Bulletin of the Geological Survey of Japan, vol.56 (1/2), p. 1- 8 , 2005

On the oxidized and reduced granites found in quarries of Okayama city, Southwest Japan

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Shunso Ishihara, Shin-ichi Yoshikura, Shigeo Horikawa, Masatsugu Ogasawara, Itaru Nishio and Shigeru Terashima (2005) On the oxidized and reduced granites found in quarries from Okayama city, Southwest Japan. *Bull. Geol. Surv. Japan*, vol. 56 (1/2), 1-8, 6 figs, 2 tables.

Abstract: Two different granites in their oxygen fugacity during the formation, are seen in the drill cores from the northwestern part of Okayama City. One is coarse-grained granite having no or little rock-forming magnetite, while the other is fine-grained aplitic granite occurring in sheet-like form and containing coarse-grained magnetite in miarolitic aggregates of K-feldspar and quartz. The major and trace elements chemistry indicates that the aplitic granite is crystallized from fractionated melts of the coarse-grained granitic magma. It is suggested that the fractionated magma became rich in H_2O which dissociated into O_2 and H_2 and the hydrogen diffused out to the roof rocks; then the oxygen fugacity was increased to form magnetite. Löllingite was discovered along cracks of the coarse-grained granite at one quarry, reflecting a low sulfur fugacity of the post-magmatic hydrothermal activity of these granites.

Keywords: Late Cretaceous, ilmenite series, magnetite series, miarolitic texture, magmatic fractionation, oxidation

1. Introduction

Late Cretaceous to Paleogene granitoids of the Inner Zone of Southwest Japan, forming the largest batholith in the Japanese Islands (Fig. 1), are divided into the inner (Japan Sea side) magnetite series of the Molybdenum Province of the Sanin Belt and the outer ilmenite series of the Tungsten Province of the Sanyo Belt and also outer ilemenite-series of the Barren Province of the Ryoke Belts (Ishihara, 1971). Within the magnetite-bearing granitoids of the Sanin Belt, magnetic susceptibility is inversely correlated with SiO_2 contents.

Coarse-grained biotite (± hornblende) monzogranite is widespread in Okayama City, which is situated in the middle of the Sanyo Belt. The granite is well known building stone for the beautiful cherry color (Ishihara and Sato, 1993). Besides the coarse-grained main rock type mined for building stones, fine-grained aplitic facies was recently discovered by drilling (Cho *et al.*, 2003). This aplitic granite having high SiO_2 contents yield much higher magnetic susceptibility than the main facies granite, which is rather unusual in the granitic terranes of Southwest Japan.

In a given pluton, magnetic susceptibility, i.e., content of magnetite, is higher in mafic phase than the felsic phase, because of availability of iron in the gra-

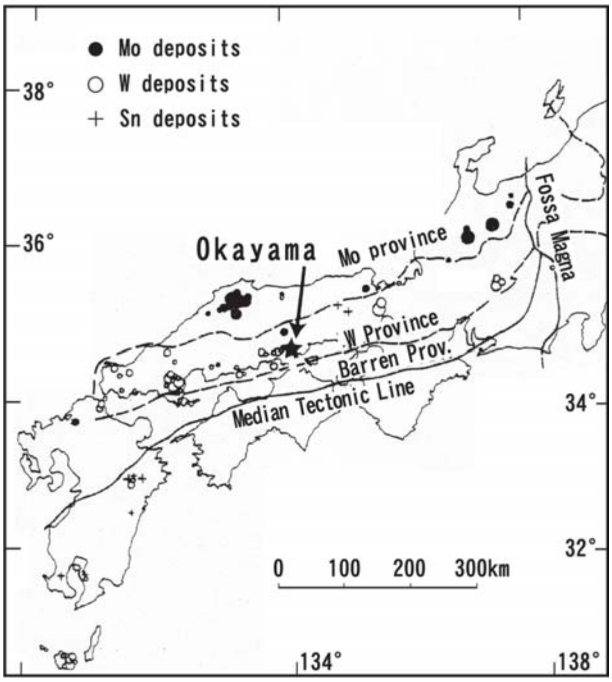

Fig. 1 Mo, W and barren metallogenic provinces of the Inner Zone of Southwest Japan and location of Okayama City.

