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Compositional variations of some constituent minerals of the late Mesozoic to early Tertiary granitic rocks of southwest Japan

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Abstract: The late Mesozoic to early Tertiary granitic rocks on the inner side of Southwest Japan are divided into three zones, Ryoke, Hiroshima and Sanin, in the order of emplacement. They are arranged from south to north with E-W extension. The three zones do not show any great difference in whole rock chemistry, except for oxidation ratio. However, there are comparatively marked differences in the chemical composition of the constituent minerals, such as hornblendes, biotites and K-feldspars. The Fe/Mg ratios in both hornblendes and biotites, and the Al-content of hornblendes become lower towards the north zone. The Or-content of the K-feldspars is highest in the granitic rocks of the Ryoke zone and lowest in the Sanin zone.

These data suggest the highest temperature and the shallowest emplacement for the formation of the Sanin zone, and the reverse for the Ryoke zone. This implies that the level of erosion deepens from north to south. The isotopic dating data seem to agree with this conception, because the interval between the mineral age and the whole rock or whole rock-mineral isochron age becomes shorter northward.

INTRODUCTION

In an earlier paper (Murakami, 1974), the writer divided the late Mesozoic to early Tertiary granitic rocks of Southwest Japan into three zones, Ryoke, Hiroshima and Sanin from south to north. Each of the zones has a roughly E-W elongation (Fig. 1). The emplacement of these granitic rocks is youngest in the Sanin zone (about 40 to 60 m.y.) and oldest in the Ryoke zone (70 to 100 m.y. for the Ryoke and the Hiroshima granites). There is no great difference in isotopic age between the Ryoke granites and the Hiroshima granites, but the former is always intruded by the latter, and emplaced into the high-grade metamorphic terrain different from that of the latter which is unmetamorphosed except for low-grade contact metamorphic aureoles.

Recently, the writer investigated the whole rock chemistry and the chemical composition of the biotites, hornblendes and K-feldspars of these granitic rocks and some data on their isotopic meneral and whole rock isochron ages.

WHOLE ROCK CHEMISTRY

As already described by Aramaki and Nozawa (1974), and Ishihara (1971;1973), notable differences in major chemical constituents can not be found among the three zones of granitic rocks except for the oxidation ratio. This is indicated in Figures 2 and 3 in which the variations of K_2O and CaO are plotted against SiO_2. Although granodioritic to quartz dioritic rocks are most prominent in the Ryoke zone, there is no great difference in chemistry of similar rocks among the three zones. In this respect, it must be noted that the granitic rocks under consideration are clearly different in the variations of major chemical constituents from those in some other batholiths of the Circum-Pa-

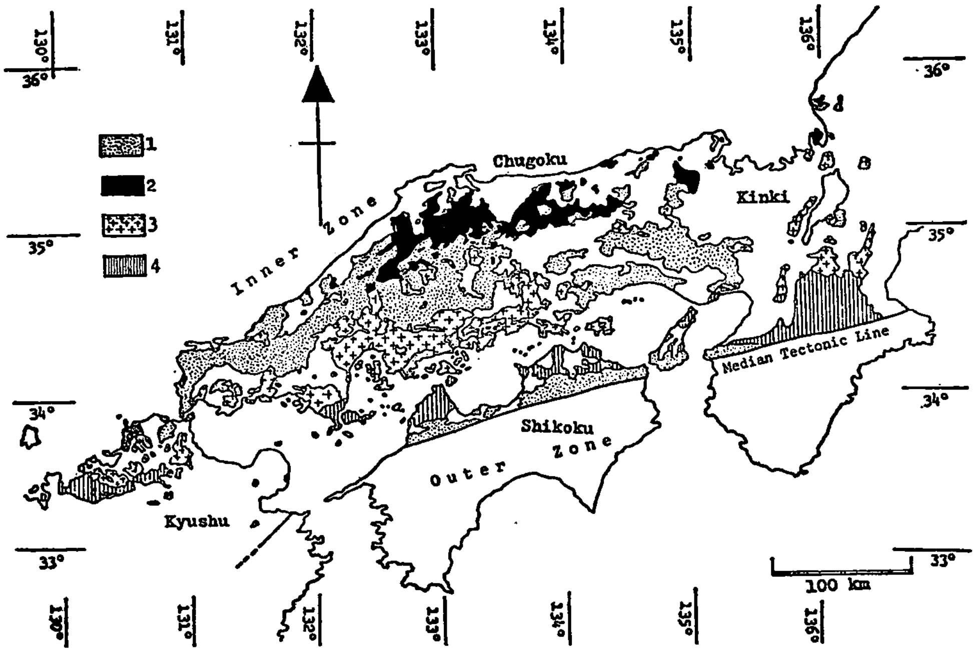

Fig. 1. Distribution of late Mesozoic to early Tertiary granitic rocks on the inner side of Southwest Japan. 1: Late Mesozoic to early Tertiary volcanic rocks. 2: Sanin granites. 3: Hiroshima granites. 4: Ryoke granites.

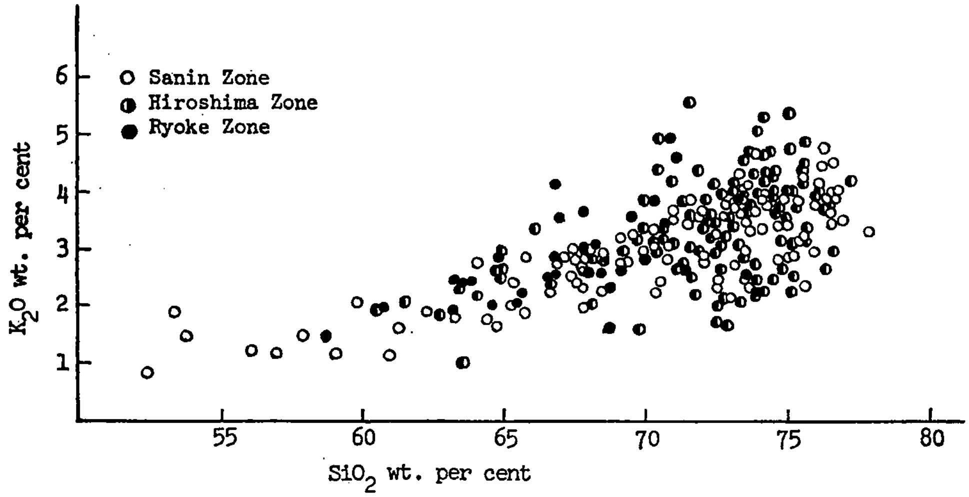

Fig. 2. Variation of K_2O vs. SiO_2 in whole rocks.

