Econ. Environ. Geol.
Vol. 27, No. 5, p. 459-467, 1994.

Petrochemical Study of the Gadaeri Granite in Ulsan Area, Kyeongsang Province

Choi, Seon-Gyu* and Wee, Soo-Meen**

ABSTRACT: The Gadaeri granite near Ulsan mine is an oval-shape isolated granitic body, and is genetically related to the iron-tungsten mineralization. The Gadaeri granite exhibits calc-alkaline and I-type characteristics, and generally shows the micrographic texture which indicates the shallow depth of emplacement. Consideration of the stratigraphic thickness of Ulsan formation and minimum-melt compositions suggests that the bulk magma crystallized at pressure of 0.5~2.0 kbar under water saturated condition. The evolutionary trend observed in the studied rocks represents that feldspar fractional crystallization has been a major magmatic process at the Gadaeri granite pluton. Different chemical characteristics between the Gadaeri and the Masan-Kimhae granites cannot be explained by fractional crystallization or different degrees of partial melting, and it reflects that the magma source for Gadaeri granite was different from that of the Masan and Kimhae granites.

INTRODUCTION

The Mesozoic granitic rocks and associated volcanics in South Korea have been studied in considerable detail by many previous workers (Won, 1968; Cha, 1976; Jin, 1980, 1985, 1988; Choo and Kim, 1986; Hong, 1987; Lee, 1991; Lee, 1992) who have been concerned with the detailed geologic mapping, tectonic and structural setting, and the petrographic and geochemical variations. On the basis of these studies, Korean granitic rocks are mainly divided into two main groups and they show distinct differences in the source material and the regional distribution. The Daebo granitic rocks (early Jurassic to early Cretaceous) were originated from partial melting of crustal materials and occur throughout the major geotectonic provinces except the Kyeongsang basin. The Bulgugsa granitic rocks (late Cretaceous to early Tertiary) were generated from igneous protolith at lower crust or upper mantle and they crop out mainly in the Kyeongsang basin (Jin, 1980; Hong, 1987).

The Gadaeri granitic pluton located in the southeastern part of Kyeongsang basin, is an oval-shape isolated granitic body about 6 km wide and 10 km long, and yielded the K-Ar biotite age of 58 Ma (Lee and Ueda, 1977). This investigation is mainly focused on the Gadaeri granitic rocks in the Ulsan area.

*Dept. of Geology, Korea Univ., Seoul 136-701, Korea
**Dept. of Earth Sciences, Korea Nat. Univ. of Education, Chungbuk 363-792, Korea
The present studies were supported by the Basic Science Research Institute Program, Ministry of Education, 1993, Project no. BSRI-'93-511.

Many studies have been done to clarify petrological and geochemical characteristics of the granitic rocks in the southern part of the Kyeongsang basin (Cha, 1976; Jin, 1985, 1988; Lee, 1991, 1992; Yang and Kim, 1993). However, the previous studies of the Gadaeri granite are few (Lee and Ueda, 1977; Choi and Imai, 1983) and cannot be used to evaluate the petrochemical variations within the pluton, and petrogenetic relationships between the Gadaeri granite and the other Tertiary granites such as Masan and Kimhae granites in the Kyeongsang basin remain questionable. The iron deposits in southeastern Korea have been considered to genetically intimate relation to the micrographic and granophyric granites, such as Masan and Kimhae granites. The Gadaeri granites and the Masan-Kimhae granites show the micrographic texture, and they have the characteristics indicating shallow depth emplacement and are commonly associated with iron deposits. Thus, the purposes of this study are to evaluate the petrochemistry of the Gadaeri granite and to constrain the genetic relationships between the studied rocks and the Tertiary granitic rocks, which are associated with the iron mineralization, from the other part of the Kyeongsang basin.

