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Geochemistry of the Kiyosaki Granodiorite in the Ryoke Belt, central Japan

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三河地方領家帯の清崎花崗閃緑岩の地球化学的研究

沓掛俊夫*

(Abstract)

The Kiyosaki Granodiorite pluton, 5 km × 4 km, is one of the Older Ryoke granitoids in central Japan. Ten new analyses of the major rock-types are presented for major and trace elements, including REE. The geochemical characteristics of the Kiyosaki Granodiorite are as follows: calcic (Peacock's index ≒ 63), higher Mg/Fe ratio than other Older Ryoke granitoids, very low Fe^{+3}/Fe^{+2} ratio, peraluminous and metaluminous, low Rb content (mostly < 100 ppm) and moderately fractionated REE patterns. They are exclusively plotted within the volcanic arc granite field of some discrimination diagrams using trace elements for the tectonic setting of the granitoids. The magma of the Kiyosaki Granodiorite had been derived from the protoliths of basaltic andesite composition of the lower crust through the dehydration melting, leaving hornblende as a major residuum.

Introduction

Geochemical investigations on the Ryoke granitoids have been performed by several authors (Honma, 1974; Ishihara and Terashima, 1977; Kutsukake et al., 1979). I carried out the geochemical, along with the petrological, study on the Kamihara Tonalite, Tenryukyo Granite and Mitsuhashi Granite in the Mikawa district of central Japan (Kutsukake, 1993, 1997) and also the Ryoke granitoids in the Kinki district of southwest Japan (Kutsukake, 2001). For the Kiyosaki Granodiorite several analyses only for major elements have been made (Koide, 1958; Nakai, 1976). Here, I present new ten analyses of the representative rocks, and examine the geochemical variation and characteristics of this pluton plotting them onto several variation and discrimination diagrams, mentioning their petrogenetic implications.

Detailed petrographic descriptions of the Kiyosaki Granodiorite can be seen in Koide (1958) and Nakai (1976). And also, Nakai (1965) studied the morphology of zircons contained in the dark enclaves of this granitoid pluton.

Geological setting

The Ryoke Belt had been located on the continental-margin of the Eurasian continent before the opening of Japan Sea. The Inner Zone of southwest Japan constituted a segment

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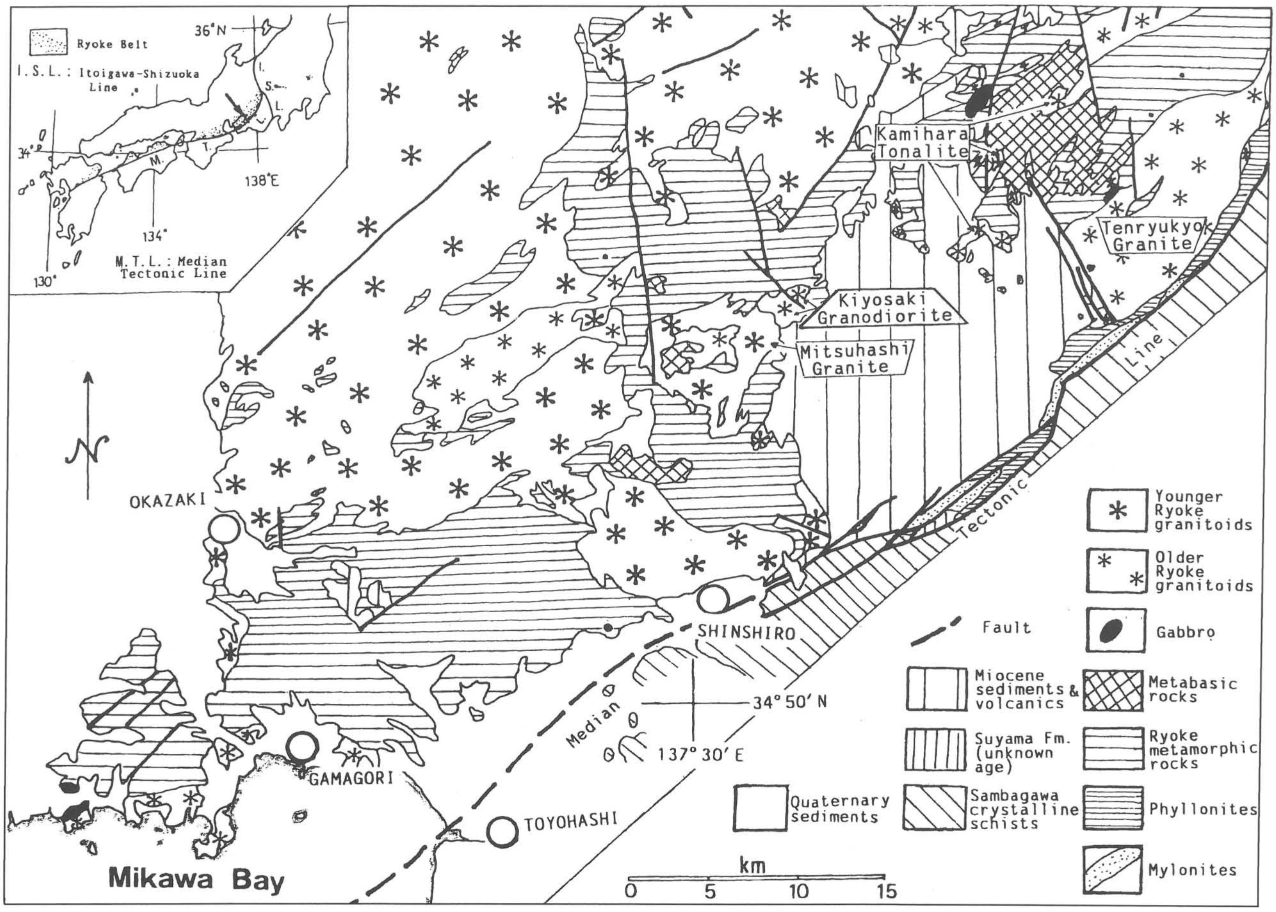

Fig. 1. Geological outline-map of the Ryoke Belt in the Mikawa district, central Japan.

of the Cretaceous-Paleogene batholithic belt of the Pacific Asia. The Ryoke Belt, composed of high T/low P metamorphic rocks, several types of mafic rocks and extensively developed granitoids, represents its magmatic front. The Ryoke granitoids exceed the metamorphic rocks in extent. They have been classified into the Older and Younger groups, according to their field relations and petrographic characteristics (Fig. 1). The Kiyosaki Granodiorite is one of the representative Older Ryoke granitoids in the Mikawa district of central Japan (Koide, 1958).