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nitic magmas. This new observation containing magnetite in leucocratic facies is therefore an interesting subject to study. In this short communication, we report microscopic observation and chemical analyses of the representative granites of the two types and present our interpretation on the oxidation of granitic rocks at high level. A new discovery of löllingite at the quarry will be supplemented.

2. Late Cretaceous granitoids of southern Okayama Prefecture

The southern Okayama region of the eastern Chugoku District, is underlain by Permian sedimentary and meta-igneous rocks of the Maizuru Group, Permian and Jurassic accretionary complex, and the late Cretaceous felsic volcanic rocks, coeval generally with the nearby granitoids. Rb/Sr whole rock ages of the granitoids are determined to be 84.0 ±3.7 Ma with the Sr*I* ratios ranging from 0.7072 to 0.7083 in the Sanyo Belt of the southern Okayama Prefecture (Kagami *et al.*, 1988).

Granitoids of the southern Okayama Prefecture consist essentially of coarse- to medium-grained phases having biotite and/or hornblende-biotite mineral assemblages, and minor fine-grained granitic stocks with varying composition and muscovite-biotite leucogranites occurring at the margin of the coarse-grained biotite granite batholith and related to tungsten ore deposits (Ishihara, 1971; 2002). Amphibole gabbroids occur very sporadically as blocky inclusion bodies in the coarse- to medium-grained granitoids.

Nureki *et al.* (1979) classified the coarse- to medium-grained granitoids into types I to IV, and unclassified fine-grained phases, as follows:

Type I: coarse- to medium-grained biotite syenogranite. K-feldspar is pinkish and has highest triclinicity, thus close to microcline.

Type II: coarse- to medium-grained hornblende-biotite monzogranite. K-feldspar is pink and porphyritic or glomeroporphyritic. This type has an intermediate triclinicity between microcline and orthoclase, and shows no microcline lattice texture under the microscope. The dimension stone mined in the Okayama City belong to this type. By point-counting on 14 stained slabs from three quarries, Nureki *et al.* (1979) revealed the average modal compositions of plagioclase 23.7-31.3 vol. %, K-feldspar 29.1-35.6 vol. %, quartz 28.2-31.2 vol. % and mafic silicates, mostly biotite, 9.6-11.5 vol. %.

Type III: coarse- to medium-grained hornblende-biotite monzogranite similar to the type II, but quartz is rounded and glomeroporphyritic. K-feldspar is pink with low degree of triclinicity.

Type IV: coarse- to medium-grained hornblende-biotite monzogranite. Plagioclase is porphyritic to glomeroporphyritic. K-feldspar is white and glomeroporphyritic.

At north of Kurashiki, 10 km due west of Okayama City, fine-grained muscovite-biotite leucogranite occur at margin of the coarse-grained granites and are related to As-bearing wolframite-quartz veins deposits. This granite is very fractionated (Rb/Sr = 34, Ishihara, 2003) and high in F (1,300 ppm, Ishihara and Terashima, 1977).

3. Quarries of Okayama City

The pink granites are exposed in the hilly granite mountain called the Yasaka-yama of Okayama City. History of the quarrying goes back to the Tenpou Era (1830-1843), but commercial mining began during the Meiji Era (1868-1912), in following privatization of the mountain under the Meiji Government. People are still mining a homogeneous coarse-grained biotite monzogranite with cherry-colored K-feldspar. Commercial name for the darker colored rock is called Ryu-o stone (Fig. 2A), while paler colored one is called Man-nari stone.

Small pegmatitic patches are sporadically seen (Fig. 3A), and mafic enclaves are very rare but present in the quarries with a sharp outline and alkali-feldspar phenocryst (Fig. 3B). Some sulfides may be observed in the quarries in filling cracks without significant hydrothermal alteration. Molybdenite, occurring along cooling joint with a little hydrothermal alteration, has been discovered at two quarries. This molybdenite has $\delta^{34}S$ value of –3.9‰. This negative value is characteristic of the ore minerals related to ilmenite-series granitic magmatism of the Tungsten Province (Ishihara and Sasaki, 2002).