GEOLOGIC SETTING

Regionally the study area occupies the southeastern part of the Kyeongsang basin, where the Cretaceous molasse-type sediments are intercalated with intermediate to acidic volcanics. Also, volcano-plutonic complexes of granitic composition, which belong to the Bulgugsa granite series in a broad sense ranging from Cre-

Table 1. Modal composition of Gadaeri granite

Specimen no.	902	907	910	810207	810310	810312	810313	810314	810315
quartz	29.6	26.6	31.1	26.1	32.9	26.0	27.9	29.7	28.2
alkali feldspar	45.0	42.4	50.8	36.7	45.0	52.6	41.5	40.8	42.8
plagioclase	22.9	27.7	14.9	29.2	18.1	18.3	28.3	23.8	23.2
amphibole	0.5	0.4	0.4	1.4	0.7	0.6	0.1	1.6	0.5
biotite	1.0	1.2	2.0	2.9	2.3	0.7	1.3	2.0	2.5
chlorite	0.1	0.7	0.1	2.2	0.5	0.4	0.2	1.2	1.5
clinopyroxene	-	-	-	-	-	0.4	-	-	-
sphene	0.1	0.1	tr	0.2	0.1	0.2	0.1	0.1	0.2
apatite	tr	tr	tr	0.1	tr	tr	tr	0.1	0.1
opaque mineral	0.8	0.9	0.7	1.2	0.4	0.8	0.6	0.7	1.0

taceous to early Tertiary in age, crop out extensively within this sedimentary basin. General geology of the study area has been described in detail by Park and Yoon (1968), Choi et al. (1980), and Choi and Imai (1983). Thus, a brief description of the rock units is given in this section. The rocks of the area consist mainly of sedimentary rocks of Ulsan formation which is intruded and mantled by Chisulryeong and Yucheon volcanic complex and granitic rocks(Fig. 1).

Ulsan formation has been subdivided into three formations on the basis of the lithostratigraphic sequence and sedimentary environments by Choi et al. (1980); Guyeongri formation, Sayeonri formation and Joilri formation. They consist mainly of reddish and grey to greyish green shale and sandstone, and tuffaceous sandstone and thin conglomerate. The Ulsan formation is intruded by intrusives and covered by extrusives and thermally metamorphosed owing to the Tertiary granitic intrusions. The Chisulryeong volcanic complex consists of welded tuff, lapilli tuff, and rhyolite. It intruded into the Ulsan formation and is in turn intruded by hornblende biotite granite. The Yucheon volcanic complex, which is intruded by hornblende biotite granite, consists of rhyolite, tuffaceous agglomerates, and volcanic breccias. The rhyolite shows the flow texture and porphyritic texture with plagioclase phenocrysts.

The Bulgugsa granites are widely exposed in the western and northern parts of the Ulsan area. These granites intrude a sequence of gently dipping Ulsan formation and volcanic complex. The granitic intrusive, called the Gadaeri granite, is exposed widely in the western part of the Ulsan mine, and yielded the K-Ar biotite age of 58 Ma (Lee and Ueda, 1977). The Gadaeri granitic body is an oval-shaped in map view, with dimension of about 6 by 10 km.

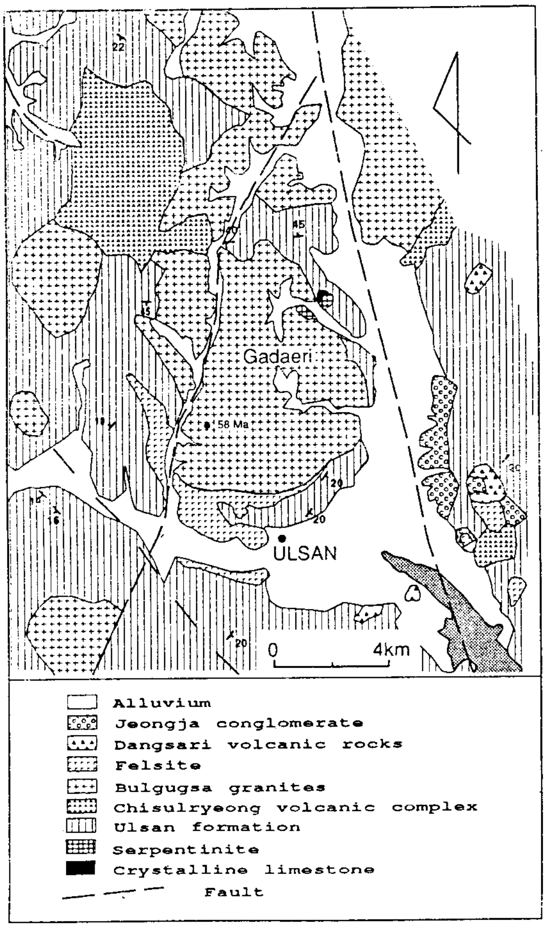

Fig. 1. General geologic map of the Ulsan area modified from Park and Yoon (1968)

