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HYDROTHERMAL EXPERIMENTS AND PETROGENESIS OF THE TAKAKUMAYAMA GRANITE, KAGOSHIMA PREFECTURE, JAPAN

By

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Abstract

Hydrothermal experiments on the Takakumayama granite, Kagoshima Prefecture, Japan, composed of a granodiorite of the Shinkoji-type and an aplitie granite of the Sarugajo-type, were carried out in the presence of 10 weight per cent water at temperatures up to 750°C. and 1 kbar. With increasing temperature, quartz disappeared in the lower temperature range than potassium feldspar, and biotite in the aplitic granite disappeared in the lowest temperature, 680°±10°C. The solidus temperatures of both the granodiorite and the aplitic granite are 720°±10°C., and agree approximately with the minimum melting point in the 'Granite System' experimentally determined by TUTTLE and BOWEN (1958). Petrogenesis of the Takakumayama granite was considered in the present paper.

Introduction

A number of experimental studies have been reported on the 'Granite System' (*e g.*, TUTTLE and BOWEN, 1958; LUTH *et al.*, 1964), the 'Granodiorite System' (*e.g.*, VON PLATEN, 1965; WHITNEY, 1975) and natural rock-water systems (*e.g.*, PIWINSKII and WYLLIE, 1968, 1970; ROBERTSON and WYLLIE, 1971a, b). These studies have provided many useful petrogenetical informations and indicated the importance of experimental works on granitic rocks from individual intrusive body in the presence of water at lower pressures.

The Takakumayama granitic stock (KAWACHI, 1961; ISHIHARA and KAWACHI, 1961), one of intrusive bodies in the western district of the Shimanto terrain, is located in the central part of the Ōsumi Peninsula, Kagoshima Prefecture, Japan (Fig. 1). Granitic rocks in the district are compositionally characterized by the higher K_2O/Na_2O and FeO/CaO ratios, and have common association with volcanic rocks (ŌBA, 1967). Few experimental studies, however, have been reported on granitic rocks having such properties. Therefore, hydrothermal experiments on the Takakumayama granite were carried out in the presence of about 10 weight per cent water at temperatures ranging from 670°C. to 750°C. and 1 kbar, in order to obtain genetical informations near solidus temperature.

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A number of geological and petrological studies of the granite have been reported (ŌBA, 1958, 1963; SHIBATA *et al.*, 1960, 1966; ISHIHARA and KAWACHI, 1961; KAWACHI, 1961, 1969; OTA and KAWACHI, 1965; TSUSUE, 1973; YAMAMOTO, 1975), and the crystallization of granitic glasses made by fusion of the granite has been studied by YAMAMOTO (1976).

The Takakumayama Granite

The Takakumayama granitic stock (Fig. 1) intrudes the Takakumayama Formation (OTA and KAWACHI, 1965) of Late Mesozoic geosynclinal sedimentary complex regionally metamorphosed to the greenschist facies. A roof pendant of the Formation still remains on the top of the Nanatsu-dake. The granite is lithologically divided into two types of Shinkoji and Sarugajo (ISHIHARA and KAWACHI, 1961; KAWACHI, 1961). Rock of the Shinkoji-type is a granodiorite which forms the core of the stock, and that of the Sarugajo-type is an aplitic granite which forms the roof. The