Whitish-yellow mineral with metallic luster grown in columnar shape on cracks in the coarse-grained granite (Fig. 2C) was found at one quarry during the A3 field excursion for the 5th Hutton Symposium in 2003, and was identified by energy-dispersion type XRF to be löllingite having As 72.8% and Fe 27.2 % (Fig. 4). Arsenopyrite is common and löllingite may be found in the high temperature-type ore deposits of the Tungsten Province (e.g., Naegi, Sakamaki *et al.*, 1961; Itaga, Ishihara and Sasaki, 1995). The discovery implies that the post-magmatic activity of the granites was low in sulfur fugacity, which is characteristic of hydrothermal activities related to the ilmenite-series granitoids in the Southwest Japan.

4. Two types of granites in the drill hole

Two types of granites observed in a borehole drilled down to 750 meters are composed of essentially coarse-grained biotite granite. Aplitic granite was locally found as sheet or planar dikes below 500 meters from the surface. The boundary between the two units is sharp at mesoscopic glance but irregular as in details seen in Plate 1B. Pink K-feldspar derived from the host granite may be contained in the aplitic granite. This rock is different from the granite exposed on surface,

Fig. 2 Photographs of rock slabs for the studied granitic rocks, NW Okayama City.
A: Polished slab of the Ryu-o stone (x 1). Fine aggregates of biotite may be well digested mafic enclave.
B: Aplitic granite with an irregular boundary of coarse-grained granite (x1). Note micrographic intergrowth of K-feldspar cored by mafic silicate and quartz. –595 m.
C: Löllingite (white) radiating on crack of the coarse-grained granite without any hydrothermal alteration (x1).

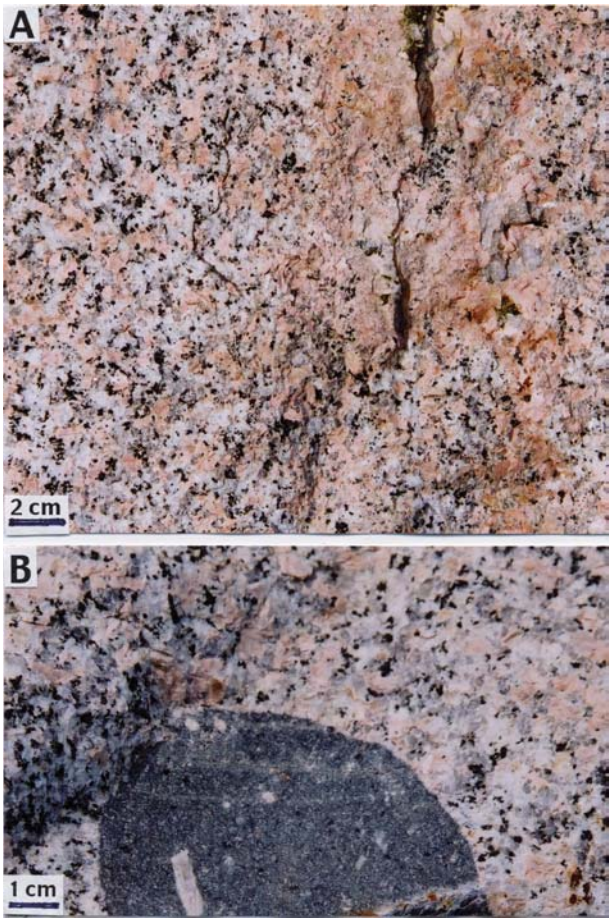

Fig. 3 A: Medium-grained biotite monzogranite with pegmatitic patch along crack. B: The same granite containing mafic enclave with a sharp outline.

because it generally contains magnetite while the exposed rock is magnetite-free, and has not plutonic but micrographic to miarolitic texture (Fig. 2B).

Two pairs of the representative rock types were studied chemically and microscopically, as follows.

Locality 542

301Gb: 541.9-542.0 meters. Coarse-grained biotite granite. The magnetic susceptibility is 0.73 x 10^{-3} SI unit.