PETROGRAPHY

The Gadaeri granite pluton is nearly equigranular in the center of the stock and porphyritic and granophyric toward the periphery, particularly near the Ulsan mine. The plutonic body mainly consists of a fine to medium grained porphyritic hornblende-biotite granite. This granite is composed of K-feldspar, quartz, plagioclase

Table 2. Whole rock chemical compositions of Gadaeri granites

Specimen no.	902	907	910	810207	810310	810312	810313	810314	810315	925
SiO_2	75.78	74.64	73.92	71.50	75.20	74.84	74.23	72.37	73.07	72.13
TiO_2	0.38	0.35	0.25	0.55	0.18	0.36	0.31	0.38	0.31	0.36
Al_2O_3	14.41	14.26	13.43	13.35	12.59	14.03	14.16	13.65	13.95	14.34
FeO^*	1.23	2.34	1.13	3.39	0.91	1.74	1.93	2.07	2.24	2.13
MnO	0.01	0.06	0.06	0.10	0.05	0.07	0.05	0.11	0.05	0.08
MgO	0.15	0.47	0.17	0.61	0.15	0.33	0.35	0.44	0.33	0.43
CaO	0.08	0.88	0.60	1.08	0.82	1.34	1.08	1.37	1.02	0.72
Na_2O	3.25	3.69	3.56	3.83	3.43	4.00	3.83	3.84	4.11	4.63
K_2O	3.91	3.96	4.60	3.75	4.83	4.34	4.01	3.95	3.90	4.00
P_2O_5	0.00	0.00	0.00	0.02	0.00	0.00	0.00	0.01	0.00	0.08
Total	99.20	100.65	97.72	98.18	98.16	101.05	99.95	98.19	98.98	98.90
Zr	50.62	23.46	11.37	22.67	26.68	46.82	20.00	27.78	64.21	117.17
Nb	16.82	15.14	16.46	9.48	14.08	13.86	14.56	14.38	13.22	17.47
Ba	389.99	690.44	677.52	1057.86	367.97	774.26	705.90	815.11	761.55	696.19
Rb	108.96	108.91	143.03	67.17	97.21	102.28	92.95	123.02	98.47	91.00
Sr	50.80	110.31	52.99	84.43	50.24	85.02	106.85	106.99	132.25	80.00
Y	18.16	31.86	38.32	23.36	10.90	24.12	22.98	41.68	28.38	33.58
Cr	1.90	1.68	2.04	3.20	3.00	1.85	1.83	2.16	2.13	2.00
Co	0.48	37.63	36.31	132.51	108.48	65.31	61.75	64.63	98.99	24.00
Ni	1.22	0.74	17.05	1.52	0.91	0.90	0.95	1.03	1.13	1.00
La	56.00	54.47	55.71	22.57	13.42	22.19	23.47	49.22	30.19	42.49
Ce	100.97	86.35	66.70	41.98	41.02	33.70	49.32	91.61	51.95	84.22
Nd	51.70	50.27	53.87	26.35	17.98	25.33	29.47	44.38	30.68	41.41
Sm	9.47	8.88	10.31	5.21	3.30	5.04	5.50	8.37	6.02	7.89
Eu	1.57	1.49	1.22	0.92	0.42	0.77	0.93	1.39	1.04	1.16
Gd	7.56	7.91	9.37	4.94	3.09	4.87	5.25	7.79	5.88	7.08
Tb	0.85	1.11	1.38	0.75	0.49	0.79	0.81	1.18	0.87	1.07
Dy	4.92	6.52	8.74	4.77	3.16	4.75	5.26	7.09	5.38	6.56
Yb	1.81	3.35	4.73	3.25	2.02	3.39	3.06	4.17	3.27	4.38
Lu	0.28	0.52	0.65	0.44	0.28	0.46	0.42	0.55	0.47	0.67
Hf	1.53	0.81	0.54	0.79	0.98	1.53	0.81	1.00	1.81	4.59
Pb	4.15	13.06	15.72	14.76	14.90	13.73	16.21	19.21	11.14	24.18
Th	9.98	10.79	11.08	5.95	11.74	7.81	11.32	11.18	9.64	11.15
Cu	1.28	2.43	1.07	7.92	2.74	5.15	7.11	4.54	3.46	16.00

FeO^* represent total iron

and minor amounts of biotite, hornblende, ilmenite, magnetite, hematite, sphene, and apatite. It shows a variety of porphyritic texture characterized by strong development of micrographic and/or granophyric intergrowth. The modal compositions of the granite are listed in Table 1. Small-scale variation in the relative abundances of alkali feldspar, quartz, and plagioclase might be due to the textural variation such as porphyritic texture and micrographic and/or granophyric texture.

