyses of these granitic rocks indicate that they are all plotted in the region of granite in the IUGS classification. The granite porphyry is characterized by the presence of phenocrysts, up to 3 mm in size, of quartz, plagioclase and K-feldspar with minor amounts of biotite and muscovite. This porphyry is sandwiched between the biotite granite host and the orebodies and contains a xenolith of the biotite granite, in places. It becomes smaller in grain-size towards the orebodies (fine-grained granite porphyry), and is gradually changed into aplite, which is eventually graded into the orebodies. However, due to their strong similarity, the fine-grained granite porphyry and aplite are put together and called aplite (I), surrounding the orebodies. In the aplite (I), layered or massive pegmatites composed of quartz and K-feldspar are locally developed on a small scale, especially in the vicinity of the orebodies. As will be mentioned later, mineral compositions of ore change irregularly within the pegmatite deposits, and thus a clear mineral zonation is not observed. Aplite dikes or veins and quartz or quartz-muscovite veins commonly occur cutting the orebodies and related granitic rocks.

In the Umanotani-Shiroyama mine (Fig. 2), there

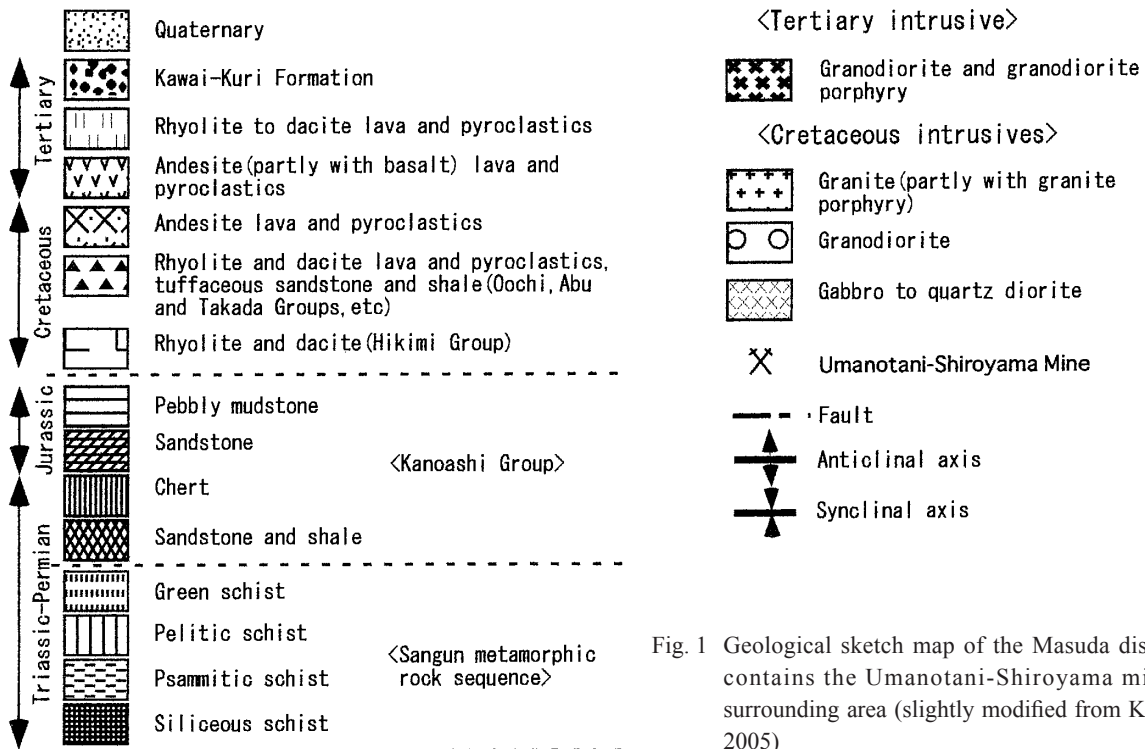
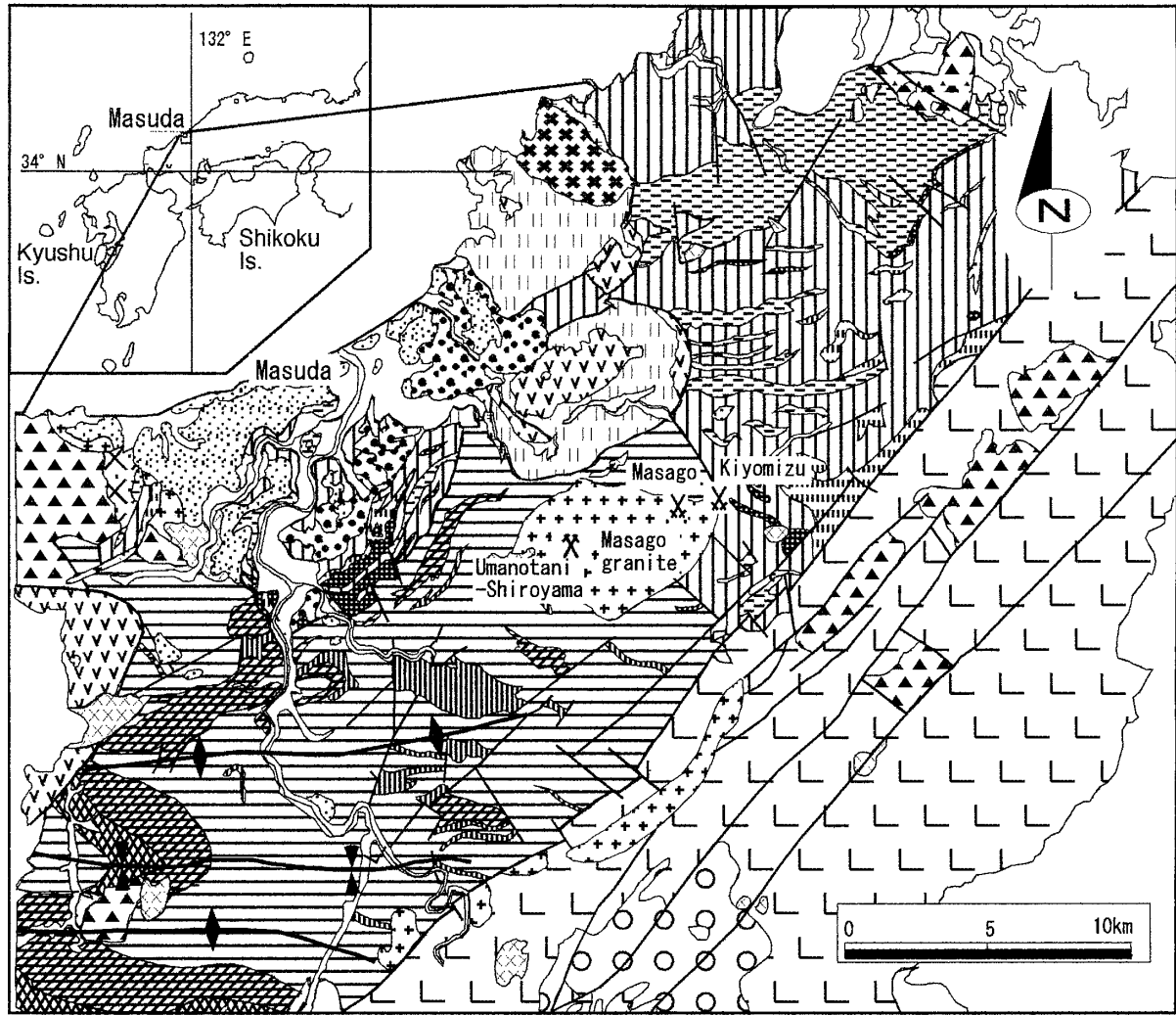


Fig. 1 Geological sketch map of the Masuda district which contains the Umanotani-Shiroyama mine and its surrounding area (slightly modified from Kihara *et al.*, 2005)