301Ga: 542.9-543.0 meters, Fine-grained aplitic granite, sheet-like body with 30^{o} dip, not sharp but irregular boundary with the host biotite granite. The magnetic susceptibility is 9.4 x 10^{-3} SI unit.

Locality 595

302Gb: 599.2-599.3 meters. Coarse-grained biotite granite. The magnetic susceptibility is1.2 x 10^{-3} SI unit.

302Ga: 595.0-595.1 meters. Fine-grained aplitic granite, sheet-like body with 15^{o} dip, has the magnetic susceptibility of 4.5 x 10^{-3} SI unit.

Under the microscope, the coarse-grained rock is monzogranite to granodiorite (plagioclase >K-feldspar) containing abundant biotite (Fig. 5A). Hornblende is of very small amount and partly biotitized. Biotite has Z-axis color of greenish brown. One or two grains of

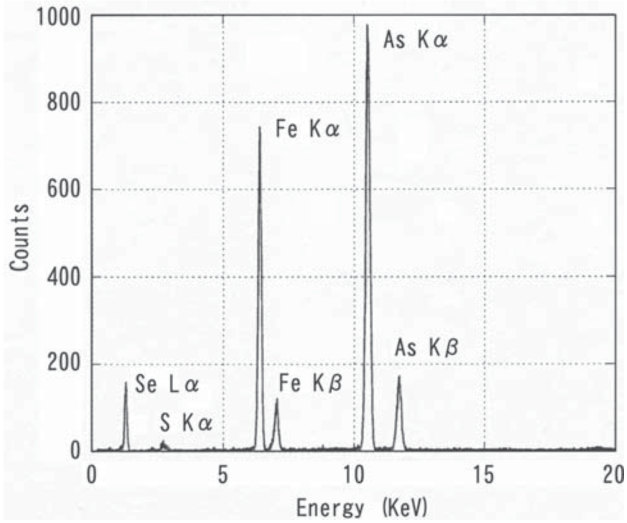

Fig. 4 X-ray spectrum of löllingite obtained with energy dispersive x-ray spectrometer (JEOL JSX-3201).

magnetite inclusion may be seen in one polished thin section of this rock (Fig. 5B), while ilmenite grains are ten to twenty times more in the grain number. One grain of allanite is found. Plagioclase is moderately sericitized (Fig. 5A), and K-feldspar is also dusty due to fine specks of altered minerals.

The aplitic granite contains much more K-feldspar than plagioclase (Fig. 5C); thus syenogranitic. K-feldspar is dusty due to alteration products. Several large grains of magnetite are found in one polished thin section. The magnetite has irregular anhedral outlines and contains no hematite blade (Fig. 5D). Biotite is small in amount and allanite is seen.

Major elements of these granites are shown in Table 1. The aplitic granites are higher in SiO_2, Na_2O (and K_2O), but are lower in TiO_2, Fe_2O_3, MgO, MnO, CaO and P_2O_5. Thus, the aplitic granites are enriched in later crystallizing components, which are also shown by the following trace element components.

Among trace elements, volatile components such as F and Li are decreased in the aplitic granite. The other decreased elements are those substituting feldspar components (Cs, Sr, Ba, not Pb). Rb is not much different between the two granites, but Rb/Sr ratio increases. Zn decreases in following decreasing of ferromagnesian components. Among those contained in accessory minerals, Zr decreases but Hf increases, so that the Zr/Hf ratio decreases in the aplitic granite. Both Th and U increase; light rare earth elements (LREE) but Y decreases in the aplitic granite.