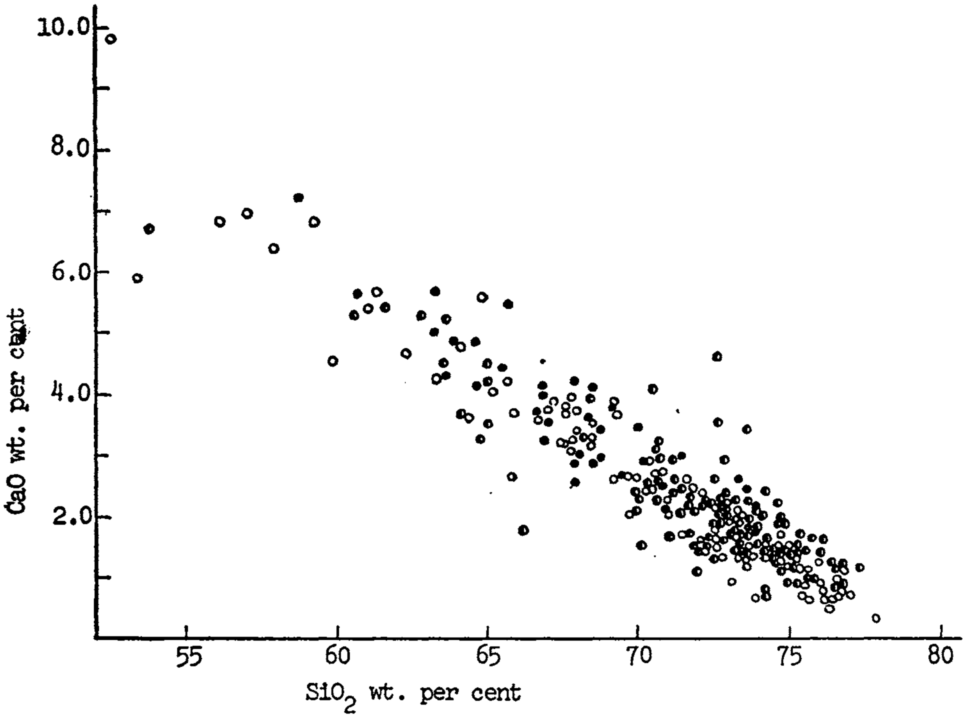

Fig. 3. Variation of CaO vs. SiO_2 in whole rocks. Symbols are the same as in Fig. 2.

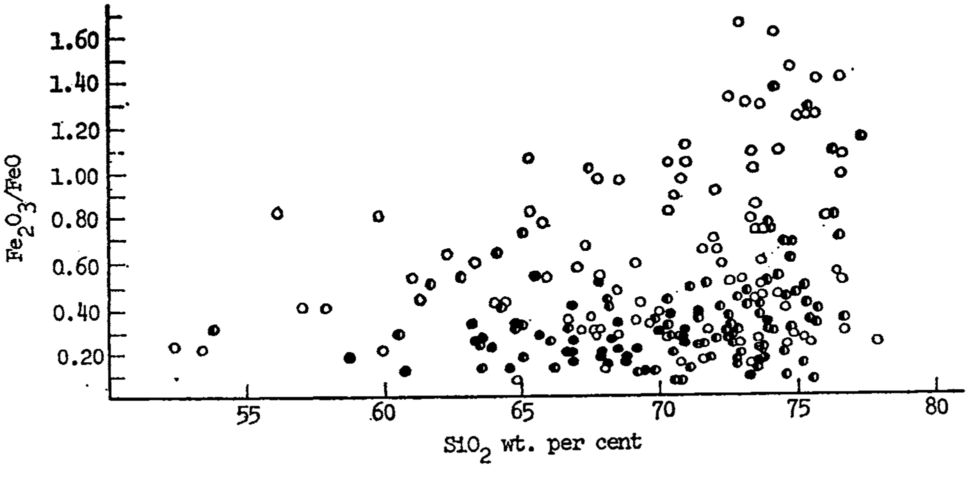

Fig. 4. Variation of Fe_2O_3/FeO ratio vs. SiO_2 in whole rocks. Symbols are the same as in Fig. 2.

cific region, such as in the Sierra Nevada (Bateman and Dodge, 1970) and in British Columbia (Roddick and Hutchison, 1972), where the inlandward increase of potash of these batholiths is pointed out. The Dickinson's model (Dickinson, 1970) on the generation of granitic magma along the subduction zone may not be appropriate to the granitic rocks of Southwest Japan.

The variation of ferric to ferrous iron ratio (oxidation ratio) with respect to SiO_2 (Fig. 4), as cited before, is highest in the Sanin zone and lowest in the Ryoke zone. Although the distinction between the Ryoke and the Hiroshima zones is not clear, the low-ratio rocks are more predominant in the former. This divergence is connected with the constituent opaque minerals, as investigated by Tsusue and Ishihara (1974) who confirmed that the Sanin granites are characterized by a fair abundance of magnetite, whereas ilmenite is the dominant accessory mineral in the Ryoke and Hiroshima zones, although scanty in the Ryoke zone. These reflect the marked difference in magnetic susceptibility among the three zones (Kanaya and Ishihara, 1973).

MINERAL CHEMISTRY

1. *Fe/Mg ratio in biotites and hornblendes*

There is a great difference among the three zones in the Fe/Mg ratio of constituent biotites and hornblendes in the granitic rocks. (Fig. 5). The Fe/Mg ratio in biotites and hornblendes in the Ryoke and the Hiroshima granites increases with the increase of the ratio in the whole rocks. On the other hand, in the Sanin zone this tendency is reversed. Comparing the rocks of similar chemical composition, the ratio is lowest in the Sanin zone, especially in the acid rocks. If equilibrium was roughly attained among the constituent biotites, hornblendes and iron ores during the formation of the granites, this fact indicates that the oxygen partial pressure was highest in the Sanin zone and lowest in the Ryoke zone (Murakami, 1969). This conclusion agrees with that drawn from the oxidation ratio in the whole rocks.