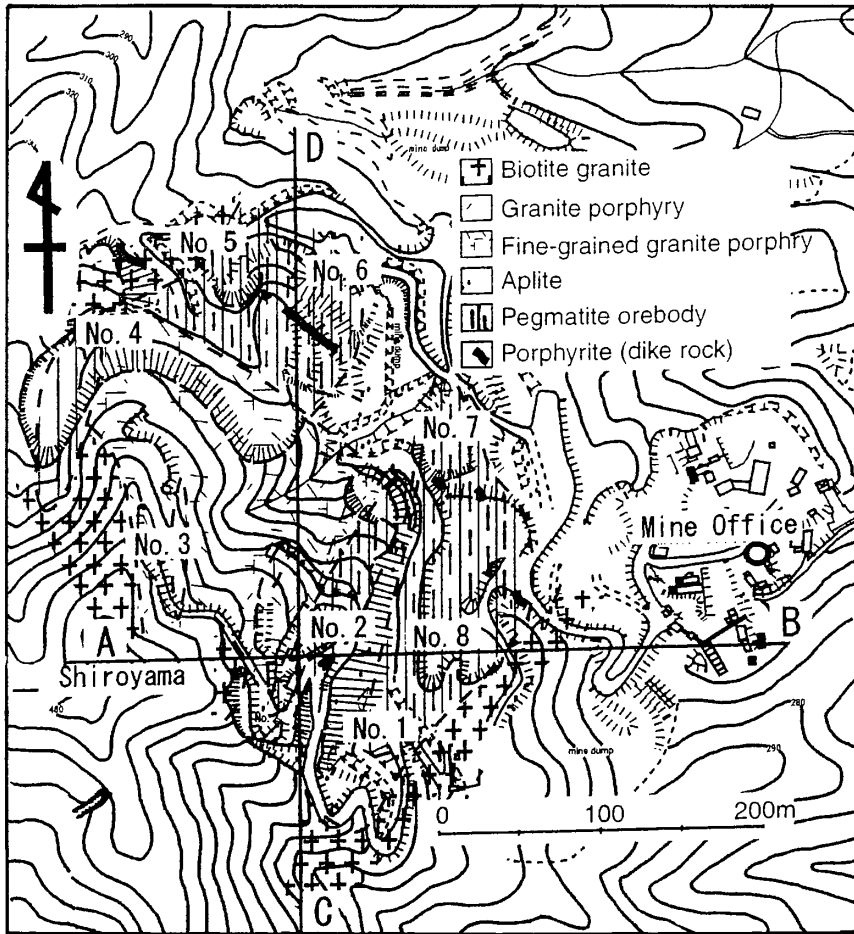


Fig. 2 Geological plan of the Umanotani-Shiroyama deposits (slightly modified from Kihara *et al.*, 2005).

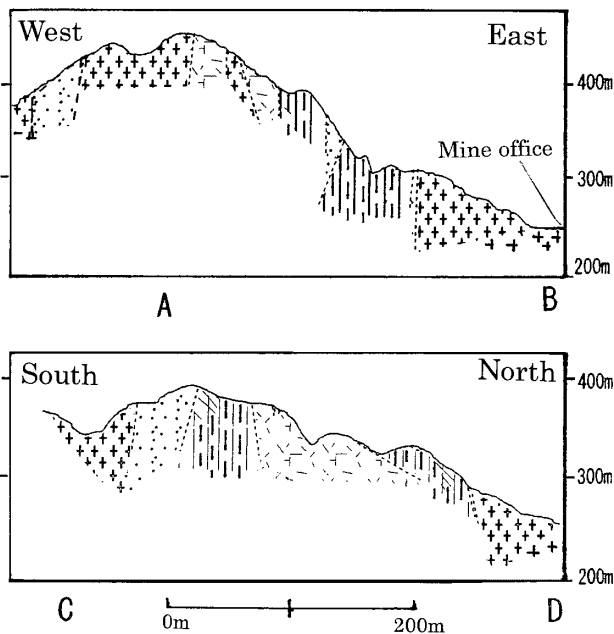


Fig. 3 Geological sections of the Umanotani-Shiroyama deposits (slightly modified from Kihara *et al.*, 2005). The section lines and the rock symbols are given in Fig. 2.

occur mainly five thick lenticular orebodies within the area of 400 m (N-S) × 300 m (E-W), with their arched distribution on the northeastern hillside of the Shiroyama Mountain (458 m above sea level). All the ore lenses have a long axis of mainly N-S direction with about 40° dip eastward (Fig. 3). Small-scaled lenticular orebodies, up to about 7 m in thickness and 20 m in length, are composed exclusively of K-feldspar, while much larger lenticular orebodies, up to about 50 m in thickness and 150 m in length, are composed of quartz, K-feldspar and biotite-free aplite. Such aplite, occurring as a matrix, is only observed within the orebodies, here called aplite (II), which is likely to have originally been the aplite (I). Quartz occurring as ore (here called “ore quartz”), semi-transparent or white with pale grayish tint, ranges from about 5 cm to 6 m in size with a hexagonal short prismatic form and also occurs as massive ores. Sometimes in the orebodies quartz shows a macroscopic “graphic intergrowth” with K-feldspar (Fig. 4). K-feldspar occurring as ore (here called “ore K-feldspar”), white to grayish white, sometimes pink in color, is euhedral to subhedral, with occasional development of Carlsbad twin, and also occurs



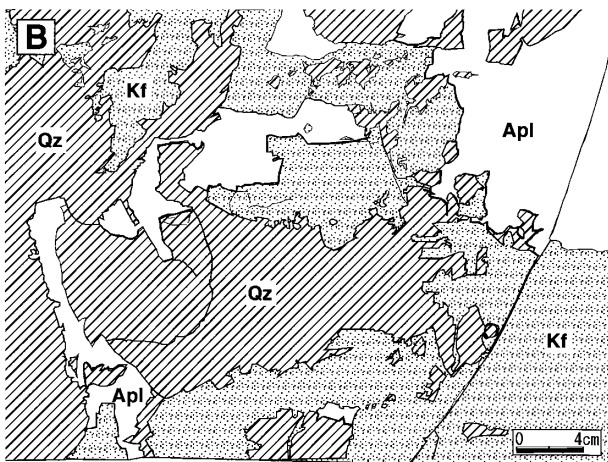
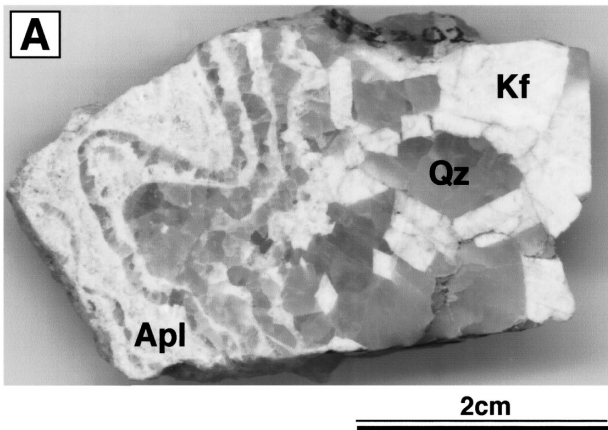


Fig. 4 Photographs showing a “graphic intergrowth” of quartz (Qz) and K-feldspar (Kf) observed in the orebodies.  
 (A) Well-crystallized quartz (gray to black) and K-feldspar (white and euhedral) with curving bands of quartz and aplite (II) (white to gray with Apl).  
 (B) Approximately equal amounts of quartz and K-feldspar with remnants of aplite (II) (Apl), forming “graphic intergrowth”.

as massive ore. Occurrence of muscovite includes: (1) quartz-muscovite veins; (2) muscovite films formed on the surface of euhedral quartz (rock crystal) and filling interstices of K-feldspar; and (3) euhedral crystals up to about 5 mm in size included in K-feldspar.

Using the in-situ modal analyses and classification criteria, the following three ore-types were identified: “feldspar ore” (>50%Kf, <30%Qtz); “quartz ore” (>60%Qtz); and “feldspar-quartz ore” (30<Qtz%<60). The “feldspar ore” is typically observed in the No. 2 orebody (Fig. 2). The “quartz ore” occurs usually in the central part of the orebodies. Gigantic rock crystals, up to 10 m in diameter and 17 m in length, are observable on the surface and underlying hidden “quartz ore”(unpublished mines data). In drusy parts formed by tectonic shearing, hexagonal short prismatic crystals of

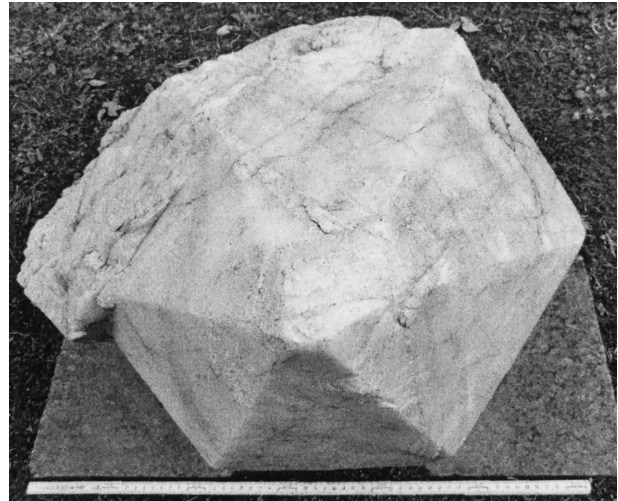


Fig. 5 Gigantic crystal of euhedral quartz from the “quartz zone”; its diameter is 35 cm. Collection of the Geological Museum, Tsukuba.

quartz, 55 cm along the c-axis in size (Fig. 5), are seen in the brecciated matrix of K-feldspar and the aplite (II). Growth of muscovite films is commonly observed on the surface of the large euhedral crystal of quartz. Usually, the “feldspar-quartz ore” is well developed in the Nos. 1, 2, 3 and 4 orebodies (Fig. 2). In places, macroscopic “graphic structure” is recognized in the “feldspar-quartz ore”. Also, locally, the aplite (II) is abundant in this zone. Massive “quartz ore”, composed predominantly of massive quartz with small amounts of K-feldspar and aplite (II), occurs only in the southern margin of the No. 1 orebody (Fig. 2).