Quartz makes up 26 to 32.9% of the mode, and commonly forms a micrographic texture with potassium feldspar. Potassium feldspar is the most dominant mineral phase and it comprises from 36.7 to 52.6 % of the rock. The potassium feldspar grains often show perthitic texture. Plagioclase is subordinate to potassium feldspar and generally occurs as phenocryst which comprises 14.9 to 29.2% of the rock. Plagioclase composition is near An_{25} and slightly zoned. The compositional zoning of plagioclase phenocrysts shows calcic cores (An_{30}) to sodic rims (An_{20}). Most plagioclase grains are altered to sericite, and are often partially replaced by chlorite. Also, the radiate intergrowths of quartz and alkali feldspar are arranged along euhedral plagioclase phenocryst (Fig. 2). Biotite occurs as a principal mafic mineral, and is reddish brown to dark red in color. It generally forms less than 3% of the rock, and is associated with small amounts of hornblende, magnetite, ilmenite, hematite, apatite and rarely sphene. Most biotite grains are partially altered or rimmed to chlorite. Hornblende is greenish to light brown in color and occurs as euhedral to subhedral form and it makes up less than 2% of the rock, and most of them are altered to chlorite and sericite. In the classification scheme of Streckeisen (1967), the rocks belong to granite in a broad scale but range from granite

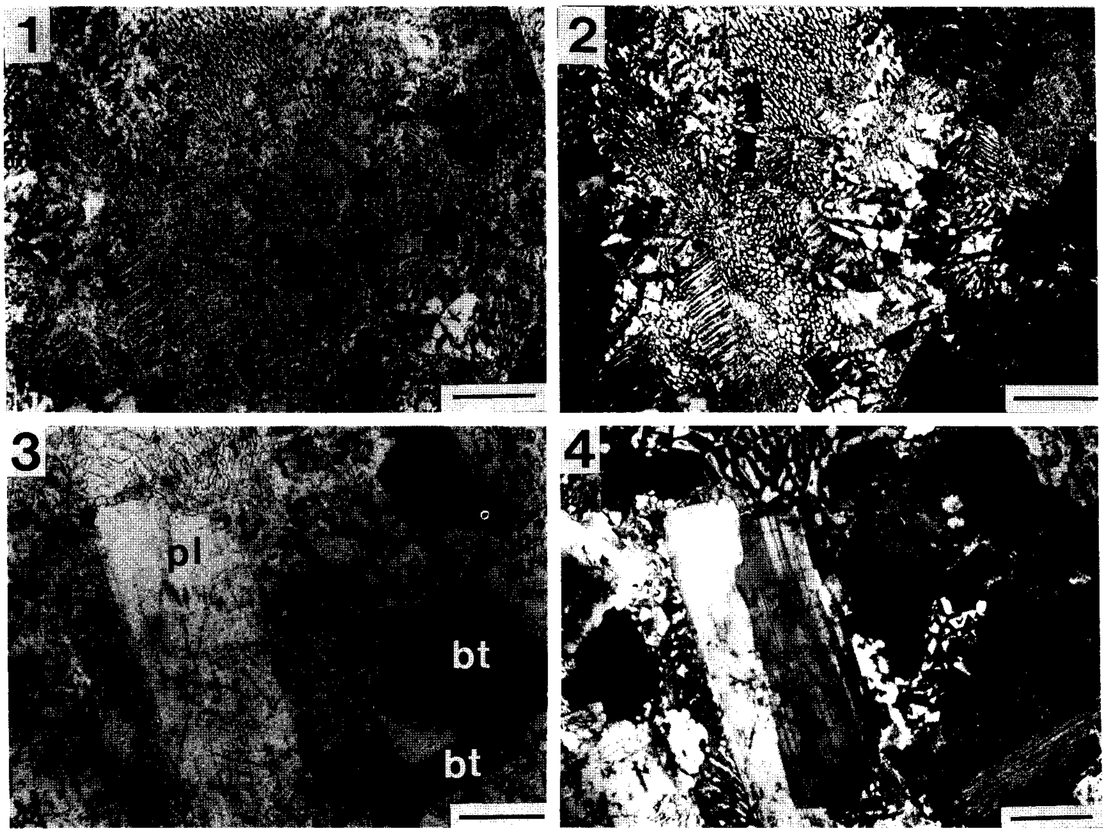

Fig. 2. Photomicrographs showing porphyritic, micrographic, and granophyric texture in Gadaeri granite. 1. Plane polarized light, 2. Ditto, crossed polars, Specimen US902, 3. Plane polarized light, 4. Ditto, crossed polars, Specimen US810310, Bar scale represents 0.2 mm in length.