In the Fe^{+3}–Fe^{+2}–Mg diagram (Fig. 6) the biotites from the Ryoke zone are plotted within the comparatively narrow area near Fe_2SiO_4–SiO_2–Fe_3O_4– and Ni–NiO–buffered biotites of Wones and Eugster (1965). In contrast, most of the biotites in the Hiroshima and Sanin zones are higher in Fe^{+3} and plot near the Fe_3O_4–Fe_2O_3–buffered biotites. These data suggest that the Ryoke granites formed under more reducing conditions than the Hiroshima and Sanin granites.

2. *Bulk chemical composition, optical angle and obliquity of K-feldspars*

The K–feldspars from the Sanin zone (Fig. 7) are most sodic, ranging approximately from Or 50 to Or 80, whereas those from the Ryoke zone are most potassic ranging from Or 80 to Or90. The potassic part of the K–feldspars from the Hiroshima and the Ryoke zones is less calcic than the sodic part, though only slightly.

In addition to this difference in bulk composition, it must be stressed that the trend of evolution of the K–feldspars in the Sanin granites is quite reversed to that in the Hiroshima granites. The K–feldspars in the Tamagawa granites (Fig. 8) belonging to the Sanin zone become more sodic with increasing Ab-content of coexisting plagioclases. The final composition is Or 48.0, Ab 48.9, An 3.1 in mole per cent (Murakami, 1971a). On the contrary, the trend line of K–feldspars from the Kuga granites belonging to the Hiroshima zone connects the Or-poor side to the Or-rich side. The final composition is Or 90.1, Ab 8.4, Or 1.5 (Murakami, 1971lb; Murakami, *et al.*, 1964). During the crystallization of granite magma, the composition of the K–feldspars becomes

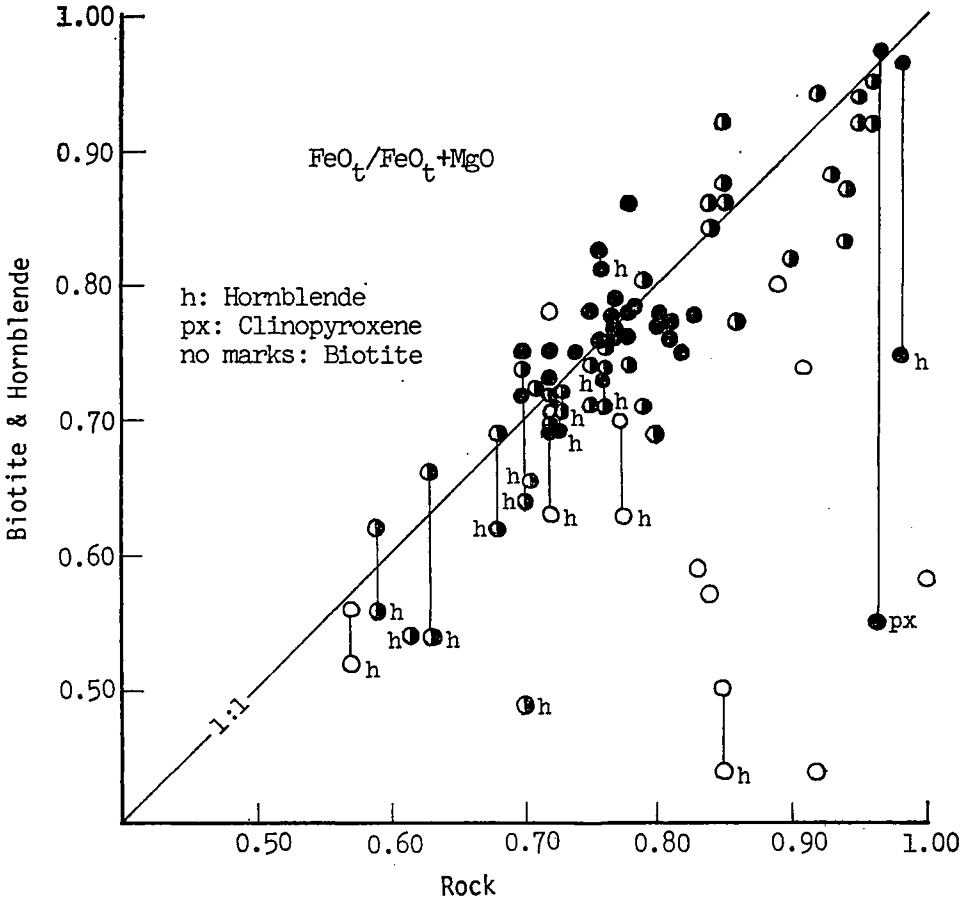

Fig. 5. Relation between FeOt/FeOt + MgO in whole rocks and that in constituent biotites and hornblendes. Coexisting biotites and hornblendes are connected by tie lines. Data from Murakami (1969) and Tachikawa and Fujimoto (1967), including unpublished information. Symbols are the same as in Fig. 2.