The ore zones occurring in the mine are classified into the following three zones based on the distance from the biotite granite inwards: (1) marginal ore zone; (2) transitional ore zone; and (3) central ore zone.

The marginal ore zone is defined as a zone being adjacent to the biotite granite host, including the No. 2 orebody and the upper part of the No. 6 orebody. The ore zone changes gradually from the biotite granite through the aplite (I) to the No. 2 orebody. This zone is characterized by the presence of bamboo sprout-shaped rock crystals up to 30 cm wide with fine-grained aggregates of muscovite flakes distributed on their surfaces. Anhedral K-feldspar interstitially fills such rock crystals. Euhedral quartz crystals up to about 10 cm in size are common in the matrix of the aplite (II), which occurs as relics-like material. Euhedral muscovite up to 5 mm in size occurs in K-feldspar in the marginal ore zone.

In the transitional ore zone, being essentially the same as the marginal ore zone, quartz increases in size, up to about 50 cm, with increasing amounts of K-feldspar, while the aplite (II) decreases significantly in amount in the transitional ore zone. Quartz in the zone

occurs as massive aggregates formed in the aplite (II) and also as euhedral crystals. Massive aggregates of K-feldspar also occur in the aplite (II). Muscovite occurs as films on the surface of euhedral quartz crystals.

The central ore zone corresponds to the No. 8 orebody and its vicinity. Quartz in this zone occurs as a massive ore, characterized by its gigantic size more than 5 m with poorly developed euhedral habits. K-feldspar with anhedral habits occurs as interstice-fillings of the massive quartz. Neither the aplite (II) nor muscovite is observed in the zone.

### 3. Chronology of magmatic and hydrothermal events

Regarding the isotope age of the Masago Granite, Kawano and Ueda (1966) first showed a biotite age of 92 Ma, followed by Shibata and Ishihara (1974) in which K-Ar age of  $92.1 \pm 3.7$  Ma was obtained for muscovite separated from the greisen envelope of the Masago wolframite-quartz veins occurring in the northern margin of porphyritic phase of the Masago Granite. Ohira *et al.* (2004) indicated Rb-Sr whole-rock isochron age of  $96.7 \pm 3.4$  Ma and SrI of  $0.706498 \pm 0.00276$  for nine whole-rock samples, including 5 Masago Granites, 3 aplites (outside the mine), and 1 aplite (in the mine). Concerning the Umanotani-Shiroyama pegmatite deposits, a K-Ar age of  $88.9 \pm 2.7$  Ma was reported by Watanabe *et al.* (1998) for “ore K-feldspar”. Later, Kihara *et al.* (2005) have established chronology of the magmatic and hydrothermal events at the Umanotani-Shiroyama mine and its surrounding area based on K-Ar dating. According to them, the ilmenite-series biotite granite and granite porphyry, hosting the ore deposits, and biotites separated from these rocks have yielded K-Ar ages ranging from  $89.0 \pm 2.0$  to  $81.4 \pm 1.8$  Ma and  $95.2 \pm 2.1$  to  $93.7 \pm 2.1$  Ma, respectively (Fig. 6). Thus, the whole-rock K-Ar ages obtained are much younger than the biotite ages.

Since K-feldspar is the most abundant K-bearing phase in these granitic rocks, the resultant whole-rock ages obtained are assumed to be those of K-feldspar. Such difference in the ages may be ascribed to the difference in closure temperatures between biotite (mica) and K-feldspar, suggesting that both granitic rocks and associated pegmatite orebodies cooled slowly. Muscovite and K-feldspar separated from the ore zone have yielded K-Ar ages with the range of  $96.2 \pm 2.3$  to  $93.1 \pm 2.0$  Ma and  $87.3 \pm 1.9$  to  $80.7 \pm 1.8$  Ma, respectively. Muscovite in quartz-muscovite veins in the ore zone and in the granite porphyry have yielded K-Ar ages of  $90.4 \pm 2.0$  Ma and  $76.3 \pm 1.7$  Ma. K-feldspar is much younger in age than coexisting muscovite. It is noted that the K-Ar ages of biotite separates and the whole-rock ages are identical to those of muscovite and K-feldspar in the ore zone, respectively. The analyti-

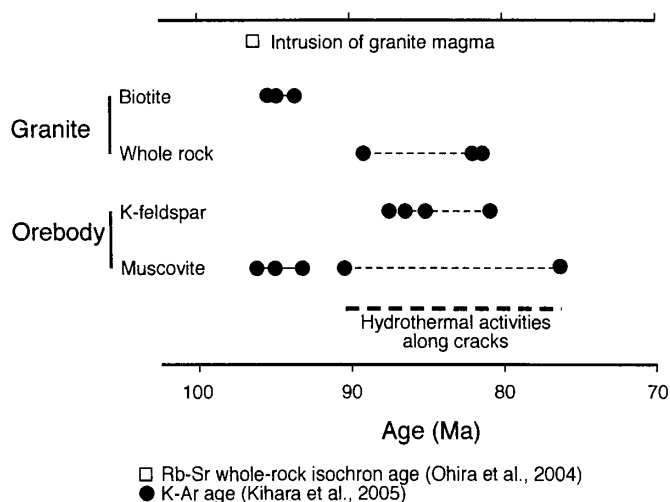


Fig. 6 K-Ar ages of magmatic and hydrothermal events in the Umanotani-Shiroyama mine and its surrounding area (slightly modified from Kihara *et al.*, 2005). Rb-Sr whole-rock isochron age measured on the Masago Granite by Ohira *et al.* (2004) is also shown.

cal results obtained are schematically drawn in Fig. 6, along with the Rb-Sr whole-rock isochron age by Ohira *et al.* (2004).

After these magmatic and hydrothermal events, there occurred emplacement of andesitic breccia dike and felsic dike, probably in Tertiary time.

### 4. Experimental procedures

To determine major and trace element abundances in the related igneous rocks and “ore K-feldspar”, a fully automated wavelength-dispersive Rigaku zsx-101e XRF system, equipped with a 3kw generator and Rh/W dual anode X-ray tube, was used following the method described in Shipboard Scientific Party (2001). Microprobe analyses were made on K-feldspar, plagioclase and muscovite, as well as on melt inclusions, using a JEOL JCMA-733 II electron-microprobe analyzer installed at the Instrument Center for Chemical Analysis, Hiroshima University. Microthermometries of fluid inclusions trapped in quartz and melt inclusions were done using a Linkam heating/freezing stage, TH-600RH. Following the method described by Kita and Matsubaya (1983), oxygen isotopic ratio measurements of samples, including (1) various related granitic rocks (whole rocks), (2) their constituent minerals (K-feldspar and quartz concentrates), and (3) ore (K-feldspar and quartz concentrates) were carried out using a mass spectrometer MAT 250. The reproducibility of this measurement is better than  $\pm 0.2\%$ , and through this analysis we got results that the  $\delta^{18}\text{O}$  of NBS-28 is  $+9.8\%$  relative to SMOW (average of 6 measurements).

## 5. Presentation of data

### 5.1 Chemistry of related igneous rocks, “ore K-feldspar” and muscovite

Analytical results of some major elements and some minor elements of the related granitic rocks are plotted against  $\text{SiO}_2$  contents in Fig. 7. For comparison, the chemical data for the Kanamaru deposit (Nanbu *et al.*, 1997) are also shown in the figure, together with the average compositions of the Japanese granitoids (S. Ishihara, unpublished data). Compared with the Japanese granitoids, the studied granitic rocks are high in both  $\text{K}_2\text{O}$  and Rb for all the  $\text{SiO}_2$  range, but slightly low in the total  $\text{Fe}_2\text{O}_3$ ,  $\text{MgO}$ ,  $\text{CaO}$ ,  $\text{TiO}_2$  and  $\text{P}_2\text{O}_5$  and significantly low in Sr. Some of the aplite II are very depleted in  $\text{Fe}_2\text{O}_3$ ,  $\text{MgO}$  and Sr, because of later hydrothermal alteration.

Except for  $\text{P}_2\text{O}_5$  and Rb, both the Umanotani-Shiroyama and Kanamaru deposits show very similar chemical variations. The ratios of Rb/Sr of igneous rocks, roughly indicating a degree of differentiation, are in the range of 2 to 69 (mostly less than about 10) in the biotite granite and of 16 to 84 (mostly larger than about 30) in the aplite (II). Rubidium and Sr contents of “ore K-feldspar” are in the ranges of 279 to 878 ppm (mostly 330 to 530 ppm) and of 6 to 49 ppm (mostly 15 to 35 ppm) with their Rb/Sr ratios ranging from 7 to 89 (mainly 25 to 65).