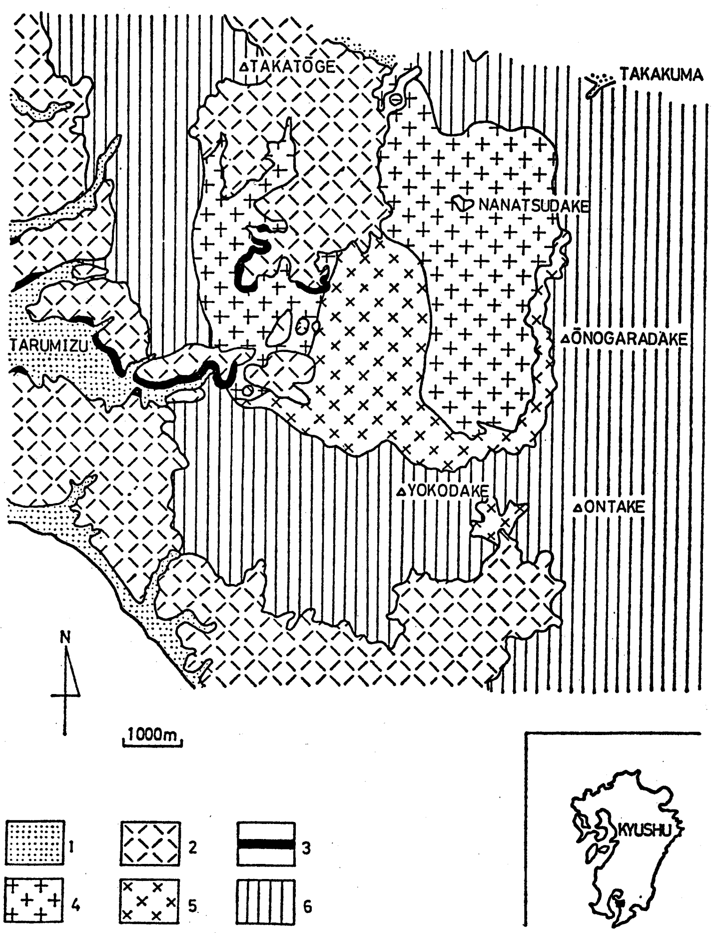

Fig. 1. Index and geologic maps of the Takakumayama granitic stock and surrounding region. Stratigraphic sequence: 1, Alluvial deposits; 2, Pyroclastic flow deposits; 3, Onobaru sandstone and conglomerate and Tarumizu sand and gravel bed; 4 and 5, Takakumayama granite (4, Shinkoji-type; 5, Sarugajo-type); 6, Takakumayama Formation.

contact between the two types is gradational across a zone several hundred meters wide. The K-Ar age determination on biotite (MILLER *et al.*, 1962) indicates that the granite was emplaced during Late Miocene (16 m.y.). Boundaries of the stock for the Formation show sharp and discordant contacts which dip to outward from the stock. Xenoliths are found throughout the stock. Mineral assemblages in the highest thermally metamorphosed pelitic rocks are characterized by appearance of garnet, andalusite or potassium feldspar (OTA and KAWACHI, 1965).

The granodiorite is a light gray-colored and coarse- to medium-grained rock with granular texture. Constituent minerals comprise plagioclase (An_{35}), quartz, potassium feldspar ($Or_{76.5}$), and biotite, with zircon, apatite, ilmenite and tourmaline as common accessories. The aplitic granite is a light-colored and medium- to fine-grained rock with slightly porphyritic texture. Constituent minerals comprise quartz, potassium feldspar ($Or_{80.5}$), plagioclase (An_{17}), biotite and small amounts of garnet and muscovite, with zircon, apatite, ilmenite and tourmaline as common accessories.

A number of compositions of the Takakumayama granite have been reported in modal by ŌBA (1958), OTA and KAWACHI (1965), TSUSUE (1973) and YAMAMOTO (1975) and in chemical by SHIBATA *et al.* (1960), KAWACHI (1961), ŌBA (1963), TSUSUE (1973) and YAMAMOTO (1975). In modal analyses, amounts of plagioclase and biotite