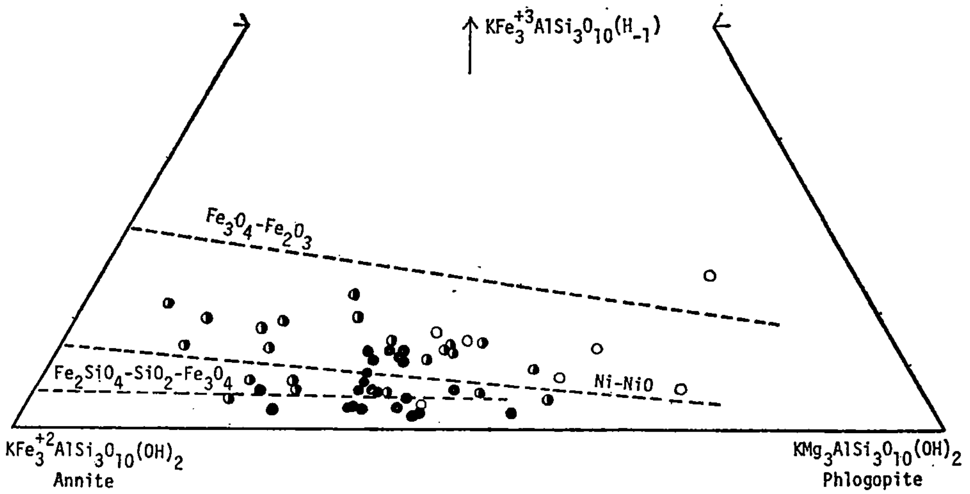

Fig. 6. Fe^{+3}-Fe^{+2}-Mg triangular diagram for biotites. Buffer lines after Wones and Eugster (1965). Data from Murakami (1969), including unpublished information. Symbols are the same as in Fig. 2.

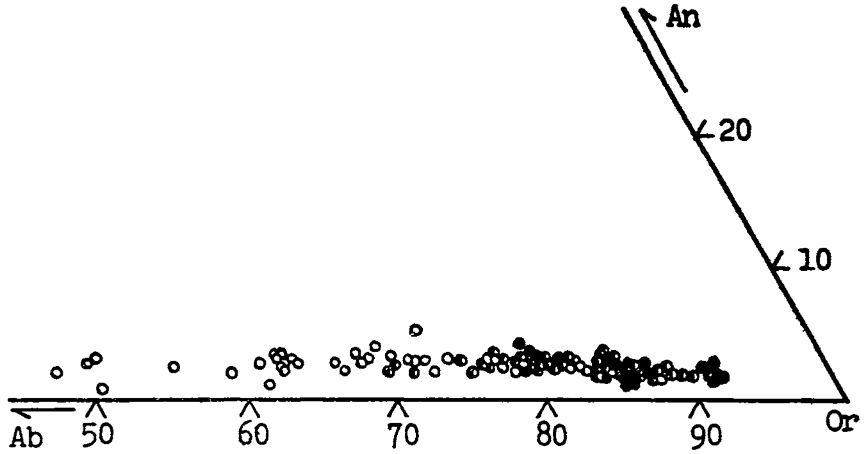

Fig. 7. Bulk chemical composition of K-feldspars. Data from Murakami *et. al.* (1964) and Murakami (1971a; 1971b), including unpublished information. Symbols are the same as in Fig. 2.

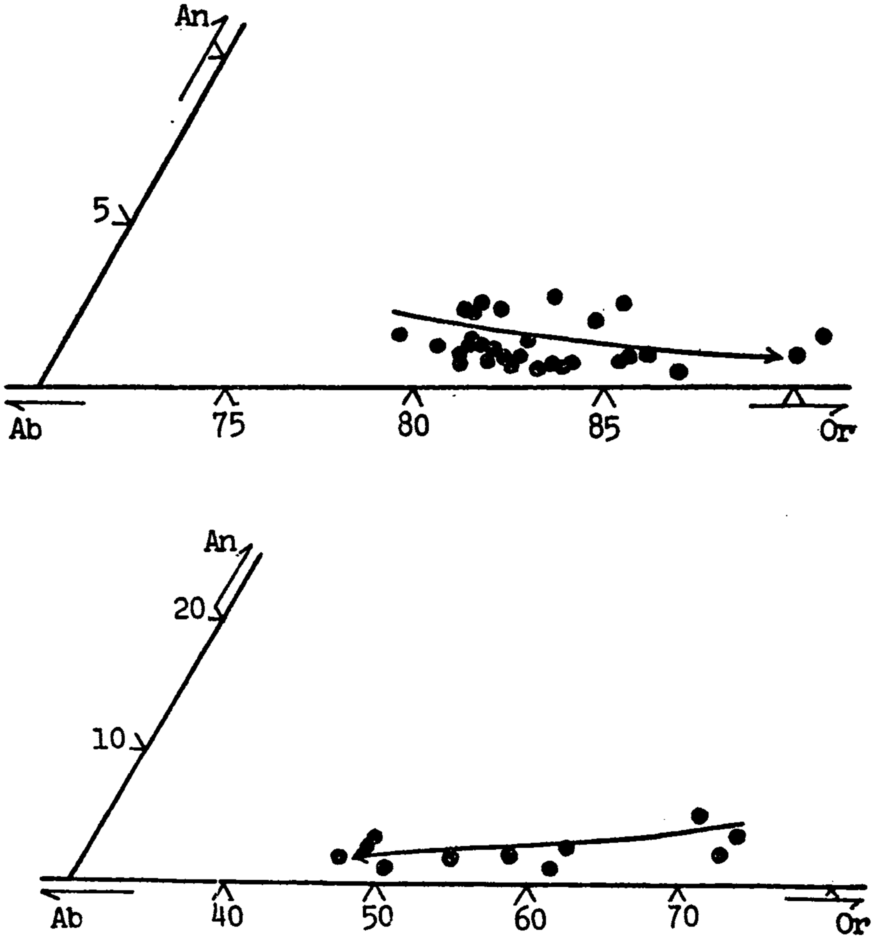

Fig. 8. Contrastive trends of evolution of K-feldspars between the Tamagawa (lower) and Kuga (upper) granites (see text). Data from Murakami (1971a; 1971b).

more albitic along the solidus line on the Ab-Or phase diagram. In the subsolidus region, however, the composition is controlled by the solvus line, becoming more and more potassic with falling temperature. Consequently, the Tamagawa granites are typical hypersolvus granites, whereas the Kuga granites are apparently subsolvus. The K–feldspars in the Ryoke granites seem to have a trend similar to that in the Kuga granites (Murakami, *et al.*, 1964), although the compositional range of the coexisting plagioclases is narrow.

The formative condition of K–feldspars is reflected by their optical angle and obliquity. The K–feldspars of the Sanin granites (Fig. 9) are lower in both optical angle and obliquity (0–0.2) than those of the Hiroshima and the Ryoke granites which have a wide range in obliquity (0–1.0) and also in optical angle. Maximum or near-maximum microcline is recognized in these two granites only.