Muscovite with K-Ar age of 90.4 Ma (Kihara *et al.*, 2005) in the quartz-muscovite vein in the transitional zone is rather coarse-grained and higher in contents of  $\text{Al}_2\text{O}_3$  averaging 30.3 wt.% (26.7 wt.%) and  $\text{Na}_2\text{O}$  averaging 0.29 wt.% (0.06 wt.%) and lower in  $\text{MgO}$  averaging 0.15 wt.% (1.40 wt.%) than muscovite with K-Ar age of 76.3 Ma (Kihara *et al.*, 2005) in the quartz-muscovite vein in the granite porphyry, which are shown in parenthesis. The  $\text{Na}/(\text{Na}+\text{K})$  values are higher in the former averaging 0.039 than the latter averaging 0.016. Based on chemical compositions, Monier *et al.* (1984) distinguished three generations of muscovite, magmatic, late- to postmagmatic and hydrothermal, from the Millevaches massif, France. It seems that the former muscovite (90.4 Ma) is closer to late to postmagmatic one, while the latter muscovite (76.3 Ma) falls in the region of hydrothermal one. Plagioclase occurs as euhedral crystals and also as perthites in the “ore K-feldspar”.

### 5.2 Perthites observed in “ore K-feldspar” and its triclinicity

Fine lamellar or stringlet intergrowths of plagioclase are commonly observed in “ore K-feldspar”, as mentioned previously. It seems likely that they are exsolution products formed during cooling, of which implications will be discussed in a later section. The chemical

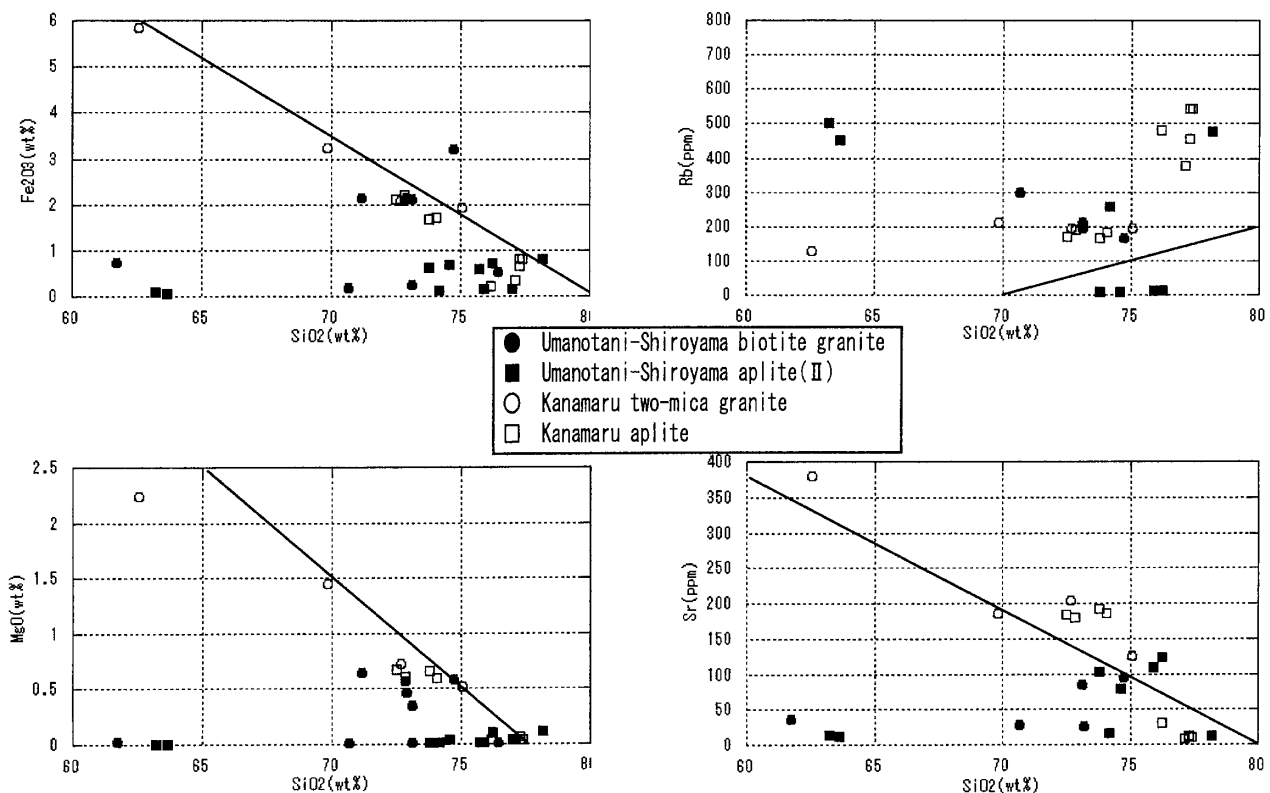


Fig. 7 Chemical compositions of some major and trace elements of the related granitic rocks plotted against  $\text{SiO}_2$  contents.



compositions for coexisting K-feldspar and plagioclase are in the following ranges: 14.49~16.71 wt.%K<sub>2</sub>O (mainly 15.4~16.5 wt.%K<sub>2</sub>O), 0.32~1.64 wt.%Na<sub>2</sub>O (mainly 0.4~0.6 wt.%Na<sub>2</sub>O), 0~0.1 wt.%CaO (mostly Ca-free) for K-feldspar (49 point analyses); and 0.11~0.48 wt.%K<sub>2</sub>O (mainly 0.11~0.19 wt.%K<sub>2</sub>O), 10.23~11.95 wt.%Na<sub>2</sub>O (mainly 10.7~11.6 wt.%Na<sub>2</sub>O), 0.06~2.34 wt.%CaO (mainly 0.1~1.4 wt.%CaO) for plagioclase (34 point analyses), respectively. Relatively large euhedral plagioclase crystals up to about 150 µm in size in association with the fine lamellar ones are rich in Ca, up to about 4.5 wt.% CaO and not of exsolution products. Though not shown here, chemical compositions of perthites observed in the biotite granite and aplite are very similar to those of "ore K-feldspar".

Triclinicity (cf. Tilling, 1968) was measured on the various K-feldspar crystals. Compared with the triclinicity values of the Kanamaru K-feldspar, 0.71 to 0.80 (Nanbu *et al.*, 1997), those of the Umanotani-Shiroyama one are very small as low as zero, except for those of the granite porphyry ( $\Delta=0.76$ ) and the feldspar-quartz zone ( $\Delta=0.30$ ). Microscopic observation of the K-feldspar with the triclinicity value of 0.30 has revealed the presence of microcline texture shown by albite twin perpendicular to pericline twin.

### 5.3 Melt inclusions observed in "ore quartz" and "ore K-feldspar"

Melt inclusions, usually less than about 15 µm in diameter with the maximum diameter up to about 30 µm, are observed not only in the biotite granite, granite porphyry and aplite (both I and II), but also in "ore quartz" and "ore K-feldspar". Representative photomicrographs of the melt inclusions are shown in Fig. 8A. In most cases, original glass has been changed into crystalline phases as clearly shown with crossed polars (Fig. 8B). Microscopic observations show three types of melt inclusions at room temperatures. They include: (1) subrounded to polygonal inclusions with abundant daughter minerals, occupying more than about 80% of the total inclusion volume, largely composed of silicate daughter minerals with or without gas bubble sometimes being a little distorted; (2) most frequently-occurring spherical to ellipsoidal inclusions with a large gas bubble in the center and radial growth of fibrous crystalline materials, occupying about 30 to 50% of the total inclusion volume, from the center towards the margin; and (3) rarely-occurring small, up to 5 µm in size, subrounded or quadrilateral inclusions composed of homogeneous "glass" and tiny gas bubble.

Microscopic observations and EPMA analyses of the melt inclusions trapped in "ore quartz" reveal that they are composed mostly of silicate phases, including K-feldspar, quartz, chlorite and muscovite, with or without somewhat distorted vapor phase. Preliminary

cooling experiments and Raman spectroscopy on those melt inclusions have not revealed the presence of appreciable amounts of CO<sub>2</sub>.

### 5.4 Two-phase fluid inclusions trapped in "ore quartz"

Microthermometries of fluid inclusions trapped in quartz in the biotite granite, granite porphyry and aplite and also of those in "ore quartz", showing occurrence in random, three-dimensional distribution, with different concentrations in adjacent zones (e.g., Roedder, 1979), were done. Fluid inclusions showing occurrence as planar groups outlining healed fractures have been measured separately. As seen in Fig. 8 (C, D), they are rounded, ovoid or quadrilateral in shape and mostly as small as 10 µm in size with the maximum size up to about 30 µm and composed of liquid+gas (two-phase type). Although CO<sub>2</sub>-rich fluid inclusions (two- or three-phase) were reported from pegmatitic minerals (e.g., Takenouchi, 1971; London, 1986a; Trumbull, 1995; Nabelek and Tarnes, 1997; Nanbu *et al.*, 1997) and from the Kiyomizu Mo-W-quartz veins hosted by the Masago Granite (Watanabe *et al.*, 1981), neither CO<sub>2</sub> nor chloride(s) have been recognized in those Umanotani-Shiroyama quartz samples. Homogenization temperatures are in the following ranges: 250~400 °C (highest mode of about 320 °C) for the marginal ore zone; 230~370 °C (highest mode of about 300 °C) for the transitional ore zone; 240~340 °C (highest mode of about 290 °C) for the central ore zone. Salinities in wt.%NaCl equivalent obtained are in the following ranges: 0.4~6.9 wt.%NaCl eq. (mainly 2~5 wt.%NaCl eq.) for the marginal and transitional ore zones. However, those for the central ore zone show a wide variation ranging from 0.9 to 12.0 wt.%NaCl eq., with bimodal distribution mainly 2~5% and 7~10 wt.%NaCl eq. In Fig. 9, the measured salinities are plotted against homogenization temperatures.