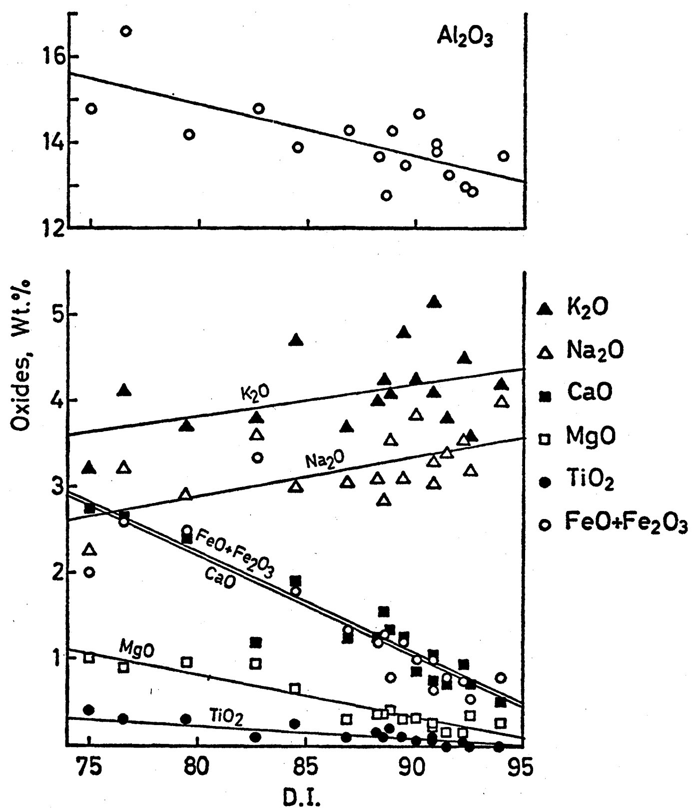

Fig. 2. Oxide contents against differentiation index (D.I.) of the Takakumayama granite. Compositions of the granite previously reported by SHIBATA *et al.* (1960), KAWACHI (1961), ŌBA (1963), TSUSUE (1973) and YAMAMOTO (1975) are included.

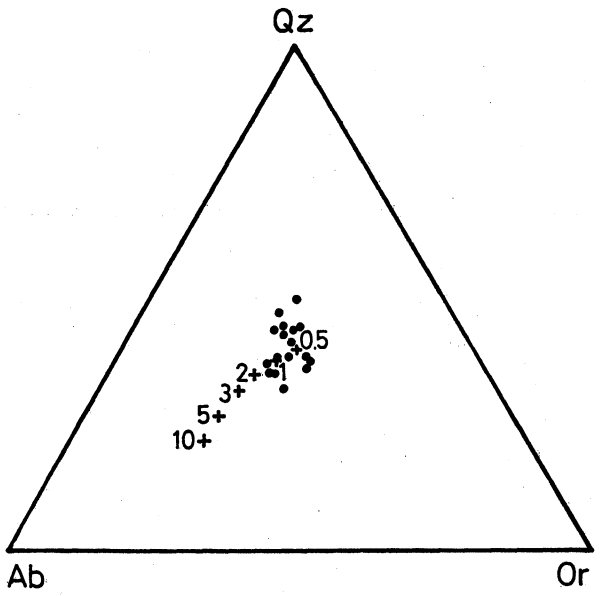

Fig. 3. Composition of the Takakuymayama granite plotted in the normative Q-Ab-Or system. Crosses represent the minimum melting compositions at indicated pressures in kbar in the $KAlSi_3O_8$-$NaAlSi_3O_8$-SiO_2-H_2O system experimentally studied by TUTTLE and BOWEN (1958) and LUTH *et al.* (1964).

decrease from the granodiorite into the aplitic granite, whereas those of potassium feldspar and garnet increase. Compositional variation of the Takakumayama granite is also found in all chemical components plotted against differentiation index (THORNTON and TUTTLE, 1960) (Fig. 2). The index varies from 75 in the granodiorite through 88 in granite of the gradational zone to 94 in the aplitic granite. Contents of TiO_2, Al_2O_3, $FeO+Fe_2O_3$, MgO, CaO and H_2O^+ decrease with increasing differentiation index, whereas those of SiO_2, Na_2O and K_2O increase. K_2O content exceeds Na_2O in all analyzed samples, and total FeO and Fe_2O_3 content agrees closely with CaO in almost all of the samples.