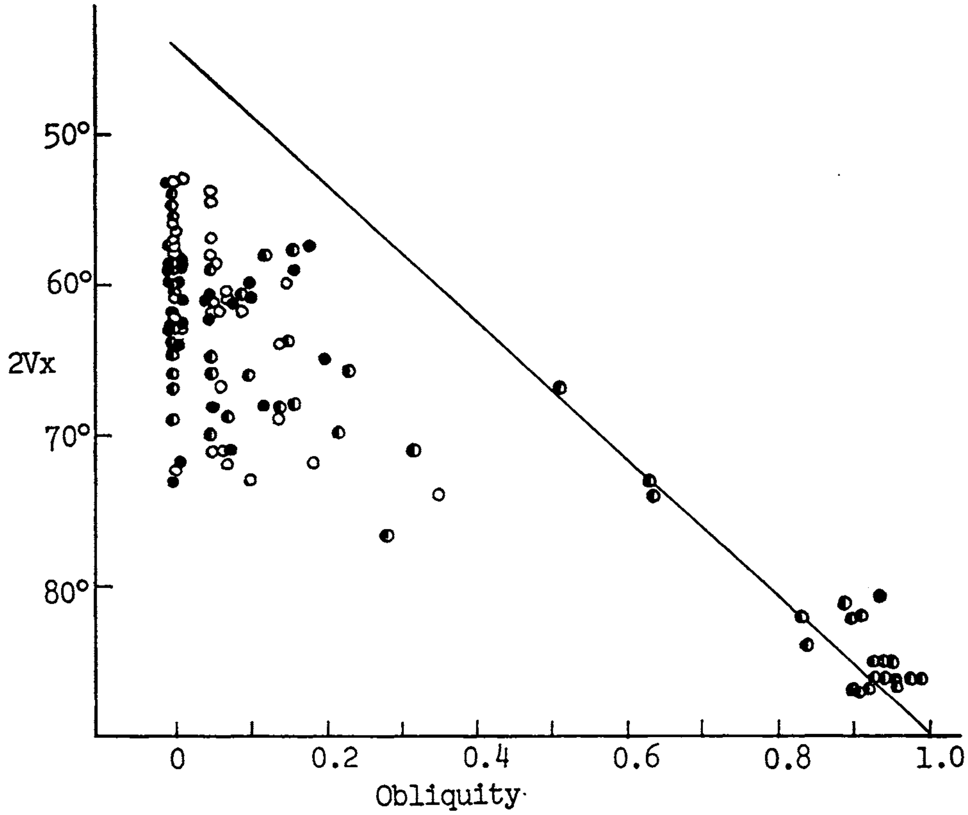

Fig. 9. Relation between optical angle and obliquity of K-feldspars. Data from Murakami *et al.* (1964) and Murakami (1971a; 1971b), including unpublished information. Symbols are the same as in Fig. 2.

3. *Al-content in hornblendes*

The hornblendes contained in the granitic rocks range widely both in Fe/Mg ratio and in Al-content which tends to vary systematically among the three zones. Most aluminous hornblendes (8–12% Al_2O_3) occur in the Ryoke zone (Fig. 10). On the other hand, all of the granites in the Sanin zone contain Al-poor hornblendes (3–6% Al_2O), as do some of the granites of the Hiroshima zone. The hornblendes in the Ryoke zone are rich in tetrahedral Al (Fig. 11), but there is no detectable difference in octahe-

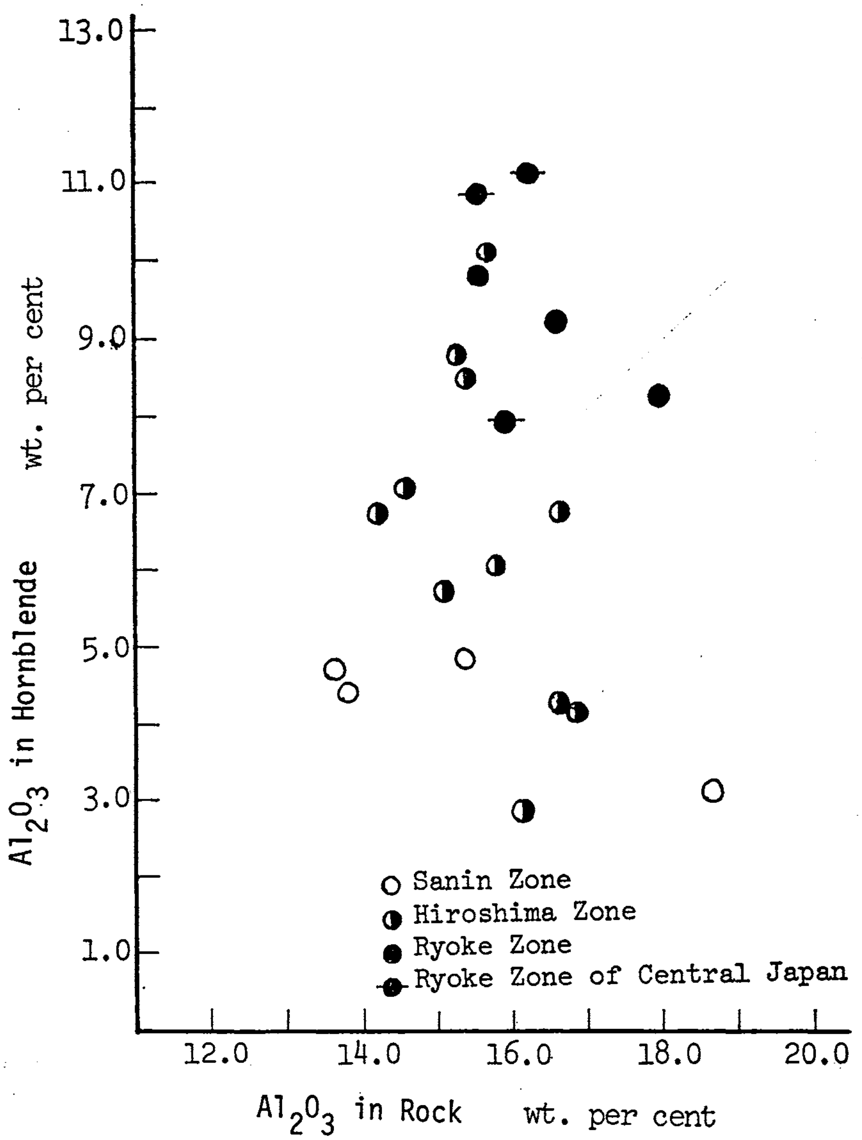

Fig. 10. Relation between Al_2O_3-content of whole rocks and that of constituent hornblendes. Data from Murakami (1969) and Shibata *et al.* (1966), including unpublished information. Data from the Ryoke zone of central Japan are also plotted for comparison.