### 5.5 Oxygen isotope compositions

The δ<sup>18</sup>O values measured relative to SMOW are given in Table 1 and depicted in Fig. 10. For comparison, those for the Kanamaru pegmatite deposits (Ishihara *et al.*, 1998) are also shown in the figure. Our results are in the following ranges: (1) +10.7‰ for the whole rock (biotite granite); (2) +12.1‰ for the igneous quartz; (3) +10.5 and +10.6‰ for the igneous K-feldspar; (4) +8.5‰ for the whole rock (aplite (I)); (5) +11.7‰ for the aplite (I) quartz; (6) +7.6‰ for the aplite (I) K-feldspar; (7) +8.2‰ for the whole rock (aplite (II)); (8) +11.9‰ for the aplite (II) quartz; (9) +7.3‰ for the aplite (II) K-feldspar; (10) for "ore quartz", +12.2‰ (marginal ore zone), +11.6‰ (transitional ore zone), +12.3‰ (central ore zone); (11) for "ore K-feldspar" with K-Ar age of 88.9±2.7 Ma, +6.9‰ (marginal ore



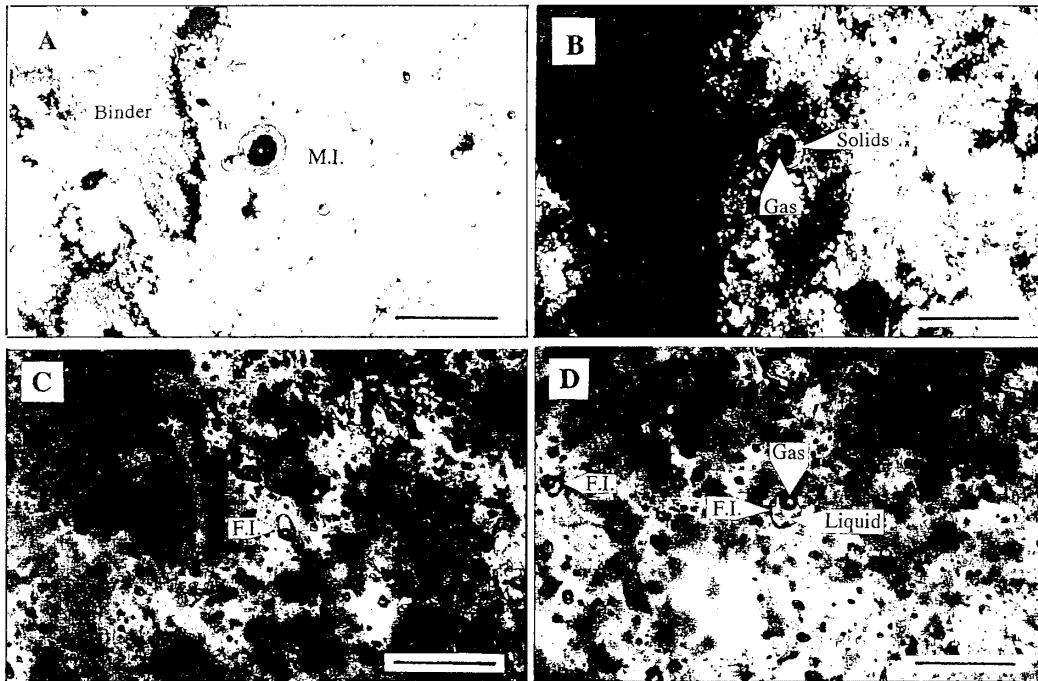


Fig. 8 Photomicrographs showing “melt inclusions” and fluid inclusions observed in “ore quartz”. Bar scales indicate 50 µm long.  
 (A) “Melt inclusions” with solid phases (devitrified) and gas in plane polarized light.  
 (B) Ditto with crossed polars.  
 (C) Relatively large-sized fluid inclusions (liquid+gas).  
 (D) Relatively large-sized fluid inclusions (liquid+gas).

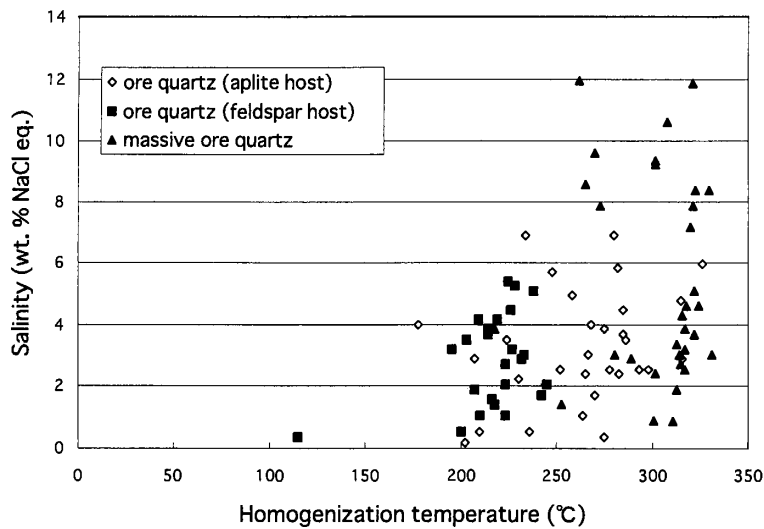


Fig. 9 Salinities (in wt.%NaCl eq.) plotted against homogenization temperatures

zone), +6.8‰ (transitional ore zone), +8.3‰ (central ore zone).

## 6. Discussions

### 6.1 Implication of the aplite (II) existing in the ore-bodies

Except for the central ore zone, the aplite (II) is

commonly observed throughout the mine. The limited distribution of the aplite (II) with pegmatitic ore quartz and K-feldspar should be noticed (Fig. 4A, B). Euhedral crystals of quartz up to about 10 cm in length and sometimes K-feldspar are well developed in the aplite (II). On the other hand, such well-developed crystals are not usually observed in the aplite (I), suggesting

Table 1 Oxygen isotopic data. Two figures underlined are duplicate results.

sample	Lithology	$\delta O$ (W.R.)	$\delta O$ (Qtz)	$\delta O$ (Kf)
(1) USGR-2WR	Medium-grained Bi granite collected at the No.2 working face	+10.7		
(2) USGR-2Qtz	Qtz separated from USGR-2WR		+12.1	
(3) USGR-2Kf	Kf separated from USGR-2WR			+10.5, +10.6
(4) USAPI-2WR	Bi-bearing aplite (1) collected at the No.2 working face	+8.5		
(5) USAPI-2Qtz	Qtz separated from USAPI-2WR		+11.7	
(6) USAPI-2Kf	Kf separated from USAPI-2WR			+7.6
(7) USAPI-2WR	Bi-free aplite (H) collected at the No2 working face	+8.2, +8.2		
(8) USAPII-2Qtz	Qtz separated from USAPII-2WR		+11.9	
(9) USAPII-2Kf	Kf separated from USAPII-2WR			+7.3
(10) US1F-1 Qtz	Qtz separated from the ore collected at the No.1 working face		+12.2	
(11) US1F-1Kf	Kf separated from US 1 F-1 Qtz			+6.8
(12) US2F-2Qtz	Qtz separated from the ore collected at the No.2 working face		+12.2, +12.2	
(13) US2F-2Kf	Kf separated from USZF-2Qtz			+6.9
(14) US5F-3Qtz	Qtz separated from the ore collected at the No.5 working face		+12.2	
(15) US5F-3Kf	Kf separated from US5F-3Qtz			+8.2, +8.3
(16) US6F-1Qtz	Qtz separated from the ore collected at the No.6 working face		+11.6	
(17) US6F-1Kf	Kf separated from US6F-1Qtz			+7.3