Chondorite normalized REE patterns of the coarse-grained granite show enrichment of LREE with flat and low values on HREE (Fig. 6). The aplitic granites have flat patterns or HREE may even be higher than LREE. The negative Eu anomalies are very strong, which may be caused by plagioclase fractionation, as compared with

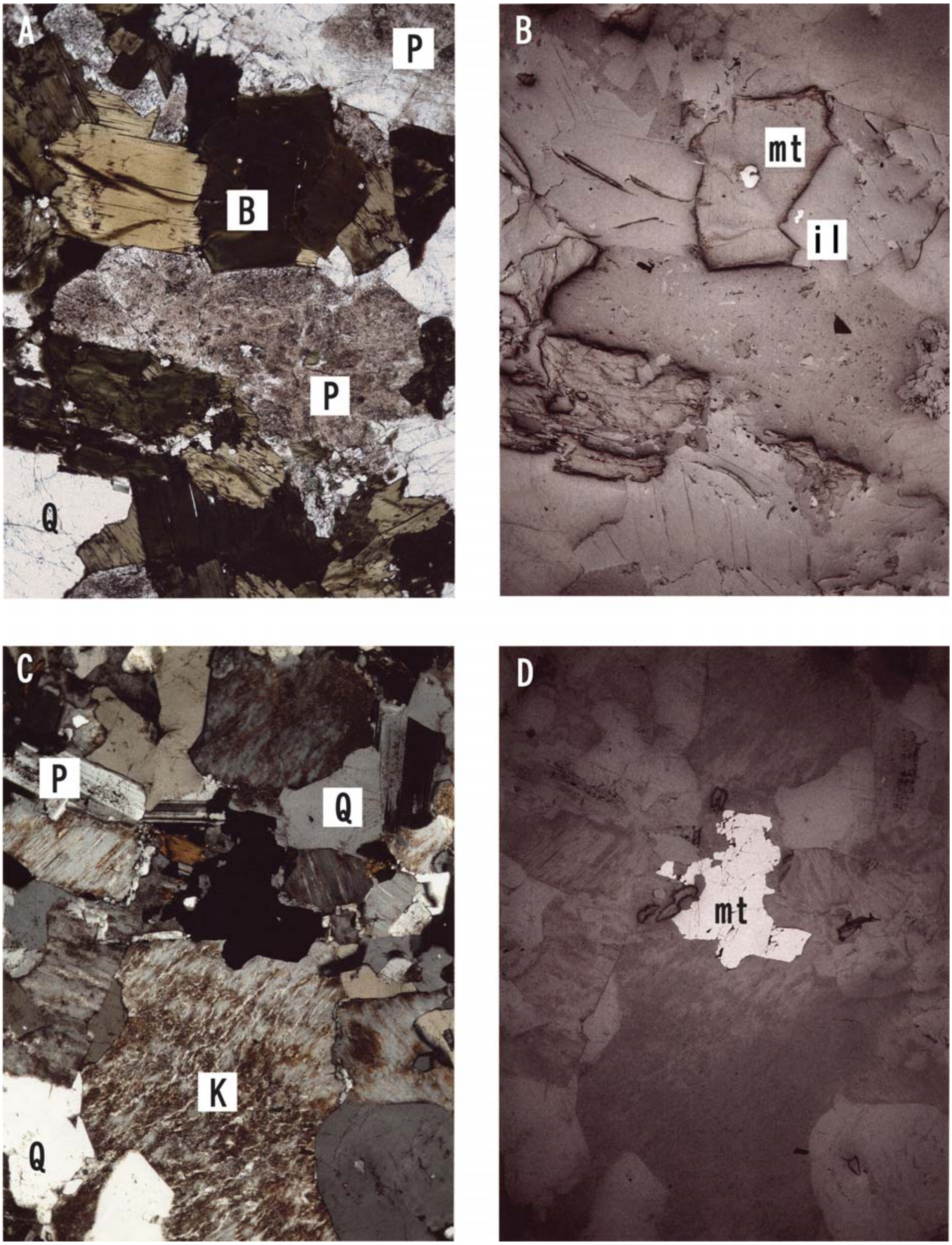

Fig. 5 Microscopic observation of the studied granitic rocks

A (transmittance) and B (reflective light): Coarse-grained granite, 301Gb (x 3.2). Plagioclase (P) is sericitized but biotite (B) is relatively fresh. Two grains of magnetite (mt) and 20 grains of ilmenite (il) are seen in this thin section. Q : quartz

C (transmittance) and D (reflective light): aplitic granite, 302Ga (x3.2).

Note abundance of K-feldspar (K) and much less plagioclase (P), compared with the coarse-grained rock and no hematitization on the magnetite. Q, quartz.