Available radiometric ages for the Kiyosaki Granodiorite are shown in Table 1. The CHIME (monazite) ages (Morishita et al., 1996) seem to be the most reasonable, as inferred from its field relations to other Ryoke granitoids (Ryoke Research Group, 1972; Nakai and Suzuki, 1996).

Table 1. Radiometric ages for the Kiyosaki Granodiorite.

Dating method	Age (Ma)	Reference
K - Ar (biotite)	69.5, 70.0	Ozima et al.(1967)
CHIME (monazite)	86.6, 87.0	Morishita et al.(1996)
U - Pb (zircon)	100, 105	Banks and Shimizu (1969)

Mode of occurrence and petrography

The Kiyosaki Granodiorite pluton, 5km (E-W) × 4 km (N-S), occurs within the Ryoke metamorphic rocks of the sillimanite-grade, and is intruded by the Mitsuhashi Granite to the southwest (Fig. 2). In many places within the pluton, thin dykes and small stocks of the Mitsuhashi Granite can be seen. Due to the cover of the Miocene Shitara Group, the southern extension of the pluton is not clear.

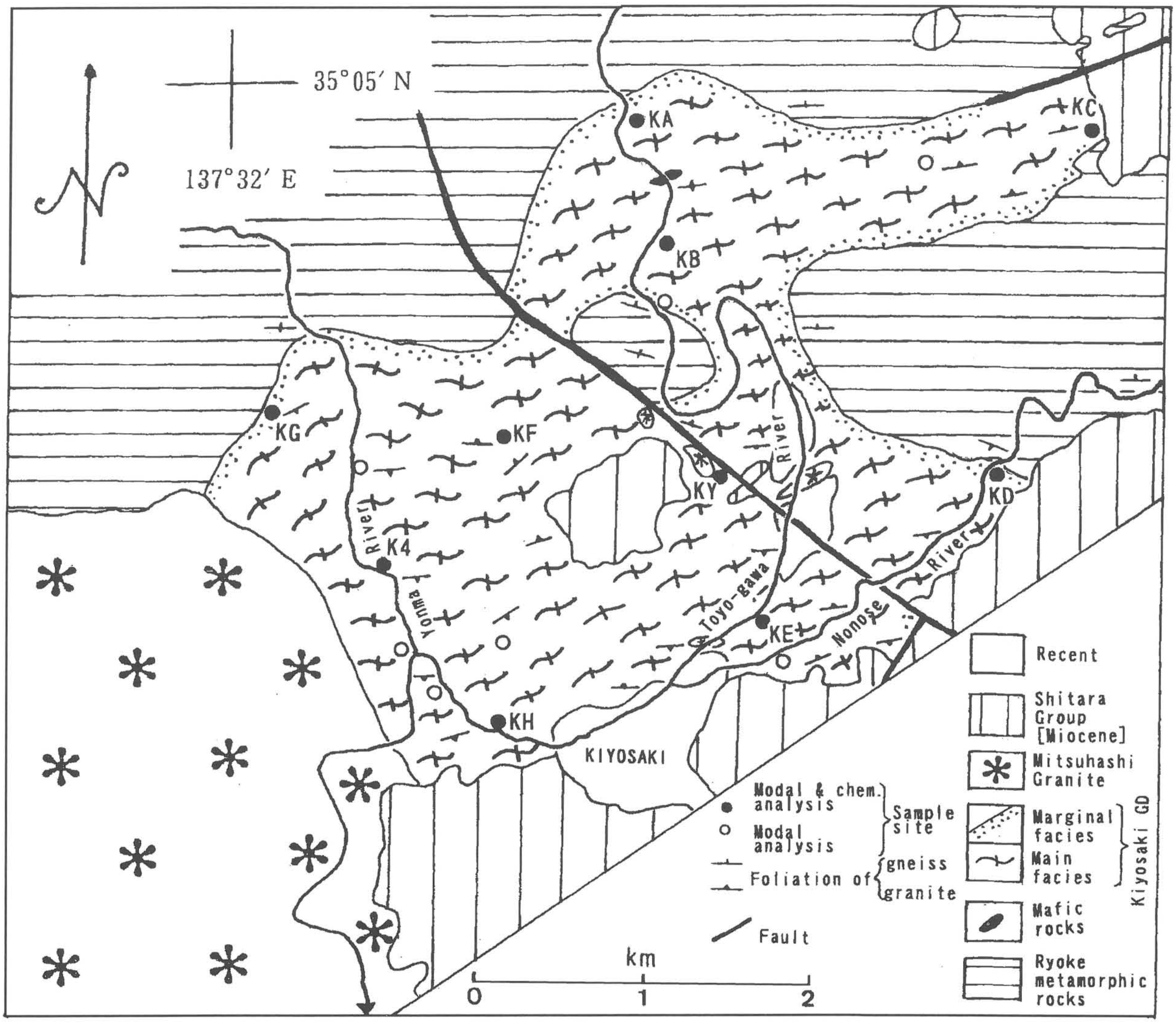

Fig. 2. Geological map and sample localities of the Kiyosaki Granodiorite pluton.

Also numerous Miocene rhyolite and andesite dykes are seen within and around the pluton, however, they are not shown in the geological map for simplicity. The shape of the pluton, as far as represented by the present exposures, looks like a pelican.