dral Al among the three zones. The Al-content of hornblendes is generally considered to be affected by both formative conditions and whole rock chemistry (Leake, 1965; Raase, 1974). Yet no relationship can be discerned between the Al-content and the whole rock chemistry in the Sanin zone where the hornblendes are always poor in Al even in the aluminous rocks, nor in the Ryoke granites where hornblendes are poor in Al even in the silica-rich rocks. These facts conflict with the ordinary variation tendency

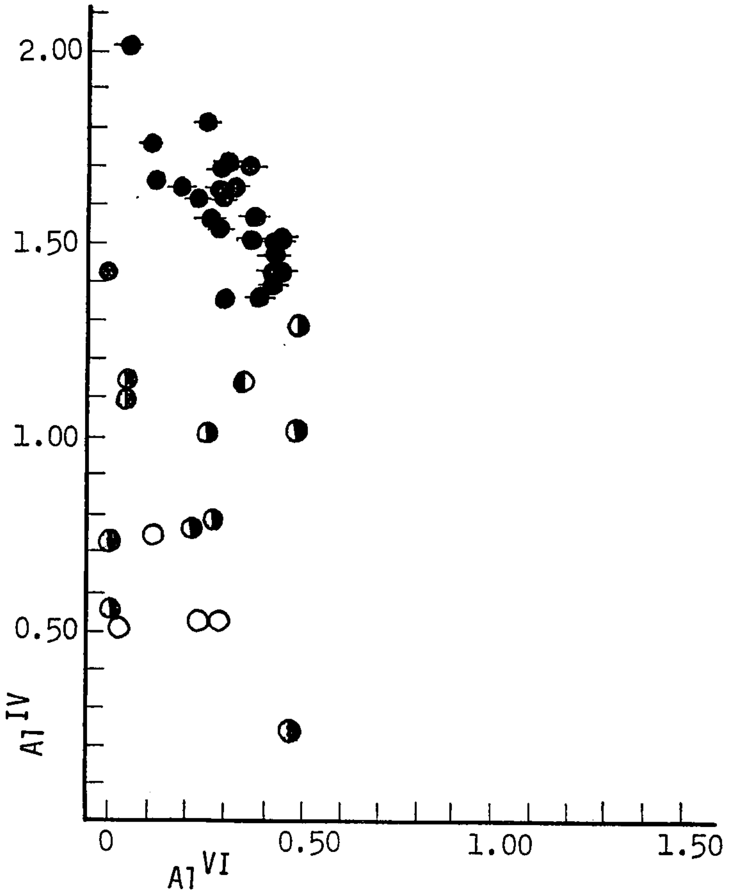

Fig. 11. Relation between Al^{IV} and Al^{VI} of hornblendes. Data from Murakami (1969), Shibata *et al.* (1966) and Kanisawa (1975), including unpublished information. Symbols are the same as in Fig. 10.

of the composition of hornblendes in which a decrease of Al^{IV} is usually accompa- a nied by an increase in the Fe/Mg ratio as differentiation progresses in granitic magm (Nockolds, 1947). On the contrary, the hornblendes investigated show a reverse tendency, as seen in Figure 12 where the Al^{IV}-content of hornblendes increases with increasing Fe/Mg ratio. This seem to suggest the intimate connection of Al^{IV} with oxygen partial pressure during the formation of hornblendes.

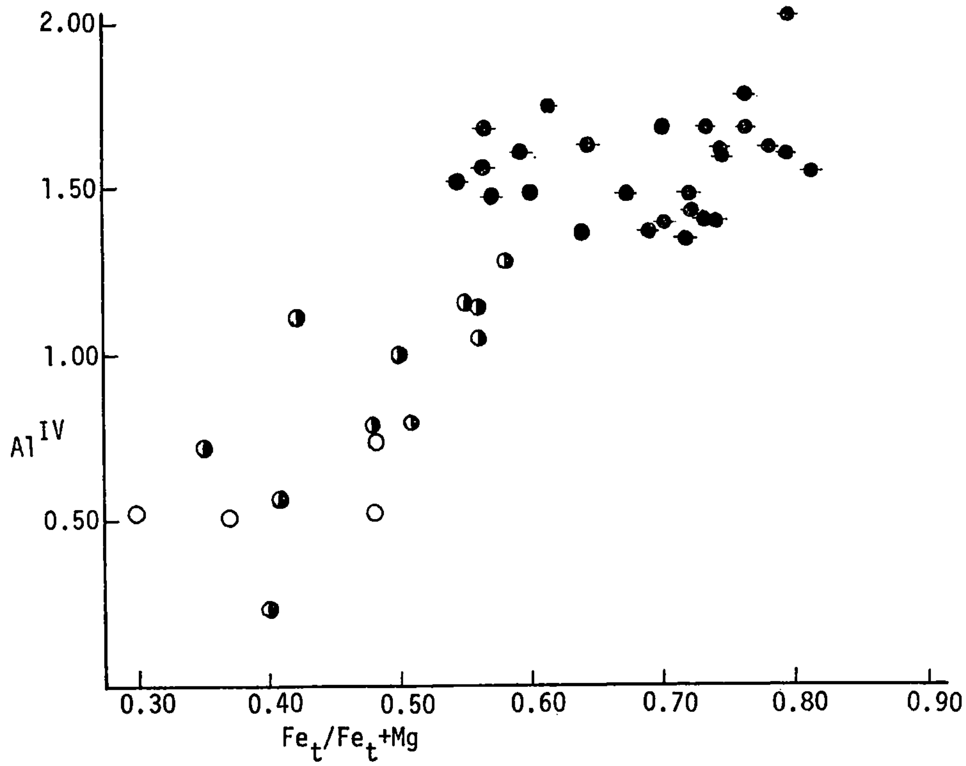

Fig. 12. Relation between Al^{IV} and Fe_t/Fe_t+Mg of hornblendes. Data from Murakami (1969), Shibata *et al.* (1966) and Kanisawa (1975), including unpublished information. Symbols are the same as in Fig. 10.