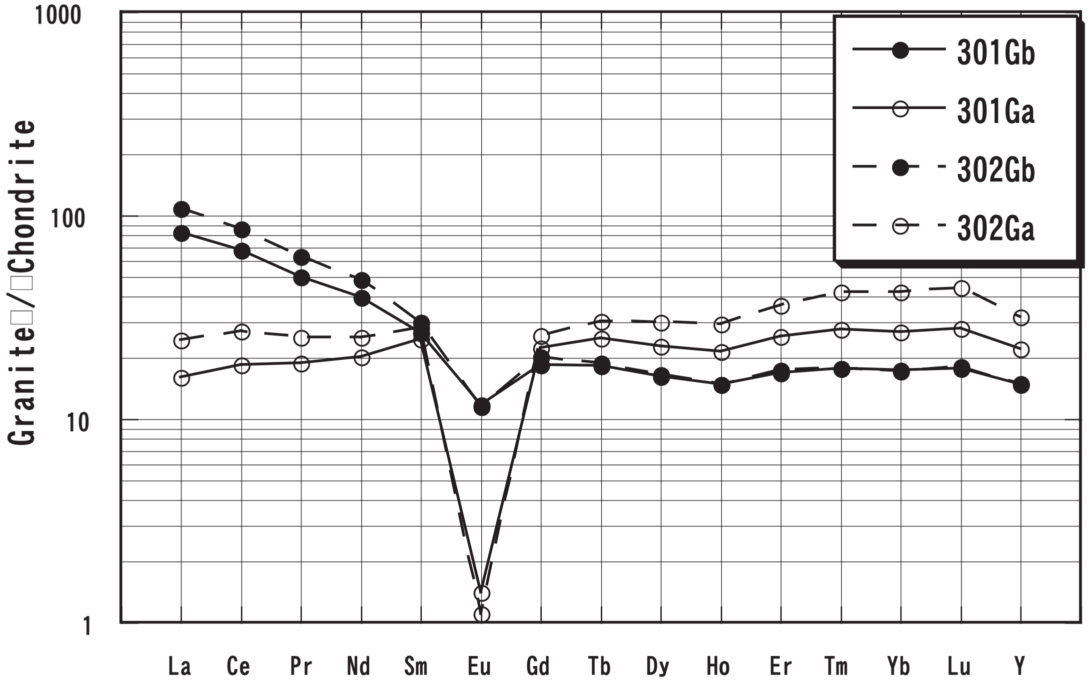

Fig. 6 REE patterns of the studied granites.

weak negative anomalies on the coarse-grained granite.

The aplitic granites are often miarolitic, micrographic (Fig. 2B), and sometimes pegmatitic up to a few cm in the crystal length. In a miarole, pink colored K-feldspar and quartz are main components, which may be cored by mafic silicates and magnetite. The matrix is fine-grained aggregates of sodic plagioclase, K-feldspar and quartz. Magnetite is not polygonal-euhedral crystals but crystals with irregular outline (Fig. 5D). This observation indicates that the magnetite was crystallized at the latest stage in the solidification of the aplitic granitic magma.

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5. Genetic consideration

Compared with the coarse-grained granite, the aplitic granite contains less plagioclase and mafic silicates, implying that the aplitic granite represents a fractionated melt of the coarse-grained granite magma and intruded as gently dipping sheet in the coarse-grained granite. Such fractionated facies are often observed at margin (e.g., very fine-grained Nukame facies of the Naegi Granite, Ishihara, 1971) or aplitic top of the Okueyama granitic pluton.

Boundary between the two facies is not sharp (Fig. 2B) and xenocrystic K-feldspar from the host granite indicates a small time gap and weak fracturing between the solidifications of the coarse-grained granite and aplitic granite magmas.

In their chemistry (Tables 1, 2), F and Li are distributed more in the early coarse-grained facies, implying that these elements have smaller distribution coefficient to the residual melts. Strong decreasing of Ba, Sr, Cs, Cr, Zn, Zr, La and Ce in the aplitic granite indicate the same tendency. On the other hand, such heavy elements as Y, Th, U, Nb, and Hf are distributed preferentially in the residual melts.