Normative C was calculated in the samples. Figure 3 shows normative composition of the Takakumayama granite plotted in the Q-Ab-Or system. Crosses represent the minimum melting compositions at indicated pressures in kbar in the $KAlSi_3O_8$-$NaAlSi_3O_8$-SiO_2-H_2O system experimentally determined by TUTTLE and BOWEN (1958) and LUTH *et al.* (1964). The granite is plotted within a small area around the minimum melting compositions at pressures below 1 kbar.

Experimental Method and Results

1. Experimental Method

Two powdered rock samples, the granodiorite No. TK01 of the Shinkoji-type and the aplitic granite No. TK09 of the Sarugajo-type, were used for these experiments. Modal and chemical analyses and CIPW norms of the samples are presented in Table 1. The experiments were all performed in standard 'cold seal' pressure vessels (TUTTLE, 1949). The fluid pressure, always equals to the total pressure in runs, was maintained within ±20 bars at 1 kbar. The temperature was measured with chromel-alumel thermocouples, and was regulated to a precision of ±5°C.

The charge of the starting material plus water put into an Ag-Pd capsule and was sealed by electric or gas torch welding. In order that equilibrium is attained, run duration at each temperature was not less than that in the experimental studies by PIWINSKII and WYLLIE (1968, 1970).

Table 1. Modal and chemical analyses and CIPW norms of the Takakumayama granite used for hydrothermal experiments

No.	TK01	TK09
Modal analyses		
Quartz	24.6	34.4
Potassium feldspar	27.6	34.2
Plagioclase	40.1	28.6
Biotite	6.7	1.2
Garnet	-	1.3
Muscovite	-	tr.
Tourmaline	tr.	tr.
Chlorite	0.7	tr.
Opaque minerals	0.2	0.2
Chemical analyses		
SiO_2	71.20	75.82
TiO_2	0.30	0.09
Al_2O_3	14.22	13.94
Fe_2O_3	1.01	0.19
FeO	1.47	0.48
MnO	0.07	tr.
MgO	0.97	0.25
CaO	2.40	0.74
Na_2O	2.88	3.30
K_2O	3.70	4.10
H_2O^+	1.14	0.32
H_2O^-	0.26	0.24
P_2O_5	0.08	0.03
Total	99.70	99.50

	TK01	TK09
CIPW norms		
Q	33.2	38.8
Or	21.9	27.9
Ab	24.4	27.9
An	11.4	3.5
C	1.3	2.8
En	2.4	0.6
Fs	1.5	0.6
Mt	1.5	0.3
Il	0.6	0.2
Ap	0.2	0.1

TK01: Biotite granodiorite of the Shinkoji-type.

TK09: Garnet-bearing biotite aplitic granite of the Sarugajo-type.

Quenching was all achieved by first removing pressure vessel from furnace, and then the vessel was cooled down to room temperature. Run products were removed from charge containers, dried at 60°C. in air, and subsequently examined by means of a petrographic microscope and an X-ray powder diffractometer employing CuKα radiation ($\lambda=1.5418$ Å).

2. Experimental Results

Results of hydrothermal experiments on the Takakumayama granite are listed in Table 2.

In the granodiorite TK01 of the Shinkoji-type, the same mineral assemblage as the natural rock remained in the run for 18 days at 730°C. Glass was not produced in the run for 28 days at 710°C., but for 18 days at 730°C. Quartz disappeared in the run for 25 days at 750°C. Small amounts of hematite and magnetite were formed in runs at temperatures above 690°C.