Within some tens to two hundreds meters from the contact a marginal facies is developed. The marginal facies rocks are more leucocratic than the main facies, and include the xenoliths of the host metamorphic rocks. Gneissic foliation, defined by the parallel alignment of mafic minerals, is not so distinctive in the southern portion, however, it fairly develops in the northern portion of the pluton. Frequently porphyritic alkali-feldspar crystals, 2 cm or more across, are seen. Ovoidal mafic enclaves, attaining 10 cm or more across, are ubiquitous.

The main facies comprise a medium-grained hornblende-biotite granodiorite and diopside-bearing hornblende-biotite granodiorite. Diopside-bearing rocks occur mainly in the southern portion of the pluton. Together with the above-mentioned rock, hornblende-diopside-biotite tonalite and diopside-biotite granodiorite without hornblende occur. There also occurs a biotite granodiorite, mainly in the northern portion.

The composition of diopside, estimated from the optics by Koide (1958), is of $Wo_{46}En_{37}Fs_{17}$. Hornblende is a green variety, and sometimes a replacement product of the diopside. Biotite (Z = grayish brown) forms a clot of several individuals. Apatite, monazite, allanite and zircon are common accessories.

Plagioclase has a composition of An_{33-47} in the diopside-bearing hornblende-biotite tonalite and granodiorite, An_{33-44} in the hornblende-biotite granodiorite, and An_{32-42} in the biotite granodiorite (Koide, 1958).

The marginal facies rocks are medium-grained biotite granite and muscovite-biotite granite.

The modal analyses for seventeen samples, of which fourteen are of main facies and three of marginal facies, have been undertaken using the thin sections, and their results are plotted onto the IUGS 's (1973) triangle (Fig. 3). The main facies rocks mostly fall in the granodiorite, and subordinately tonalite fields, whereas, the marginal facies rocks exclusively fall within the granite field.

Geochemistry

Ten samples, of which eight are of main facies and two of marginal facies rocks, are selected for ICP and XRF analyses. The analyses were undertaken at the Activation Laboratories, Ltd., Ontario, Canada. The results are shown in Table 2.

1. Major elements

SiO_2-range is rather narrow from 62.8 to 69.2 for the main facies rocks, whereas, the marginal facies rocks are more silicic, and contain 72.3 and 79.0 % of SiO_2. Calculated D.I. (differentiation index; Thornton and Tuttle, 1960) ranges from 58.5 to 75.1 for the main facies. Fe^{+3}/Fe^{+2} ratios are usually very low, ranging from < 0.1 to 0.3. This is so in other Ryoke granitoids (Kutsukake, 1993, 1997, 2001), which belong to the ilmenite-series of Ishihara (1977).

The analyses are plotted on the alkalies and lime vs. silica diagram (Fig. 4a). Regular decrease of CaO and increase of alkalies with increasing SiO_2 can be recognized for the main facies, and it seems to possess an alkali-lime (Peacock's) index of ca.63, although it is not certain because the less silicic rocks are absent. This value is almost the same as that (62.6) of the Older Ryoke granitoids around Lake

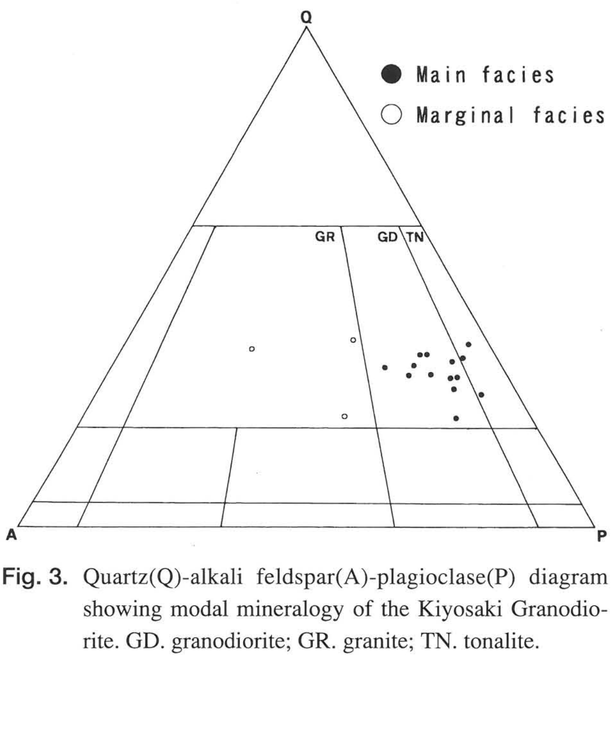

Fig. 3. Quartz(Q)-alkali feldspar(A)-plagioclase(P) diagram showing modal mineralogy of the Kiyosaki Granodiorite. GD. granodiorite; GR. granite; TN. tonalite.

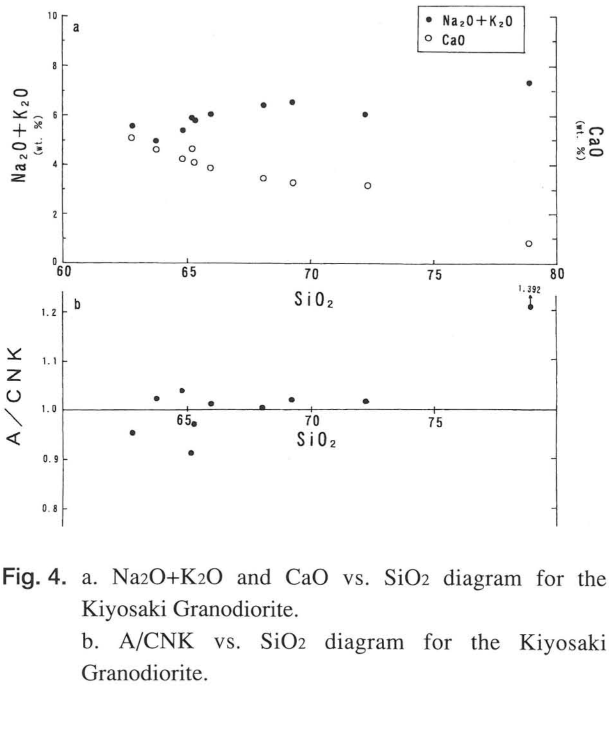

Fig. 4. a. Na_2O+K_2O and CaO vs. SiO_2 diagram for the Kiyosaki Granodiorite.
b. A/CNK vs. SiO_2 diagram for the Kiyosaki Granodiorite.