In the southern Okayama Prefecture, magnetic susceptibility measurement indicates that some granitoids have boundary values of the magnetite and ilmenite series, although the region belongs to the ilmenite-series (see Fig. 1 of Ishihara *et al.*, 2003). Such example is also found in the coarse-grained granite, and two grains of magnetite specks are found in the drill core sample of 301Gb (Fig. 5B). Large phenocrystic magnetites (Fig. 5D), however, are seen with miarolitic feldspar and quartz in the aplitic granite. This observation indicates that the magnetite is the latest crystallized mineral.

Magnetite in granitoids occurs usually as euhedral crystals contained in mafic silicates, which is a real evidence of magnetite series (Tsusue and Ishihara, 1974). In the aplitic granite, the magnetite occurs as anhedral, phenocrystic form associated with miarolitic salic minerals. The magnetite must have been crystallized at the end of the granite solidification having increasing of oxygen fugacity.

Two possible mechanisms can be considered to increase the oxygen fugacity in the solidifying granitic magmas: (1) concentration of H_2O in the residual melts, then dissociation of H_2O and quick defusing out of H_2 into the roof rocks, and (2) involvement of small amounts of meteoric water which supplied free oxygen. Although we have no O- and H-isotopic data yet, meteoric water interaction has not been observed in normal batholithic environment (Ishihara and Matsuhisa, 2002). We tentatively conclude the first interpretation may be the case for the studied granites.

Table 1 Chemical compositions of the representative coarse-grained granite and fine-grained aplitic granite.

	542 Locality		595 Locality	
	301Gb	301Ga	302Gb	302Ga
SiO_2(%)	73.84	74.46	74.98	76.51
TiO_2	0.17	0.04	0.17	0.02
Al_2O_3	12.98	12.79	12.66	12.57
Fe_2O_3	2.19	1.26	2.34	0.96
MnO	0.05	0.02	0.06	0.02
MgO	0.19	0.03	0.19	<0.02
CaO	1.24	0.6	1.45	0.45
Na_2O	3.65	4.32	3.87	4.02
K_2O	4.44	4.33	3.51	4.81
P_2O_5	0.05	<0.01	0.05	0.02
S	0.01	0.01	<0.01	0.01
CO_2	0.15	0.12	0.10	0.06
H_2O^+	0.62	0.34	0.56	0.34
H_2O^-	0.40	1.87	0.06	0.13
Total	99.98	100.19	100.00	99.92
Trace elements (ppm)				
F	1070	700	1240	1000
Li	31	13	41	19
Rb	168	156	134	227
Cs	7.1	4	10.9	3.8
Sr	107	11	108	9
Ba	722	12	539	12
Zr	134	48	145	110
Hf	5.4	6.5	5.5	10
Nb	9.5	9.9	10.5	17
Ta	4	4	3	5
Y	31	50	33	67
La	24	5	37	8
Ce	52	17	67	23
Cr	23	14	20	12
Co	6	8	7	8
Zn	59	28	61	24
Pb	20	32	17	35
Ga	16.1	19.0	16.4	17.4
Ge	1.7	2.0	1.3	1.9
Se	0.3	0.5	0.3	0.2
Mo	0.8	0.6	0.6	0.6
W	2	3	2	3
Sn	3.3	2.8	3.7	2.1
Sb	0.3	0.5	0.9	0.5
Tl	1.8	2.1	1.5	2.2
Bi	0.3	<0.3	0.9	1.9
Th	9.5	19.4	11.1	30
U	2.5	7.5	2.1	10.7
ASI	0.99	0.99	0.99	0.99
K_2O/Na_2O	1.2	1	0.91	1.2
Rb/Sr	1.6	14.2	1.2	25.2

The following elements are below the detection limits:
V 1 ppm, Ni 1ppm, Cu 1 ppm, As 1ppm and Cd 0.2 ppm.
Analyzed by polorized XRF at GEMOC, Macquarie Univ.
F by photometric (Terashima, 1977) and Li by AA (Terashima, 1971)

Table 2 REE and some other element abundance of the coarse-grained granite and fine-grained aplitic granite.