In the aplitic granite TK09 of the Sarugajo-type, the same mineral assemblage as the natural rock remained in the run for 32 days at 670°C., although an amount of biotite decreased considerably and small amounts of hematite and magnetite were

Table 2. Results of hydrothermal experiments on the Takakumayama granite at 1 kbar

No.	Temp., °C	Time, days	Crystalline Phases
TK01	670	32	Pl+Or+Qz+Bt
	690	24	Pl+Or+Qz+Bt+Mt+Hm
	710	28	Pl+Or+Qz+Bt+Mt+Hm
	730	18	Pl+Or+Qz+Bt+Mt+Hm+Gl
	750	25	Pl+Or+Bt+Mt+Hm+Gl
TK09	670	32	Pl+Or+Qz+Bt+Ga+Mt+Hm
	690	24	Pl+Or+Qz+Ga+Mt+Hm
	710	28	Pl+Or+Qz+Ga+Mt+Hm
	730	18	Pl+Or+Qz+Ga+Mt+Hm+Gl
	750	25	Pl+Or+Ga+Mt+Hm+Gl

Abbreviations: Pl, plaigoclase; Or, potassium feldspar; Qz, quartz; Bt, biotite; Ga, garnet; Mt, magnetite; Hm, hematite; Gl, glass.

formed. The biotite disappeared in the run for 24 days at 690°C. As in the case of the granodiorite, glass was not produced in the run for 28 days at 710°C., but for 18 days at 730°C., and quartz disappeared in the run for 25 days at 750°C.

Hematite is a light brown-colored and hexagonal-shaped grain with several microns in size under the petrographic microscope. Magnetite forms cube-shaped grain with the same grain size as the hematite. Amounts of hematite and magnetite will be related to partial or complete decomposition of biotite. It is estimated that these hydrothermal experiments were all performed in oxidizing conditions near f_{O_2} defined by the HM buffer experimentally determined by EUGSTER and WONES (1962).

The experimental results indicate that with increasing temperature, quartz disappears in the lower temperature range than potassium feldspar, and biotite in the aplitic granite disappears at the lowest temperature. They also indicate that no difference between the solidus temperatures of the granodiorite and the aplitic granite is found in the presence of excess water at 1 kbar.

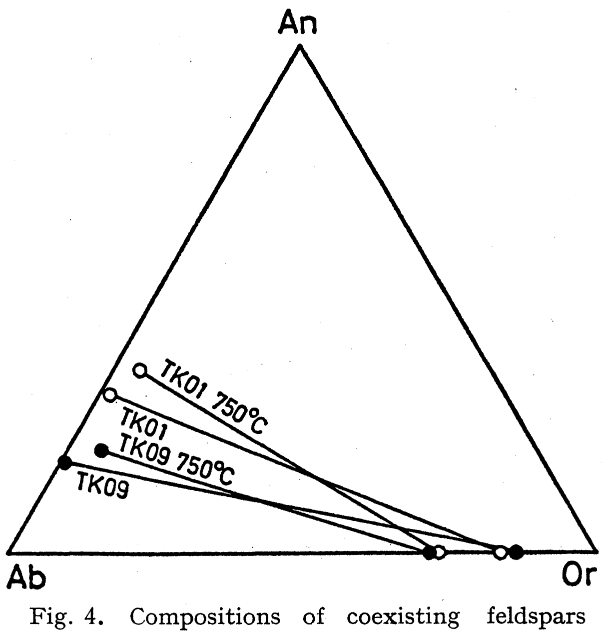

Fig. 4. Compositions of coexisting feldspars from natural granitic rocks and their run products at 750°C. and 1 kbar.

3. Reactions of Coexisting Feldspars

Figure 4 shows compositions of coexisting feldspars in the natural rock samples and their run products at 750°C. Composition of potassium feldspars was estimated from 2θ values of ($\bar{2}$01) reflection by using the determinative curve of the alkali-exchanged orthoclase by WRIGHT and STEWART (1968) and WRIGHT (1968), although An-content could not be obtained. Composition of plagioclases was also estimated from refractive indices and from 2θ values ($\bar{2}$01) reflection by using the determinative curve of the 'maximum microcline-low albite' solid solution by ORVILLE (1967). As clearly seen in Fig. 4, potassium feldspars from the run products are richer in Ab-content than the natural ones, and plagioclases also in An-content. It is suggested, therefore, that the Or-component in potassium feldspars will be the most reactive as compared to the other two components, and similarly the Ab-component in plagioclase.