W.R. = whole rock; Qtz = quartz; Kf = K-Feldspar; Bi = biotite

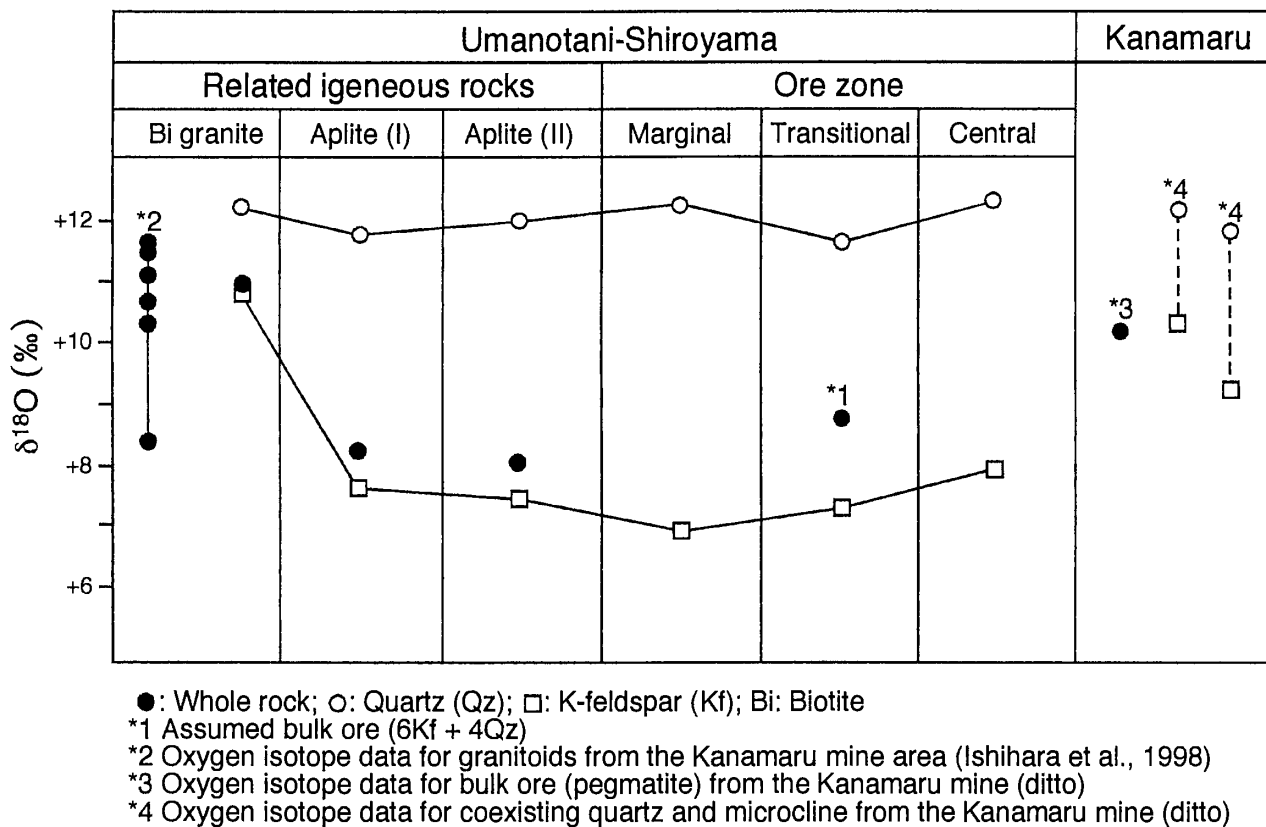


Fig. 10 Oxygen isotopic variations of related granitic rocks and ores

that the pegmatite deposits might have been formed by replacing the earlier aplite (I) during the late stage differentiation of a series of granitic magmas. In addition, it is also noted that throughout the orebodies, macroscopic “graphic intergrowth” shown by well-crystallized quartz and K-feldspar (Fig. 4A, B) is often recognized in the “feldspar-quartz ore”, suggesting its magmatic origin of K-rich granitic magma.

### 6.2 Implication of perthites observed in “ore K-feldspar”

Perthites show a regular arrangement of plagioclase (close to albite in chemical composition), which is assumed to be internally controlled by the crystal structure of alkali feldspar (K-feldspar), suggesting that they are of exsolution products formed on cooling of the alkali feldspar. If this is the case, possible temperatures of exsolution can be evaluated by using the results by Smith and Parsons (1974) who completed an experimental study on the alkali feldspar solvus at 1 kbar. The resultant temperatures range from about 500 to 300 °C, corresponding to temperatures at which diffusion of K and Na has stopped. Therefore, beginning of exsolution would have occurred at higher temperatures than those temperatures observed. However, there is a possibility that the alkali feldspar reacted with later Na-rich fluids to form an intergrowth of K-feldspar and plagioclase, as will be mentioned later. It is emphasized that plagioclase thus formed does not have a transition twin and that K-feldspar is not changed into microcline. Further detailed discussion based on crystal growth theory and electron diffraction is beyond the scope of this study.

### 6.3 Implication of melt inclusions observed in “ore quartz” and “ore K-feldspar”

As described before, melt inclusions are common in ores and related igneous rocks. Both types of inclusions, melt inclusions and two-phase fluid inclusions stated later, occur separately, suggesting that they are not related to each other and different in origin. Therefore, it is reasonable to state that only melts trapped as melt inclusions in “ore quartz” and “ore K-feldspar” were directly related to the formation of the pegmatite deposits. This statement is strongly supported by the results of oxygen isotopic ratio measurements. This is because “ore quartz” has inherited its oxygen isotopic signature from that of the related igneous rocks, though that of “ore K-feldspar” alone is not preserved.

Studying the melt inclusions trapped in quartz in F-, B- and P-rich pegmatites of the Variscan Ehlenfriedersdorf complex, Germany, Thomas *et al.* (2000) found a complete miscibility between silicate melts and hydrous fluids at low pressure. Based on microscopic and microanalytical studies, they recognized two main

types of melt inclusions at room temperature. The first type (A-type) consists of crystals of different daughter minerals such as K-feldspar, quartz, white mica, topaz, and sometimes berlinite, borates, and others with a liquid plus vapor phases of 5 to 20 vol.% of the inclusion. The second type (B-type) contains the same phases; however, liquid plus vapor occupy between 30 and 50 vol.% of the inclusion. Both types of melt inclusions were subjected to heating experiments. The partially crystallized melt inclusions were rehomogenized at 1 kbar between 500 and 712 °C by conventional rapid-quench hydrothermal experiments. The two melt inclusion types were interpreted as two coexisting melts which had been trapped simultaneously on both sides of a two-melt solvus, indicating that the former type represents a water-poor silicate melt and the latter type a water-rich melt (Thomas *et al.*, 2000).

In the Umanotani-Shiroyama pegmatite deposits, three types of melt inclusions are distinguished. The first type is corresponding to the A-type of Thomas *et al.* (2000) and the second one possibly to the B-type. However, the second type is different from the B-type in that in the former, the bubble occurs in the center of the inclusion and fibrous crystalline material is growing radially from the bubble towards the margin without euhedral to subhedral crystals. Although we have not carried out heating experiments on these melt inclusions, there is a possibility that they might represent a miscibility between silicate melts and hydrous fluids at low pressure. Heating experiments and Raman spectroscopy for H<sub>2</sub>O and CO<sub>2</sub> analysis of the melt inclusions are required for further discussion.

### 6.4 Implication of oxygen isotope data

It is interesting to notice the fact that the  $\delta^{18}\text{O}$  values of quartz from the Umanotani-Shiroyama mine, whether in the granitic host or in ores, are almost the same, about +12‰. Such  $\delta^{18}\text{O}$  values of “ore quartz” might represent a very closer approximation to the original isotopic composition of the granitic melts, probably being responsible for the pegmatite deposits. As to  $\delta^{18}\text{O}$  values of quartz in granitic rocks in Southwest Japan, it has been stated that those in the magnetite-series granitoids in the Sanin district, the Japan Sea side, are about +10‰, while those in the ilmenite-series granitoids in the Sanyo district, the Pacific Ocean side, are about +12‰ (Matsuhisa *et al.*, 1972). It is noted that the  $\delta^{18}\text{O}$  values of quartz in the medium-grained biotite granite host in the Umanotani-Shiroyama mine area accord with those of the ilmenite-series granitoids in the Sanyo district.

On the other hand, the  $\delta^{18}\text{O}$  values of K-feldspar reveal a different behavior from those of quartz. Namely, from the granitic host towards aplites (I and II), they show a striking decrease, as much as +4‰, and from



the aplites towards the ores, they are almost the same. Such decreasing tendency, but somewhat smaller spread, was also recognized at the Kanamaru pegmatite deposits (Ishihara *et al.*, 1998).

The fractionation between coexisting quartz and K-feldspar in the medium-grained biotite granite host is +1.6‰. If fractionation factors determined by Matsuhisa *et al.* (1979) are applied, this fractionation corresponds to an equilibrium fractionation at 356 °C, being much lower than the formation temperature of the granite, suggesting that reliable geothermometric data can not be obtained mainly due to isotopic exchange after their crystallization.