Table 2. Major- and trace-element analyses of the Kiyosaki Granodiorite.

	KA	KB	KC	KD	KE	KF	KG	KH	K4	KY
SiO_2	68.01	69.22	65.28	78.98	65.12	62.79	72.30	65.89	64.80	63.74
TiO_2	0.41	0.39	0.39	0.11	0.58	0.71	0.31	0.58	0.61	0.71
Al_2O_3	15.34	15.31	15.36	11.71	15.37	16.17	14.66	15.60	16.07	15.91
Fe_2O_3	0.01	0.21	0.78	0.23	0.60	0.33	0.32	1.11	0.36	0.53
FeO	3.26	2.95	2.83	0.61	3.99	4.89	2.21	3.52	4.16	4.48
MnO	0.07	0.07	0.08	0.01	0.10	0.10	0.04	0.08	0.08	0.09
MgO	1.14	1.08	1.14	0.09	2.23	2.97	0.52	2.10	2.24	2.90
CaO	3.46	3.22	4.07	0.84	4.66	5.09	3.13	3.95	4.28	4.67
Na_2O	3.57	3.65	3.63	2.53	3.41	3.01	3.71	2.95	3.36	3.04
K_2O	2.88	2.90	2.24	4.84	2.52	2.58	2.35	3.10	1.98	1.96
P_2O_5	0.10	0.10	0.13	0.03	0.15	0.16	0.12	0.15	0.15	0.15
L.O.I.	0.92	0.81	3.35	0.37	1.29	1.54	0.51	1.24	1.11	1.64
Total	99.16	99.91	99.27	100.35	100.02	100.34	100.18	100.27	99.21	99.83
Trace elements (in ppm except for Au and Ir) [I C P]										
Ag	-	-	-	-	-	-	-	-	<0.4	<0.4
As	2	3	4	4	1	3	2	2	<1	2
Au (ppb)	<2	<2	<2	<2	<2	<2	<2	<2	<1	1
Ba	585	543	525	1241	826	582	1343	848	316	504
Be	2	2	2	<1	2	2	1	2	2	2
Bi	-	-	-	-	-	-	-	-	<5	<5
Br	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
Cd	-	-	-	-	-	-	-	-	1.0	1.3
Co	8.6	7.3	7.5	1.0	12.8	17.2	3.6	12.3	13.6	15.9
Cr	8.5	9.7	8.1	1.0	57.9	79.8	2.2	52.1	49.9	78.8
Cs	4.6	7.5	5.4	2.2	2.7	4.4	1.1	2.8	4.0	4.3
Cu	-	-	-	-	-	-	-	-	13	15
Hf	4.6	4.5	4.8	4.9	4.3	4.6	9.3	4.8	4.2	4.0
Hg	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Ir (ppb)	<2	<2	<2	<2	<2	<2	<2	<2	<1	<1
Mo	<2	<2	<2	3	<2	<2	2	<2	<2	<2
Ni	-	-	-	-	-	-	-	-	15	24
Pb	-	-	-	-	-	-	-	-	10	8
Rb	120	120	86	90	74	91	54	96	91	94
Sb	<0.1	0.4	0.3	0.2	0.2	0.3	0.6	0.3	0.1	0.1
Sc	7.8	6.7	8.4	0.4	10.9	15.1	3.1	10.5	11.6	13.0
Se	<0.5	1.0	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
Sr	227	215	290	82	283	302	295	282	270	297
Ta	0.9	0.7	0.6	0.3	0.6	0.6	0.3	0.9	1.1	1.1
Th	10.2	13.2	7.5	11.5	6.4	13.8	3.3	10.9	11.1	6.6
U	1.2	3.0	1.1	1.3	0.8	3.4	1.4	1.0	1.2	1.6
V	44	46	42	<5	78	103	14	77	77	100
W	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Y	19	17	20	6	15	21	10	16	17	17
Zr	163	158	186	163	167	181	316	189	149	160
La	28.1	31.9	23.9	33.1	22.4	33.9	19.2	28.1	36.2	19.5
Ce	54	54	45	70	42	63	39	51	63	36
Nd	23	24	19	34	18	27	18	19	24	15
Sm	4.28	3.92	3.85	6.52	3.50	4.97	2.77	3.38	4.01	3.18
Eu	1.01	0.96	1.07	0.97	1.20	1.20	1.76	1.12	1.05	0.98
Tb	0.6	0.5	0.6	0.5	0.4	0.6	0.4	0.4	0.5	0.5
Yb	1.78	1.67	2.10	0.65	1.20	1.85	1.04	1.39	1.70	1.49
Lu	0.26	0.25	0.32	0.10	0.18	0.27	0.15	0.20	0.25	0.21
Trace elements (in ppm) [X R F]										
Ga	19	17	19	13	20	19	18	19	21	19
Nb	11	6	9	6	6	13	8	12	10	10
Pb	39	24	13	24	8	13	10	14	10	7
Rb	103	107	87	87	77	83	45	96	85	83
Sn	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
S	655	225	455	<50	150	150	150	80	55	180

Analyzed by the Activation Laboratories, Ltd., Ontario, Canada. -: not determined.