Genetical Considerations of the Takakumayama Granite

1. Water in the Takakumayama Granitic Magma

In the present study, the temperature for disappearance of biotite in the aplitic granite of the Sarugajo-type is 680°±10°C., and is apparently lower than that of quartz, 740°C±10°C. This is consistent with the crystallization of the granitic glass made by fusion of the aplitic granite studied by YAMAMOTO (1976) that quartz and no biotite is synthesized in the presence of excess water with the HM buffer at 700°C. and 1 kbar. However, occurrence of the aplitic granite shows that biotite is rarely included into quartz grain. According to the experimental studies in the water-deficient system of a granodiorite reported by ROBERTSON and WYLLIE (1971a, b), with decreasing water content the increase of isobaric temperature for disappearance of biotite is considerably greater than that of quartz. On the other hand, the crystallization of the granitic glass also indicates that both biotite and quartz are stable in the run with the NNO buffer at the same temperature and pressure. These facts imply that the aplitic granite has been formed either in a relatively poor water condition or in an oxidizing one. However, YAMAMOTO (in prepared) has shown that the biotite in the aplitic granite has crystallized in an oxidizing condition. It is estimated, therefore, that the aplitic granite has been probably formed in an environment near a water-saturated condition. Judging from the modal and chemical analyses that the granodiorite of the Shinkoji-type is rich in hydrous minerals and H_2O^+ as compared to the aplitic granite, it is also estimated that the granodiorite has been formed in an environment like the formation of the aplitic granite.

2. Differentiation of the Takakumayama Granitic Magma

In the present study, the temperature for disappearance of quartz was 740°±10°C. in both the granodiorite and the aplitic granite, and the quartz disappeared in lower temperature than potassium feldspar. Crystalline phases in the runs at 750°C. correspond to the mineral assemblage of the two-feldspar surface in the 'Granodiorite Tetrahedron.' As mentioned above, it is estimated that the Takakumayama granite

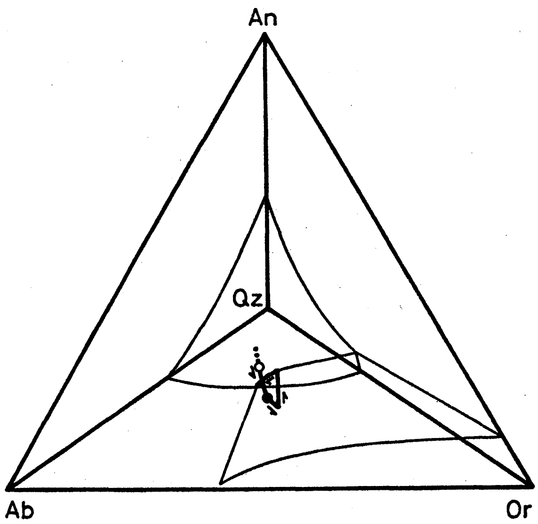

Fig. 5. Schematic diagram to illustrate a crystallization course of the Takakumayama granitic magma in the 'Granodiorite Tetrahedron.' Open and solid circles represent compositions of the granodiorite of the shinkoji-type and the aplitic granite of the Sarugajo-type, respectively. Curves with arrows represent the crystallization course.

has been formed in environments near a water-saturated condition. It is suggested, therefore, that liquid in the Takakumayama granitic magma has changed compositionally from the two-feldspar surface through the two-feldspar-quartz boundary curve toward the minimum melting point during the differentiation (Fig. 5).