In this respect, it is noted that the Ryoke granites* ranging from 67 to 73 per cent in SiO_2 are commonly devoid of hornblende except for the aluminous (and calciferous) rocks (Fig. 13). This may account for the common occurrence of the aluminous hornblendes in the Ryoke granites. The Al-poor hornblendes are thought to become unstable with decreasing Al_2O_3 and CaO, and with increasing SiO_2 of whole rocks. This feature is not recognized in the Hiroshima granites nor in the Sanin granites.

SUMMARY AND DISCUSSION

The late Mesozoic to early Tertiary granitic rocks of Southwest Japan show some systematic variations of the chemical compositions of whole rocks and of constituent minerals, as summarized below.

1) The Fe_2O_3/FeO ratio of whole rocks increases from south to north, which suggests the northward increase of oxygen partial pressure during the formation of granitic magma. This tendency is reflected also in the constituent biotites and hornblendes, especially their Fe/Mg ratio which decreases from the Ryoke zone to the Sanin zone. A similar result was obtained in the central Sierra Nevada Batholith (Dodge, 1973).

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*In these granites relic clinopyroxenes are often rimmed by biotites without hornblendes (see Fig. 5).

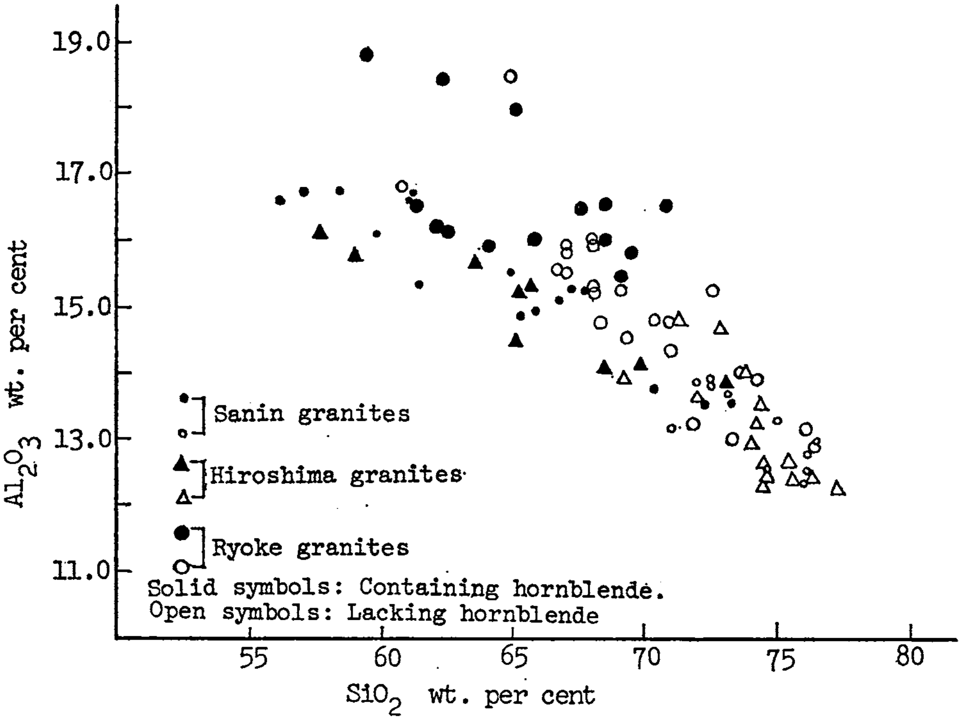

Fig. 13. Compositional relation between hornblende-bearing rocks and hornblende-lacking rocks among three granite zones. Data from Murakami (1969). including unpublished information.

2) The bulk chemical composition of K–feldspars is most potassic in the Ryoke zone, and most sodic in the Sanin zone. The evolutionary trend of the K–feldspars in the Sanin zone resembles that of typical hypersolvus granite, whereas the Hiroshima and the Ryoke granites are characterized by the subsolvus type trend, The three zones differ also in optical angle and obliquity of K–feldspars. Compared with the Sanin granites, the Hiroshima and the Ryoke granites are distinguished by the wide range of optical angle and obliquity of the K–feldspars, and by the presence of maximum to near-maximum microcline.

3) The tetrahedral Al-content of the hornblendes decreases from south to north with decreasing Fe/Mg ratio. It is highest in the Ryoke zone and lowest in the Sanin zone with some exceptions. No systematic variation of the octahedral Al-content is found among the three zones. There is an important difference in the occurrence of hornblendes among the three zones. In the range 67-73% SiO_2, the Ryoke granites usually lack hornblendes where they are low in Al_2O_3 and CaO. On the other hand, in the Hiroshima and the Sanin zones the biotites coexist with hornblendes in the rocks with a wider range of SiO_2 than in the Ryoke zone.

Consequently, the Sanin granites seem to be characteristic of hypersolvus granites, whereas many of the Hiroshima and the Ryoke granites seem to be subsolvus granites. This is verified also by the occurrence of maximum to near-maximum microcline in the latter granites, because the microcline of high obliquity cannot be stabilized under the ordinary cooling conditions in the laboratory experiment (Akizuki, 1973). This suggests that maximum to near-maximum microcline was formed by recrystallization. The Sanin granites, therefore, may have been emplaced under higher temperatures and less cover than the Hiroshima and the Ryoke granites.