KA: Specimen no.00010902; Hornblende-biotite granodiorite
KB: Specimen no.00010901; Biotite granodiorite
KC: Specimen no.99042901; Diopside-biotite granodiorite
KD: Specimen no.99050204; Muscovite-biotite granite(marginal facies)
KE: Specimen no.99033001b; Hornblende-biotite tonalite
KF: Specimen no.99121201; Diopside-hornblende-biotite tonalite
KG: Specimen no.99110701; Biotite granite (marginal facies)
KH: Specimen no.98112301; Diopside-hornblende-biotite granodiorite
K4: Specimen no.98112304; Hornblende-diopside-biotite granodiorite
KY: Specimen no.99011601a; Hornblende-diopside-biotite granodiorite

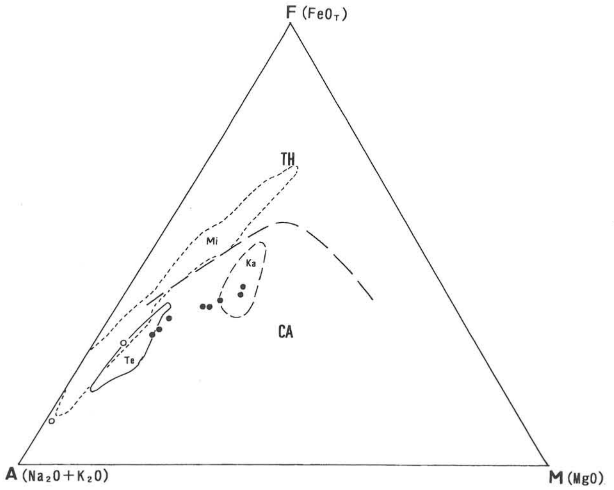

Fig. 5. AFM-diagram for the Kiyosaki Granodiorite.
The symbols are as in Fig. 3. The bold dashed line separates calc-alkaline (CA) from tholeiitic (TH) suites (Irvine and Baragar, 1971). The areas surrounded by the dashed line (Ka), solid line (Te), and dotted line (Mi) are for the Kamihara Tonalite, Tenryukyo Granite and Mitsuhashi Granite, respectively (Kutsukake, 1993, 1997).

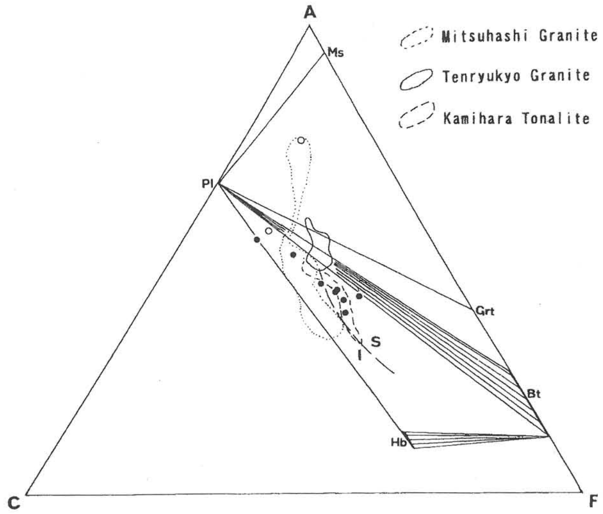

Fig. 6. ACF diagram for the Kiyosaki Granodiorite.
The symbols are as in Fig. 3. The boundary between I-type (I) and S-type (S) granitoids is after Hine et al. (1978). For the areas of other Ryoke granitoids refer to Kutsukake (1993, 1997).
Bt. biotite; Hb. hornblende; Grt. garnet; Ms. muscovite; Pl. plagioclase.

Sakuma (Kutsukake, 1993).

The analyses are plotted onto the AFM-diagram (Fig. 5). They fall exclusively within the calc-alkaline field (Irvine and Baragar, 1971) and also show a typical calc-alkaline trend as well as other Older Ryoke granitoids in the Mikawa district.

Calculated A/CNK (mol. Al_2O_3/[CaO+Na_2O+K_2O]) are plotted against SiO_2 (Fig. 4b). Most of them fall between 0.90 and 1.05, therefore, they are metaluminous and peraluminous (Debon and LeFort, 1983). A/CNK is less than 1.25 in I-type granitoids (White and Chappell, 1977), and this is so in the Kiyosaki Granodiorite.

The analyses are also plotted on the ACF diagram modified by White and Chappell (1977) (Fig. 6). Most of them fall within the field of plagioclase+biotite+hornblende, and this is consistent with the fact that they contain both biotite and hornblende. A rock including only biotite as mafic mineral consistently lies on the tie-line connecting plagioclase and biotite. Also a diopside-rich rock lies above the line connecting plagioclase and hornblende. A marginal facies rock falls within the field of those of the Mitsuhashi Granite (Kutsukake, 1997).

Half of the analyses fall in the S-type side of the boundary between I- and S-type granitoids (Hine et al., 1978). The Older Ryoke granitoids such as the Kamihara Tonalite, which can be regarded petrographically to belong to I-type, are frequently plotted in the S-type field in this diagram (Kutsukake, 1993). This is so for the Kiyosaki Granodiorite, indicating its relatively alumina-rich chemistry.

2. Trace elements

Forty-two trace elements were analyzed (Table 2). Rb content ranges from 76 to 120 ppm for the main facies. These Rb values are in the range of the volcanic arc granites of Pearce et al. (1984), and slightly lower than the average value (132 ppm at 69.1 % SiO_2) of the typical I-type granitoids of the Lachlan Fold Belt of Australia (White and Chappell, 1983).

Sn and W are under the detection limit for all of the samples; this is in accord with the fact

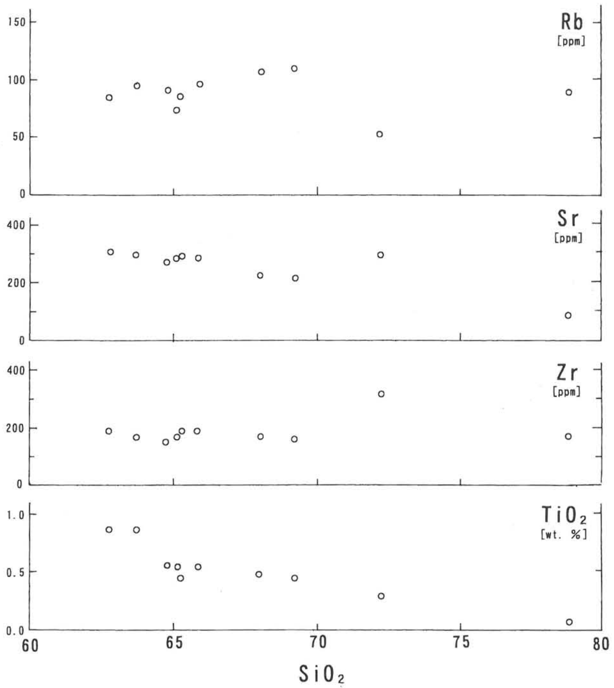

Fig. 7. Rb-Sr-Zr-Ti patterns for the Kiyosaki Granodiorite, plotted vs. SiO_2.

that the Ryoke granitoids are lacking in the mineralization of these metals. Only a marginal facies rock contains 3 ppm of Mo. In this regard it is worthwhile to mention that some of the Ryoke granitoids are related to trace Mo-mineralization (Sato and Nakai, 1991).