The following two contrast crystallization courses of granitic rocks have been recognized: (1) Potassium feldspar crystallizes after appearance of quartz (*e.g.*, PIWINSKII and WYLLIE, 1968, 1970; PIWINSKII, 1968); and (2) Quartz crystallizes after appearance of potassium feldspar (*e.g.*, GIBBON and WYLLIE, 1969; MCDOWELL and WYLLIE, 1971, ROBERTSON and WYLLIE, 1971a, b). The former includes mainly granitic rocks constituting batholith ranging in composition from tonalite to granite, and sometimes has normative Di. On the other hand, in the latter, acidic volcanics, syenitic rocks and alkali granites are mainly included and normative C is calculated from almost all of their chemical analyses. This fractional crystallization is explained by a course, syenodiorite→granodiorite→granite, according to JOHANSSEN's (1939) classification.

Therefore, the fact that quartz disappears in lower temperature than potassium feldspar is consistent with the following features of the Takakumayama granite and the related granitic rocks:

(1) Normative C is nearly always calculated from chemical analyses of the Takakumayama granite and the related granitic rocks.

(2) The aplitic granite is a slightly porphyritic rocks, and the related granitic rocks are sometimes accompanied with volcanics such as porphyries and rhyolites (*e.g.*, Satsuma Peninsula granitic rocks, YAMAMOTO *et al.*, 1970).

(3) Potassium feldspar grain in the Takakumayama granite is well crystallized rather than quartz, and euhedral one more than several centimeters in size occurs sometimes within the related granitic rocks (*e.g.*, Yakujima granite, SHIBATA *et al.*, 1960).

3. P-T Condition of the Takakumayama Granitic Magma

Evidences by which pressure at emplacement of the Takakumayama granite is estimated are as follows: (1) Occurrence of a roof pendant of the Takakumayama

Formation on the top of the Nanatsu-dake and a slightly porphyritic texture of the aplitic granite indicate that the Takakumayama granite has been emplaced at a relatively shallow depth; and (2) Normative composition in Fig. 3 shows that the granite is plotted around the minimum melting compositions at pressures below 1 kbar in the 'Granite System' experimentally determined by TUTTLE and BOWEN (1958). As mentioned previously, it is estimated that the granite has been formed in environments near a watersaturated condition. Therefore, the pressure of the Takakumayama granitic magma is considered to be approximated by that of the minimum melting composition in the 'Granite System', *i.e.*, a low pressure condition below 1 kbar.

Evidences by which temperature at emplacement of the Takakumayama granite is estimated are as follows: (1) Mineral assemblages of the aplitic granite and the highest metamorphosed pelitic rocks in the contact aureoles (OTA and KAWACHI, 1965) indicate that the Takakumayama granite has been formed at temperature corresponding to the amphibolite facies; and (2) Crystallization of granitic glasses in the presence of excess water at 700°C. and 1 kbar (YAMAMOTO, 1976) indicates that the same major constituent mineral assemblages as the host rocks are obtained, without the HM buffer. In the present study, glass was not produced in runs at 710°C., but at 730°C., and the solidus temperature at 1 kbar was 720°C$\pm$10°C. in both the granodiorite and the aplitic granite. As mentioned above, it is estimated that the Takakumayama granite has been formed at pressures below 1 kbar. Therefore, the solidus temperature of the Takakumayama granitic magma is considered to be approximated by that obtained by the hydrothermal experiments, *i.e.*, 720°C., and agrees with the minimum melting point of 720°C. at 1 kbar in the 'Granite System' experimentally determined by TUTTLE and BOWEN (1958). It is suggested, therefore, that the eutectic reaction may have taken place in the formation of the granite.

Conclusion

The present study gave the hydrothermal experiments on the Takakumayama granite, Kagoshima Prefecture, Japan. Petrogenetical informations were obtained from combinations of the field observations, the petrography, the petrochemistry and the hydrothermal experiments. It is concluded that the granite has been formed in environments near a water-saturated condition at pressures below 1 kbar and temperature about 720°C.

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