	542 Locality		595 Locality	
Elements	301Gb	301Ga	302Gb	302Ga
Rb	170	155	136	230
Sr	99	8	101	7
Ba	711	14	528	12
Cs	6.8	4.3	6.1	3.7
Ga	16	18	16	18
Ge	1.5	2.5	1.9	2.5
Ag	1.6	0.9	1.6	1.3
La	25.6	5.00	33.8	7.61
Ce	54.4	15.0	70.0	21.9
Pr	6.10	2.30	7.70	3.09
Nd	23.7	12.2	29.1	15.2
Sm	5.19	4.84	5.87	5.49
Eu	0.866	0.103	0.850	0.082
LREE	115.856	39.443	147.320	53.372
Gd	4.80	5.79	5.26	6.71
Tb	0.87	1.19	0.90	1.43
Dy	5.22	7.36	5.33	9.65
Ho	1.06	1.55	1.07	2.11
Er	3.53	5.36	3.64	7.63
Tm	0.576	0.896	0.576	1.36
Yb	3.64	5.60	3.61	8.79
Lu	0.572	0.903	0.585	1.44
Y	32.5	48.8	32.9	69.7
HREE	52.768	77.449	53.871	108.82
Zr	171	54	181	132
Hf	4.8	3.3	4.9	6.5
Nb	12.5	16.8	12.5	20.5
Ta	0.9	1.7	0.8	2.6
Tl	1.2	1.5	1.2	1.7
Pb	9	24	13	25
Th	10.4	20.2	13.7	36.0
U	2.8	6.5	3.0	11.8
Rb/Sr	1.7	19.4	1.4	32.9
La/Yb	7.0	9.4	0.9	0.9
Zr/Hf	35.6	16.4	36.9	20.3
Th/U	3.7	3.1	4.6	3.1
L/HREE	2.2	0.51	2.7	0.49

Analyzed by ICP-MS at Actlabs.

6. Conclusions

Magnetite from the magnetite-bearing aplitic granite discovered in the drill cores of the northwestern part of Okayama City is considered as the latest crystallized mineral from the residual melts of the late Cretaceous granitic activities, for the mode of occurrence in the miarolitic part and anhedral morphology. The residual melts were rich in H_2O, and dissociation of H_2O and preferential diffusion of hydrogen brought the local oxidation of the aplitic granite magma.

Acknowledgements: The authors wish to acknowledge Dr. Akio CHO for the samples provided, and Dr. Jun-ichi Itoh for energy dispersive XRF analysis.

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Received May 26, 2004
Accepted February 24, 2005

西南日本，岡山市の採石場に見られる還元型／酸化型花崗岩類

石原舜三・吉倉紳一・堀川滋雄・小笠原正継・西尾　格・寺島　滋

要　旨

岡山市内には多数の採石場があり,チタン鉄鉱系のピンク花崗岩を古くから採石している．この度,ボーリング岩芯から磁鉄鉱系に属する花崗岩の共存例が発見されたので,それらの特徴を記載し成因を考察した．これは地下500～700 mで見られ，粗粒チタン鉄鉱系花崗岩中に磁鉄鉱系アプライト質花崗岩が混在するものである．このアプライト質花崗岩は岩床状の産状を示し,ミアロリチック組織や小規模ペグマタイトを伴う．磁鉄鉱は粗粒であるが,他形結晶でミアロル部分に産出する．アプライト質花崗岩の主成分・微量成分は固結末期濃集成分に著しく富んでおり，粗粒花崗岩マグマの分化相と考えられる．粗粒チタン鉄鉱系花崗岩マグマの固結最末期に水に富む少量の残マグマが形成され，その水のO_2，H_2への解離とH_2の上方への選択的な拡散によって，酸素フュガシティが上昇し，磁鉄鉱系アプライト質花崗岩が晶出したものと解釈された．また近傍の採石場で粗粒花崗岩の割れ目から見いだされた黄白色の柱状結晶は，化学分析によって砒鉄鉱と同定された．この発見は花崗岩マグマ最末期の熱水活動の硫黄フュガシティが低かったことを示唆する．