Some of the trace elements are plotted against SiO_2 (Fig. 7). Slight increase of Rb and decrease of Sr with increasing SiO_2 can be recognized for the main facies rocks. Zr stays almost constant for the main facies. Generally TiO_2 decreases with increasing SiO_2.

In the Sr vs. modal plagioclase plots (Fig. 8a), a positive correlation is not seen. Therefore, the plagioclase fractionation could not be a major factor in the formation of lithologic variations of the Kiyosaki Granodiorite, as Sr enters Ca sites of plagioclase. This is also supported by the TiO_2 vs. Zr plots (Fig. 8b), which indicate neither

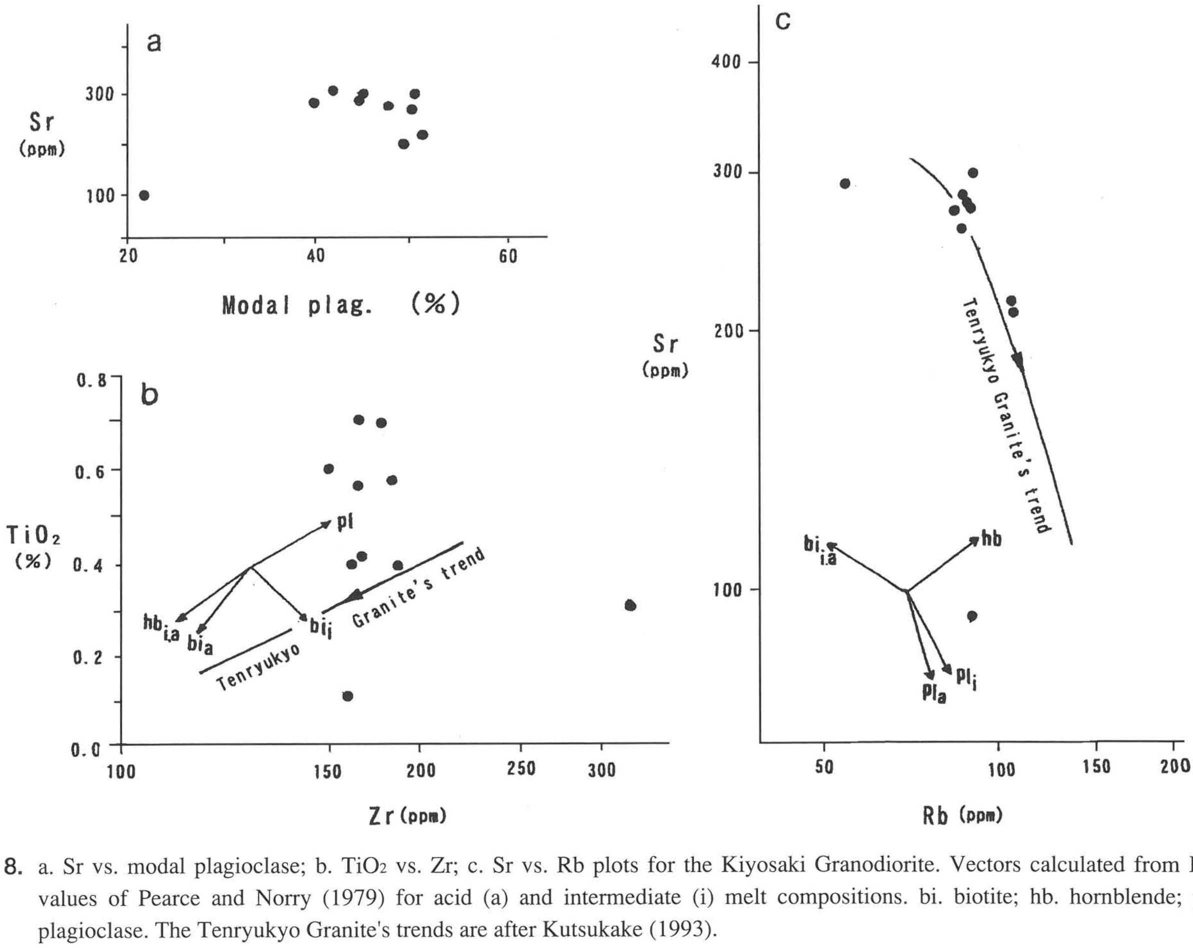

Fig. 8. a. Sr vs. modal plagioclase; b. TiO_2 vs. Zr; c. Sr vs. Rb plots for the Kiyosaki Granodiorite. Vectors calculated from K^D values of Pearce and Norry (1979) for acid (a) and intermediate (i) melt compositions. bi. biotite; hb. hornblende; pl. plagioclase. The Tenryukyo Granite's trends are after Kutsukake (1993).

plagioclase nor biotite fractionation to be responsible to this trend, although the Tenryukyo Granite exhibits a clear plagioclase fractionation trend (Kutsukake, 1993). Rb-Sr relations (Fig. 8c) do not show any particular tendencies of fractionation. These trace element relations do not provide any clues to the elucidation of lithologic variations of the Kiyosaki Granodiorite.