to adamellite in a narrow sense (Fig. 3).

GEOCHEMISTRY

Sampling and Analytical Technique

Ten whole rock samples were analyzed for major, trace and rare earth elements (REE). Sample powders for the analysis were prepared by grinding in a tungsten carbide shatter box. Most of the samples are fresh and altered parts were avoided. The whole rock data are listed in Table 2. Philips (PW 1480) X-ray fluorescence spectrometer was used to determine major element composition of the whole rock. REE and selected trace elements were analyzed by Inductively Coupled Plasma mass spectrometer (VG Elemental-PQ II Plus) at Korea Basic Science Center. Among the samples, specimens 902, 907, 925, and 910 were obtained from the outcrops of the Gadaeri granite, and specimens 810207, 810310, 810312, 810313, 810314, and 810315 were obtained from the drill cores of boreholes US 81-2 and US 81-3 near the Ulsan mine, which were drilled by Korean Mining Promotion Co.

Major Element

The Gadaeri granite has a restricted silica range with SiO_2 contents of 71 to 76 wt. % (Table 2). Variations of major elements with increasing differentiation are indexed with weight percent of SiO_2 and are shown in Fig. 4. The Bulgugsa hornblende biotite granites from other localities (Masan and Kimhae hornblende biotite granites; data from Lee, 1991) were also plotted for comparison. Harker plots for major element of the Gadaeri, Masan, and Kimhae granites show relatively clear correlations between major element concentrations and silica except Na_2O and MnO. The abundance of K_2O increase with increasing differentiation index, while Al_2O_3, FeO, CaO, TiO_2, and MgO decrease regardless of the sample loca-

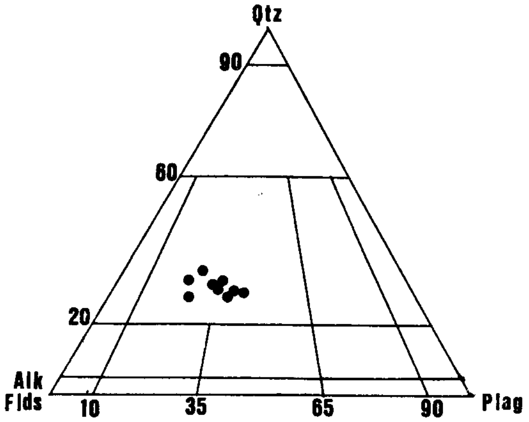

Fig. 3. Triangular diagram of modal quartz-plagioclase-alkali feldspar of the granitic rocks.

tions. Based on Fig. 5, the overall trend of the rocks corresponds to that of calc-alkaline series, and the Gadaeri granite shows more differentiated characteristics. All samples have high Al_2O_3 (>13.4 wt. % except sample 810310) and normative corundum indicating a high degree of peraluminosity.

The genetic type of granite can be classified by [Al/(Na+K+Ca)] ratio (Hine et al., 1978) and is shown in Fig. 6. All samples have less than 1.1 which is typical of I-type value. According to the White and Chappell (1983), I-type granites have greater than 3.2% of Na_2O and average mean value of Na_2O/K_2O ratio is about 0.8, while S-type granites have less than 0.6 in general. The Gadaeri granite has greater than 3.2 wt. % of Na_2O (3.25~4.63 wt. %) contents and Na_2O/K_2O ratios range from 0.73 to 1.15 with average mean value of 0.93. Based on above mentioned characteristics, the Gadaeri granite belongs to I-type granite.