On the contrary, the large fractionation in the coexisting quartz and K-feldspar pair in the ore (+3.9~+5.4‰) shows equilibrium temperatures of 120 to 170 °C, which is unreasonable for its formation temperature and strongly suggests disequilibrium between the pair. Because there is no significant difference in  $\delta^{18}\text{O}$  values of quartz among the host granite, aplites and “ore quartz”, this disequilibrium suggests some changes in  $\delta^{18}\text{O}$  values of K-feldspar through later processes. This is possibly due to selective depletion of  $\delta^{18}\text{O}$  values of K-feldspar. Such depletion could be brought about by oxygen isotopic exchange with  $^{18}\text{O}$ -depleted later hydrothermal fluids as evidenced from abundant two-phase fluid inclusions trapped in “ore quartz” and “ore K-feldspar”. If the  $\delta^{18}\text{O}$  values of quartz (+12.2‰) are least exchanged or left almost unchanged after its formation, it is estimated that “ore K-feldspar” should have  $\delta^{18}\text{O}$  values of about +10 to +11‰ for the formation temperature range of about 300 to 500 °C, as inferred from the perthite compositions.

### 6.5 Implication of abundant two-phase fluid inclusions in “ore quartz” and “ore K-feldspar”

It is characteristic that two-phase fluid inclusions are abundant in the “ore quartz” and “ore K-feldspar”, as well as in related igneous rocks and they are not related to melt inclusions. Possibly in the latest stage of the pegmatite formation, magmatic fluids would be liberated from the felsic melts and mixed with local meteoric water to form hydrothermal fluids, part of which were trapped as two-phase inclusions in both “ore quartz” and “ore K-feldspar”, as well as in the igneous minerals. The trapped two-phase fluid inclusions show a wide range variation of homogenization temperatures and salinities, that is, 230~370 °C and 0.4 to 12.0 wt.%NaCl eq., respectively. The remaining fluids were eventually lost from the magmatic-hydrothermal system through quartz±muscovite veins and fissures cutting the orebodies and related igneous rocks throughout the mine. It is possible that such fluids might have stimulated the formation of microcline and/or “perthite-like” materials developed in “ore K-feldspar”. Also, it can

be said that if both “ore quartz” and “ore K-feldspar” would have reacted with such hydrothermal fluids for a long time, both quartz and K-feldspar would have exchanged with the fluids, with the resultant  $^{18}\text{O}$  depletion. This means that life time of the magmatic-hydrothermal system would not be long enough to affect the oxygen isotopic compositions of “ore quartz”.

Using fluid inclusion data and phase-equilibrium experiments, London (1986a, b) found support for a continuous transition from magma to aqueous vapor in the Tanco rare-element pegmatite, Manitoba, Canada and in the miarolitic pegmatites of Afghanistan. However, such continuous transition has never been recognized in the Umanotani-Shiroyama pegmatite deposits.

### 6.6 Formation history and genetic model

A series of ilmenite-series felsic magmas intruded into older rocks, including the Misumi Group, the Kanoashi Group and the Late Cretaceous subaerial pyroclastic rocks. Their magmatic differentiation and crystallization started at about 97 Ma from the peripheral part towards inside, forming a pluton, the Masago Granite. At the latest stage of differentiation of the felsic magmas, the pegmatite deposits were formed closely related to aplite at about the same time (95 to 90 Ma) in the uppermost part of the Masago Granite. Parts of the differentiated granitic melts were trapped as melt inclusions in “ore quartz” and “ore K-feldspar”. Such magmatic origin of the pegmatite deposits is independently supported by Rb-Sr isotope study by Ohira *et al.* (2004). The resultant  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $^{87}\text{Rb}/^{86}\text{Sr}$  ratios measured on “ore K-feldspar”, though very few in sample number, are plotted on the same isochron obtained for whole-rock samples (5 granites and 4 aplites), suggesting that aplites and related pegmatite deposits are differentiation products of a series of ilmenite-series magmatism responsible for the Masago Granite.

Magmatic fluids derived from the crystallizing pluton were mixed with meteoric water to form hydrothermal fluids. The resultant fluids penetrated the peripheral part of the deposits and circulated through them. Part of the fluids was trapped as two-phase inclusions in “ore quartz” and “ore K-feldspar”, as well as in the rock-forming minerals of the related igneous rocks. Such fluids were eventually lost through the ubiquitous quartz±muscovite veins and fissures, cutting the orebodies and related igneous rocks. A genetic model illustrating the formation process of the pegmatite deposits is schematically depicted in Fig. 11.

One of the characteristic features of pegmatite deposits is a gigantic size of constituent minerals, including euhedral forms of quartz (rock crystal). The quartz is latest crystallized mineral, so that at the final stage of the pegmatite formation, there must have been open-space at the top of the granitic stock. Therefore, the

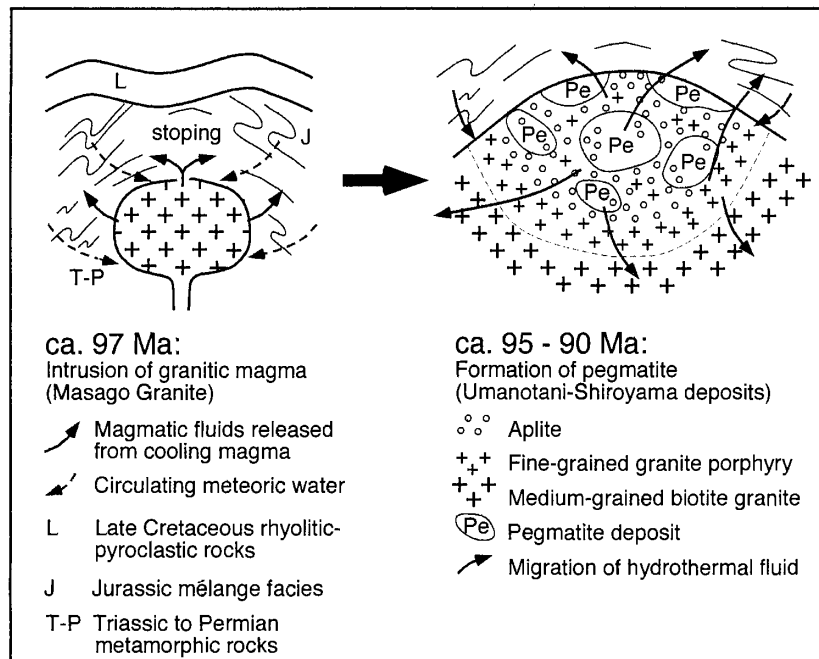


Fig. 11 Genetic model schematically illustrating the formation of pegmatite deposits on the uppermost part of the Masago Granite at the Umanotani-Shiroyama mine. (Not to scale)

emplacement level of the Masago Granite might have been shallow. We have not discussed the mechanism for such gigantic growth of constituent minerals, but only described here. This problem, though essentially important, is left for future investigation, as well as the absence or paucity of gem and strategic minerals, fluxing components and REE.

## 7. Conclusions

The evidences presented here reveal that the pegmatite deposits at the Umanotani-Shiroyama mine are of typical granitic pegmatite in origin. They include: (1) close temporal and spatial association of the ilmenite-series Masago Granite and the pegmatite deposits, (2) common occurrence of macroscopic “graphic intergrowth” displayed by quartz and K-feldspar in the ore, (3) common occurrence of melt inclusions trapped in “ore quartz” and “ore K-feldspar”, (4) common occurrence of perthites observed in “ore K-feldspar” and (5) inheritance of oxygen isotopic signature, especially of quartz, from the surrounding biotite granite. This conclusion is independently supported by Rb-Sr isotope study by Ohira *et al.* (2004). The presence of abundant two-phase (liquid+gas) fluid inclusions trapped in “ore quartz” and “ore K-feldspar”, as well as in those in the related igneous rocks, strongly indicates that hydrothermal fluids, possibly admixture of magmatic fluids released from the Masago Granite and circulating meteoric water, entered the magmatic-hydrothermal system

and circulated through the orebodies during the later phase of the pegmatite formation. It seems very likely that such hydrothermal fluids were responsible for the formation of microcline in some of “ore K-feldspar” and for its striking  $^{18}\text{O}$  depletion.