The difference among the three zones in oxygen partial pressure and Al-content of the constituent hornblendes also appears to be related to the depth of emplacement of the granites. The difference in oxygen partial pressure may be accounted for by the dissociation of water (Osborn, 1959;1962), by commingling of magma with connate ground water (Taylor, 1971), or by reduction caused by the contamination of magma by carbonaceous sediments (Ishihara and Kanaya, 1976). Of these-the possibility of co-mingling connate ground water was discounted by Ishihara and Matsuhisa (1975) on the basis of their oxygen isotope study, although further detailed investigation is needed. The paucity of data prevents choosing between the other two interpretations. However, both seem to be related to depth of emplacement. The dissociation of water is accelerated at shallower levels because the leaching of hydrogen increases along fissures or fault planes with upward migration of the granitic magma. This accounts for the higher oxygen partial pressure in the Sanin zone. On the other hand, the Ryoke zone is characterized by broad migmatic regions accompanied by amphibolite-facies metamorphic rocks. As indicated by Murakami (1974), this feature of the Ryoke zone is in contrast with the other two zones which are devoid of migmatic rocks. Then, the reduction process might have been more effective in the Ryoke zone than in the other two zones. Dodge (1973), in his investigation of the Sierra Nevada Batholith, attributed the different oxygen partial pressure to the difference in the H_2O-content of the original materials from which the granitic magma was derived, but confirmative data is lacking for the rocks presently under consideration.

In ordinary magmatic differentiation of granites, the constituent hornblendes will become poor in Al^{IV}, accompanied by high Fe/Mg ratio, as the whole-rock SiO_2 increases, but the opposite is true for the hornblendes under consideration. This anomaly may be accounted for by assuming that recrystallization took place under low-oxygen partial pressure in the Ryoke zone, as well as in a large part of the Hiroshima zone, because the granites of these two zones are possibly subsolvus. Under low-oxygen partial pressure, silica activity will be lowered as a result of limited formation of opaque minerals. This will affect the Al-content of hornblendes. Moreover, if the water vapour pressure is fairly high, biotite will replace the hornblende except that in aluminous and calciferous rocks. These processes would have been most effective in the Ryoke zone where the oxygen partial pressure was lowest and recrystallization is thought to have progressed under lowest temperature and slowest cooling conditions. In the Hiroshima zone, the oxygen partial pressure and the depth of emplacement of granites are intermediate. Consequently, the consituent hornblendes are also intermediate in composition. The Sanin zone is distinguished by the higest oxygen partial pressure and the shallowest emplacement of the granites. Therefore, the constituent hornblendes are possibly of magmatic origin. They are lowest in Fe/Mg ratio and Al^{IV}-content. Some of the hornblendes may be the relict minerals through rapid cooling, because they are frequently rimmed by biotite, The above conclusion is supported by the isotopic dating data (Fig. 14). If the isotopic mineral age marks the uplift and erosional unroofing of the pluton (Murakami, 1974), and the consolidation of magma is roughly dated by the whole rock age, the interval between the two ages must be larger in the older rocks than in the younger ones, assuming that the rate of uplift was nearly equal after consolidation. As seen in Figure 12, this interval is largest in the Ryoke granites and smal-

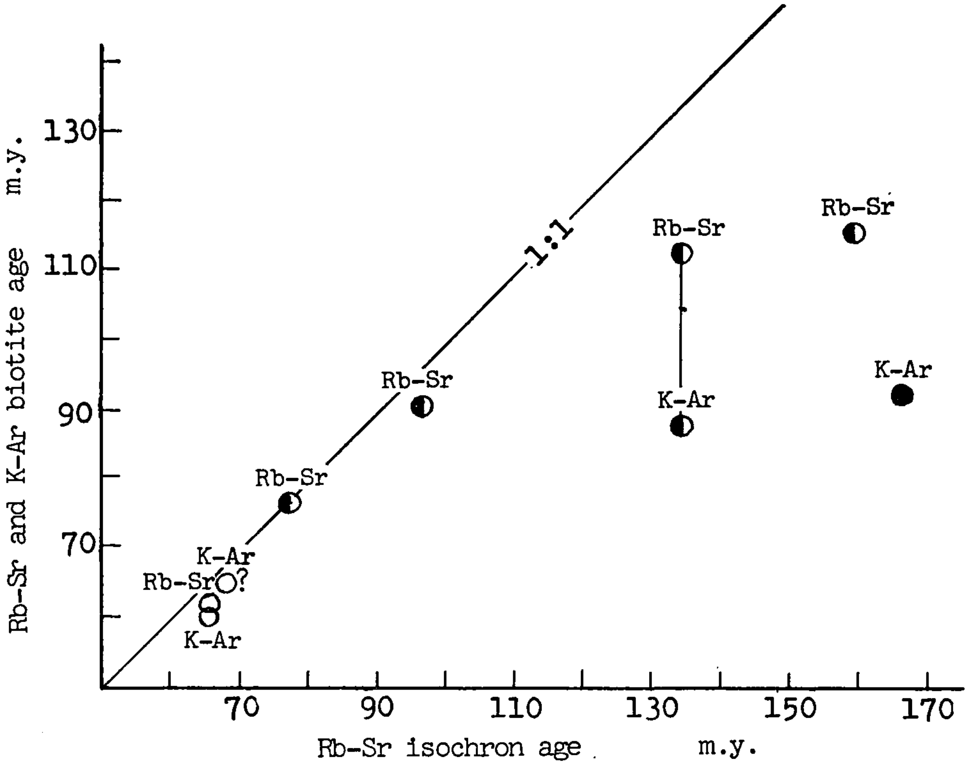

Fig. 14. Relation between mineral age and whole rock or whole rock-mineral isochron age of the granitic rocks of Southwest Japan. Two data on volcanic rocks are also plotted. Data from Hattori and Shibata (1974), Ishizaka (1971), Seki and Hayase (1974) and Yanagi (1975). Symbols are the same as in Fig. 2.

lest in the Sanin granites. As noticed by Hattori and Shibata (1974), the Sanin granites always show concordant or near-concordant K-Ar and Rb-Sr ages. This suggests fairly shallow emplacement of the granites, subject to rapid cooling. On the other hand, the Ryoke granites consolidated at the deepest level the three zones of granites under slow cooling condition and probably recrystallized.

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