Trace and Rare Earth Elements

Trace and rare earth element analytical results are listed in Table 2. The conventional Harker variation diagrams for trace elements for the Gadaeri, Masan, and Kimhae granites are plotted in Fig. 7 in order to constrain the genetic relationship among the rock groups. Variation patterns of the trace elements show a simple differentiation trends except Y and Rb which show slightly dispersed patterns. The Gadaeri granites show higher Rb/Sr ratios (0.8) compared with the others and might be caused by either fractionation of the Rb-poor and Sr-enriched plagioclase or plagioclase may not be involved in the melt phase retaining at the unmelted restite during low degree of partial melting.

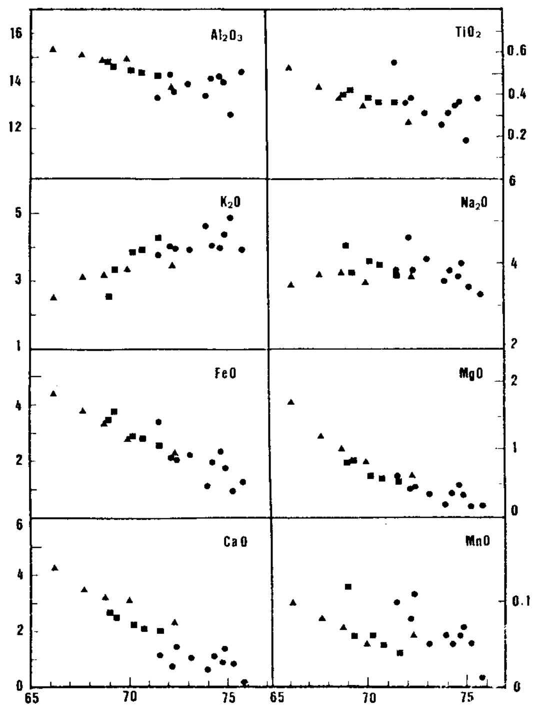

Fig. 4. Variation of major elements vs. silica. Total Fe is reported as FeO. Solid circles are Gadaeri granites, solid Triangles are Masan granites and solid squares are Kimhae granites.

Rare earth element concentrations of the rocks are characterized by LREE enrichments, which become more pronounced with increasing differentiation, consistent with fractionation of the observed phenocrysts phase assemblages for which distribution coefficient of the REE increases from La to Lu (Fig. 8).

RESULT AND DISCUSSION

The chemical features of the Gadaeri granite represent the calc-alkaline characteristics and show the hypabyssal, shallow seated, rapid cooled plutonic nature based on their petrographic evidences. The micrographic texture is usually found in highly evolved granitic rocks which have nearly eutectic compositions. The formation mechanism of the micrographic granites can be explained by the two-stage decompression model governing the degree of undercooling and can be increased by the exsolution or degassing of vapor phases along the weak zones such as faults or fractures (Lee, 1991). CIPW-normative compositions of the Gadaeri granite plotted in the ternary system demonstrate potential constraints

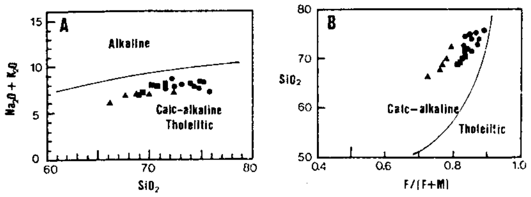

Fig. 5. A) Alkali vs. Silica diagram for the Gadaeri, Masan and Kimhae granites. B) Silica vs. FeO*/(FeO*+MgO) diagram for the Gadaeri, Masan, and Kimhae granites. Symboles are same as in Fig. 4.

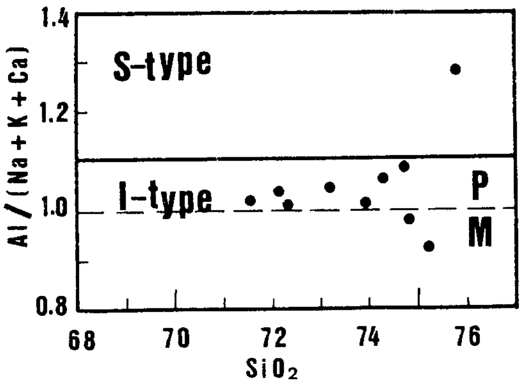

Fig. 6. Molar Al/(Na+K+Ca) vs. Silica variation diagram. I and S-type boundary is based on Hine et al. (1978). P: Peraluminous, M: Metaluminous