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## References

- Brown, G. E. Jr. and Ewing, R. C. (1986) Introduction to the Jahns Memorial Issue. *Am. Mineral.*, **71**, 233-238.
- Cerny, P. (1982) Petrogenesis of granitic pegmatites. In: Cerny, P. (ed.) *Granitic pegmatites in science and industry. Mineral Assoc. Canada Short Course*

- Handbook 8, 405-461.
- Hirowatari, F., Soeda, A. and Motomura, Y. (1981) Geology and ore deposits of the Masago granitic mass and its surrounding area. *Mining Geology Special Issue*, No. 9, 1-14 (in Japanese with English abstract).
- Ishihara, S., Hamano, K. and Ikegami, A. (1998) Isotopic evaluation on the genesis of the Kanamaru pegmatite deposit, Niigata Prefecture, Japan. *Resource Geol.*, **48**, 1-6.
- Jahns, R. H. (1982) Internal evolution of pegmatite bodies. In: Cerny, P. (ed.) *Granitic pegmatites in science and industry. Mineral Assoc. Canada Short Course Handbook 8*, 293-346.
- Kawano, Y. and Ueda, Y. (1966) K-Ar dating on the igneous rocks in Japan (V) – Granitic rocks in southwestern Japan. *Jour. Japan. Assoc. Mineral. Petrol. Econ. Geol.*, **56**, 191-211 (in Japanese with English abstract).
- Kihara, S., Hoshino, K., Watanabe, M., Nishido, H. and Ishihara, S. (2005) K-Ar ages of granitic magmatism and related pegmatite formation at the Umanotani-Shiroyama mine, Shimane Prefecture, Southwest Japan, and their bearings on cooling history. *Resource Geol.*, **55**, 123-129.
- Kita, I. and Matsubaya, O. (1983) F<sub>2</sub>-technique for the oxygen isotopic analysis of silica Minerals. *Rept. Research Inst. Underground Resources, Mining College, Akita Univ.*, **48**, 25-34 (in Japanese with English abstract).
- London, D. (1986a) The magmatic-hydrothermal transition in the Tanco rare-element pegmatite: evidence from fluid inclusions and phase equilibrium experiments. *Am. Mineral.*, **71**, 376-395.
- London, D. (1986b) Formation of tourmaline-rich pockets in miarolitic pegmatites. *Am. Mineral.*, **71**, 396-405.
- London, D. (1992) The application of experimental petrology to the genesis and crystallization of granitic pegmatites. *Can. Mineral.*, **30**, 499-540.
- London, D. (1996) Granitic pegmatites. *Trans. Roy. Soc. Edinburgh Earth Sci.*, **87**, 305-319.
- Matsuhisa, Y., Honma, H. and Matsubaya, O. and Sakai, H. (1972) Oxygen isotopic study on the Cretaceous granitic rocks in Japan. *Contrib. Miner. Petrol.*, **37**, 65-74.
- Matsuhisa, Y., Goldsmith, J. R. and Clayton, R. N. (1979) Oxygen isotopic fractionation in the system quartz-albite-anorthite-water. *Geochim. Cosmochim. Acta*, **43**, 1131-1140.
- Monier, G., Mergoïl-Daniel, J. and Labernardière, H. (1984) Générations successives de muscovites et feldspaths potassiques dans les leucogranite du massif de Millevaches (Massif Central français). *Bull. Minéral.*, **107**, 55-68.
- Nanbu, A., Hamano, K., Tanaka, R. and Ishihara, S. (1997) Chemical and mineralogical characteristics of the Kanamaru pegmatite deposit, Niigata Prefecture, Japan. *Resource Geol.*, **47**, 1-10.
- Nabelek, P. I. and Ternes, K. (1997) Fluid inclusion in the Harney Peak Granite, Black Hills, South Dakota, USA: Implications for solubility and evolution of magmatic volatiles and crystallization of leucogranite magmas. *Geochim. Cosmochim. Acta*, **61**, 1447-1465.
- Ohira, H., Suehiro, A. and Iizumi, S. (2004) Geochronological study on the Masago pluton, Shimane prefecture, Southwest Japan. *Fission Track Newsletter*, 17: 27-31 (in Japanese).
- Roedder, E. (1979) Fluid inclusions as samples of ore fluids. In: Barnes, H. L. (ed.) *Geochemistry of Hydrothermal Ore Deposits*. John Wiley & Sons, New York, 684-737.
- Shibata, K. and Ishihara, S. (1974) K-Ar ages of the major tungsten and molybdenum deposits in Japan. *Econ. Geol.*, **69**, 1207-1214.
- Shipboard Scientific Party (2001) Leg 191 summary. In: Kanazawa, T., Sagar, W. W., Escutia, C. *et al.*, Proc ODP Init Repts, 191, College Station TX (Ocean Drilling Program), 1- 49.
- Smith, P. and Parsons, J. (1974) The alkali feldspar solvus at 1 kilobar water-vapour pressure. *Mineral. Mag.*, **39**, 747-767.
- Soeda, A., Takeno, S. and Watanabe, M. (1979) Mineralogical study on the wolframite series from the Chugoku district, Southwest Japan (I) Relationships between lattice parameters and chemical compositions. *Jour. Japan. Assoc. Mineral. Petrol. Econ. Geol.*, **74**, 357-375.
- Takenouchi, S. (1971) Study of CO<sub>2</sub>-bearing fluid inclusions by means of the freezing stage microscope. *Mining Geol.*, **21**, 286-300 (in Japanese with English abstract).
- Thomas, R., Webster, J. D. and Heinrich, W. (2000) Melt inclusions in pegmatite quartz: complete miscibility between silicate melts and hydrous fluids at low pressure. *Contrib. Mineral. Petrol.*, **139**, 394-401.
- Tilling, R. L. (1968) Zonal distribution of variation in structural state of alkali feldspar within the Rader Creek pluton. *Jour. Petrol.*, **42**, 63-80.
- Trumbull, R. B. (1995) A fluid inclusion study of the Sinceni rare-element pegmatites of Swaziland. *Mineral. Petrol.*, **55**, 85-102.
- Tsuboya, K. and Ishihara, S. (1961) Uraniferous ore deposits at Masago and Komaki mines, Shimane Prefecture. *Rept. Geol. Surv. Japan*, no. 190, 309-322 (in Japanese with English abstract).
- Watanabe, M. and Soeda, A. (1981) Distribution of



polytype contents of molybdenite from Japan and possible controlling factor in polytypism. *Neues Jb. Miner. Abh.*, **141**, 258-279.

Watanabe, M., Shimada, N. and Yoshida, T. (1981) Fluid inclusion study on the granitic rocks and possibly related ore deposits in the Tsumo mining district, southwest Japan. *Mining Geology Special Issue*, No. 9, 145-162 (in Japanese with English

abstract).

Watanabe, M., Nishido, H., Hoshino, K., Hayasaka, Y. and Imoto, N. (1998) Metallogenic epochs in the Inner Zone of Southwestern Japan. *Ore Geol. Rev.*, **12**, 267-288.

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## 島根県益田市の馬の谷-城山鉍山の花崗岩ペグマタイトについて

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### 要 旨

馬の谷-城山鉍山のペグマタイト鉍床(長石および石英)は、本邦最大の生産量を有し、約 97 Ma のチタン鉄鉍系の真砂花崗岩中に胚胎する。鉍体は、母岩である黒雲母花崗岩から内側に向かってその距離に応じて、次のように分帯される：(1) 周縁帯、(2) 漸移帯、(3) 中央帯。肉眼での結晶粒径は、この順番で増大し、その結果、巨大に成長した石英の結晶は中央帯にのみみられる。馬の谷-城山鉍山の鉍石は、単純な鉍物組合せで特徴づけられ、石英とカリ長石が卓越し、微量の白雲母と斜長石を伴う。以下に述べる一連の証拠から、本鉍床が典型的な花崗岩ペグマタイトであり、真砂花崗岩をもたらした花崗岩マグマの末期の結晶・分化作用の産物であると結論される：(1) 真砂花崗岩と鉍床との密接な時間的・空間的随伴関係(約 95～90 Ma)、(2) 鉍体内で、カリ長石と石英との“文象構造”の存在、(3) “鉍石石英”および“鉍石カリ長石”中にメルト・インクルージョンの存在、(4) “鉍石カリ長石”中にパーサイト(その組成から約 500-300 °Cでの離溶を示唆)の存在、(5) “鉍石石英”の酸素同位体組成が、周りの黒雲母花崗岩のそれを受け継いでいること。二種類のメルト・インクルージョン(一つはインクルージョンの全容量の約 80 %以上をいくつかの娘鉍物が占めるもの(±気泡)と、もう一つはその全容量の 30-50 %を占める大きな気泡をもつもの)が共存することは、低圧条件で、シリカに富むメルトと流体との間の混和を示すものかもしれない。石英の酸素同位体組成( $\delta^{18}\text{O}$ )は、関連する火成岩中であれ、“鉍石石英”であれ、ほとんど同じ(約 +12 ‰)である。一方、火成岩中のカリ長石の酸素同位体組成は約 +11 ‰であるのに対して、“鉍石カリ長石”は約 +7-+8 ‰とかなり“同位体的に軽く”なっている。この事実は、石英とカリ長石の間の酸素の交換反応速度の違いによるものと考えられる。多量の気-液 2 相包有物が、“鉍石石英”、“鉍石カリ長石”および関連火成岩中に観察される。この事実は、熱水流体-恐らくは真砂花崗岩マグマから放出されたマグマ流体と循環天水の混合物とみられる-が、主にペグマタイト形成の末期に、本マグマ-熱水系に間隙を通して侵入し、鉍体の中を循環したことを強く示唆する。“鉍石石英”中にトラップされた 2 相包有物の均一化温度(圧力補正はされていない)は、以下の通りである：(1) 250-400 °C (周縁帯)、(2) 230-370 °C (漸移帯)、(3) 240-340 °C (中央帯)。また、末期の熱水流体の循環は、本マグマ-熱水系内部では、石英の  $\delta^{18}\text{O}$  値に影響を与える程には、長期間継続はしなかったと、考えられる。