In the discrimination diagrams in terms of tectonic setting of granites of Pearce et al. (1984), the rocks of the Kiyosaki Granodiorite exclusively fall in the volcanic arc granite (VAG) fields (Fig. 9). Also in the Hf-Rb-Ta diagram (Harris et al., 1986), they fall within the volcanic arc granites field (Fig. 10). These

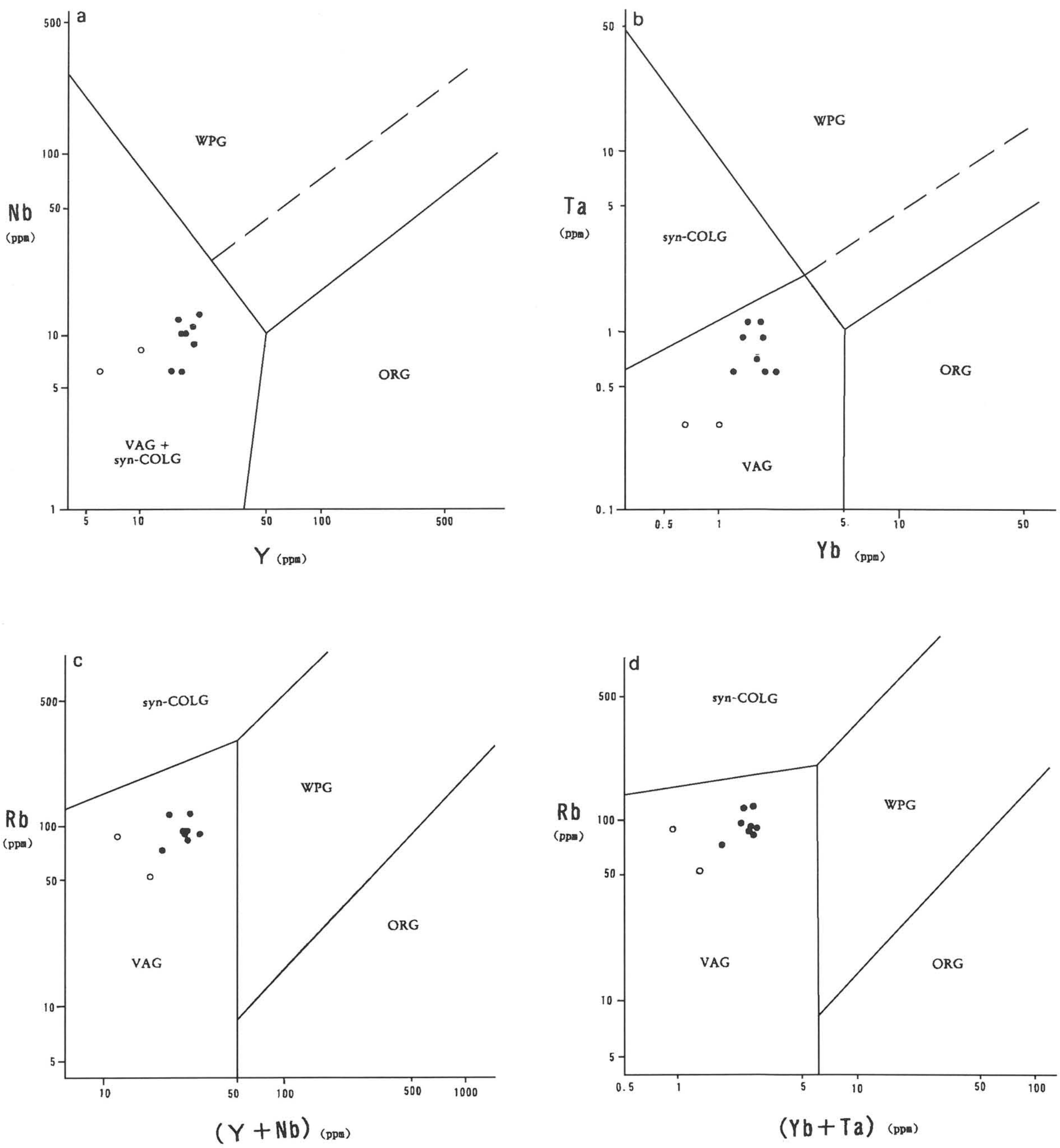

Fig. 9. a. Nb vs. Y; b. Ta vs. Yb; c. Rb vs. (Y+Nb); d.Rb vs. (Yb+Ta) discrimination diagrams (Pearce et al., 1984) for the Kiyosaki Granodiorite. The symbols are as in Fig. 3

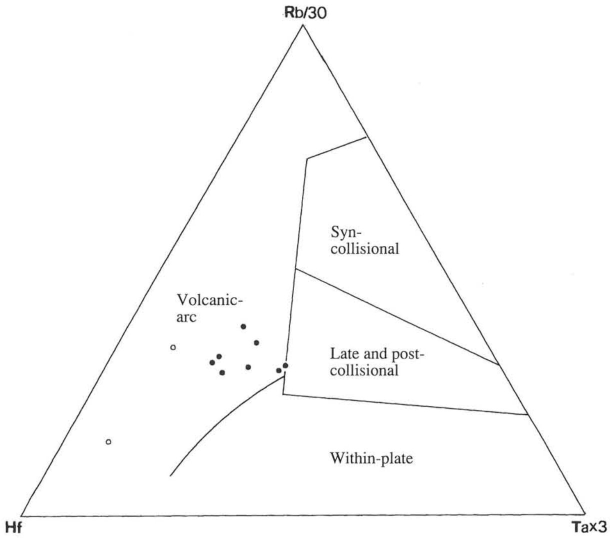

Fig. 10. Hf-Rb-Ta discrimination diagram (Harris et al., 1986) for the Kiyosaki Granodiorite. The symbols are as in Fig. 3.

are in harmony with the fact that the Ryoke granitoids are the products of a continental margin arc magmatism.

3. REE

Total REE contents of the main facies rocks range from 77 ppm to 135 ppm, whereas two marginal facies rocks contain 146 ppm and 82 ppm of REE, respectively. Chondrite-normalized $(La)_N$ = 60 ~ 110, $(Lu)_N$ = 5.5 ~ 10 and $(La/Lu)_N$ = 8.6 ~ 13.8 for the main facies rocks.

Chondrite-normalized REE patterns are shown in Fig. 11. The main facies rocks exhibit the quite similar pattern, although they are somewhat variable for heavy REE. The patterns are characteristically steeply sloped for light REE, however, they are rather flat for heavy REE. These patterns are common to other Ryoke granitoids (Kutsukake, 2001). Eu-anomalies are almost non-existent. When they are recognized, they are either slightly positive or negative.

The marginal facies rocks exhibit more fractionated, heavy REE depleted, patterns than the main facies. One sample shows the positive and another one does the negative Eu-anomalies, respectively.

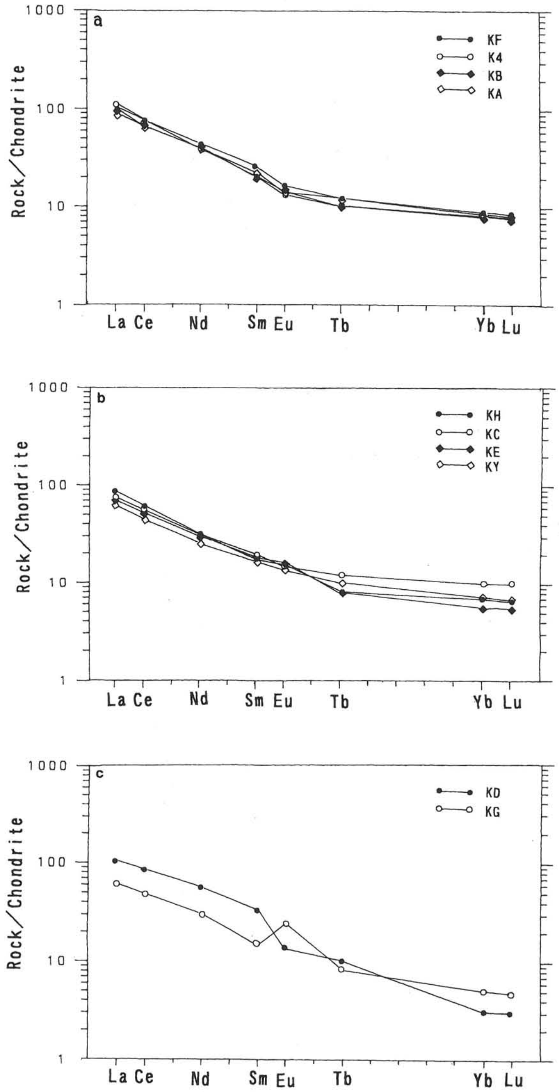

Fig. 11. Chondrite-normalized REE patterns for the main facies (a and b) and marginal facies (c) of the Kiyosaki Granodiorite. For sample number refer to Table 2.

Discussion

1. Chemical control of the crystallization of diopside

The Kiyosaki Granodiorite is unique in the frequent occurrence of diopside. However, it is not necessarily more calcic than other Ryoke granitoids. For most of the major elements, the conspicuous differences can not be detected between the Kiyosaki Granodiorite and the Kamihara Tonalite, which rarely includes diopside. Only the Mg/Fe ratio, however, differs between the two granitoids. The MgO/ total FeO ratios in the Kiyosaki Granodiorite and Kamihara Tonalite are of 0.531 and 0.476 at SiO_2 ≒ 64 wt.%, respectively (Kutsukake, 1993). The higher Mg/Fe ratio should have favored the crystallization of diopside in the Kiyosaki Granodiorite, being the crystallization conditions, such as water-vapor pressure and oxygen-fugacity, almost the same as other granitoids.

2. Trace elements' implications

The trace element contents and their relationships of the Kiyosaki Granodiorite indicate that it belongs to the volcanic arc granites, and it is in harmony with its tectonic setting, having been formed by the continental-margin arc magmatism.

Relatively narrow chemical variations among the major facies of the Kiyosaki Granodiorite indicate a limited fractionation, operated during the crystallization of the magma. The major fractionation phases are neither plagioclase nor biotite, which are the predominant fractionation phases of the Tenryukyo Granite (Kutsukake, 1993). These trace element data provide no definitive clues as regards the major fractionation phases of the Kiyosaki Granodiorite. The chondrite-normalized REE patterns also show non or quite limited plagioclase fractionation.

3. Magma genesis

The I-type characteristics, calcic nature and the predominance of granodiorite of the Kiyosaki Granodiorite require the protoliths of fundamentally basaltic or basaltic andesitic composition. The tonalitic Ryoke granitoids are most likely to have been derived from a dehydration-melting of amphibolites or garnet amphibolites (Kutsukake, 2001).

Virtual absence of Eu-anomalies suggests that plagioclase had not been a major residual mineral. And also moderate heavy REE depletion indicates that the garnet, which has large partition coefficients for HREE, had not been a major residual mineral. The hornblende should be the most probable candidate of the major residual minerals. The experimental dehydration melting of amphibolite has produced the tonalitic and granodioritic melts under 0.8 ~ 1 GPa, although these melts are slightly richer in alumina and magnesia than the tonalites and granodiorites of the Kiyosaki Granodiorite (Johannes and Wolke, 1994). The residual phases are hornblende, plagioclase, clinopyroxene and garnet/orthopyroxene in these experiments. The REE patterns, however, indicate that hornblende is the major residual phase among these residues for the Kiyosaki Granodiorite's magma.

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（要　旨）

沓掛俊夫：三河地方領家帯の清崎花崗閃緑岩の地球化学的研究.

この岩体を構成する主要な岩型につき10個のサンプルの主成分元素と42種の微量元素（REEを含む）の分析を行った．地球化学的特徴は，石灰質（Peacock's index≒63），他の古期領家花崗岩類に比べて，Mg/Fe 比の高いこと，著しく低い Fe^{+3}/Fe^{+2}, 低いRb 含有量（ほとんど, <100 ppm），中程度に分化したREEパターンなどである．微量元素を用いた花崗岩のテクトニックな分類図では，すべてVAGの領域に落ちる．REEのパターンは，Euの異常はほとんど認められず，HREEに関して平坦なパターンを示す．これらの地球化学的特徴は，清崎花崗閃緑岩のマグマが，下部地殻を構成する角閃岩の脱水溶融により生じたとして矛盾しない．