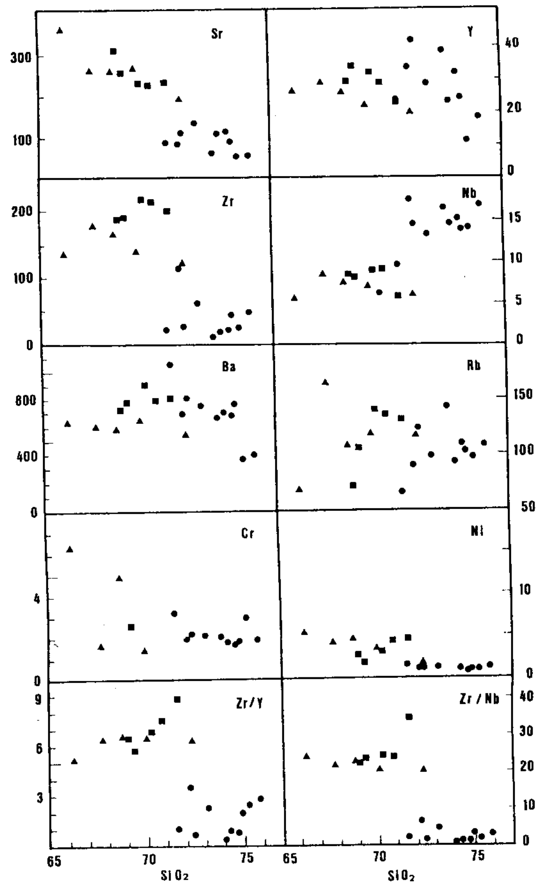

Fig. 7. Variation of trace element vs. Silica. Symbols are same as in Fig. 3.

from experimental data on pressure and fluid activity during granitic magma genesis (Fig. 9). The ternary plots are useful petrological tool for estimating the conditions of melt formation for granite composition with greater than 90% felsic components (Inger and Harris, 1993). The water-saturated ternary minimum points at various pressures and $P_{H_2O}=0.5$, $P_{H_2O}=0.3$ points (Winker, 1979; Ebadi and Johannes, 1991) are plotted for reference. The Gadaeri granite samples cluster around a composition corresponding to minimum melt at 2 to 0.5 kbar under water-saturated conditions. The Ulsan formation near the Gadaeri granite is equivalent to its maximum stratigraphic thickness of about 4,000 m (Choi et al., 1980) without accounting the pre-date erosional processes. The overburden thickness corresponds to maximum pressure at about 1.2 kb. Considering above reasons, it is thought that the pressure at the time of the final crystallization of bulk magma was about 0.5 to 2.0 kbar under water saturated conditions. The pressure condition of the Gadaeri granite is lower than that of the granite from the southern part of the Kyeongsang basin, which indicated the pressure of 1.2 to 2.6 kbars by the silicate melt inclusion study (Yang and Kim, 1993).

Several problems arise during fractional crystallization in granitic magma system. Most models to develop to explain trace element changes during fractional crystallization of intrusive assume complete separation of crystals and liquid. Feldspar fractionation in acidic magma is impeded by the lack of density contrast between the solid phase and the melt and the high viscosity of granitic liquids. However, Martin and Nokes (1989) and Koyaguchi et al. (1990) demonstrated experimentally that crystal fractionation is still possible in granitic magma system by settling out from the boundary layer of convection cells. More recently, Inger and Harris (1993) suggested that the crystal fractionation would be a efficient mechanism for differentiation of granite in case magma volume is small and country rock was probably at significantly lower temperature than melt. The volume of the Gadaeri granite source magma might be small due to the decreasing geothermal gradient of the crust (Lee,

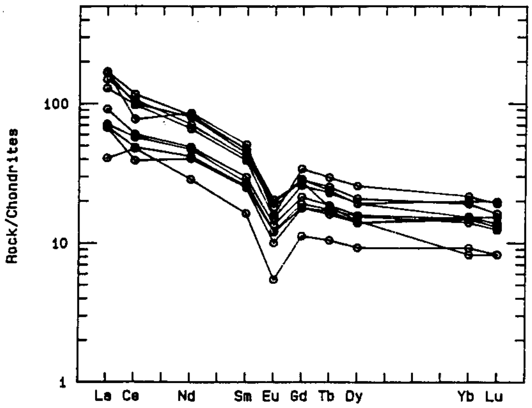

Fig. 8. Chondrite normalized REE patterns of the Gadaeri granites.

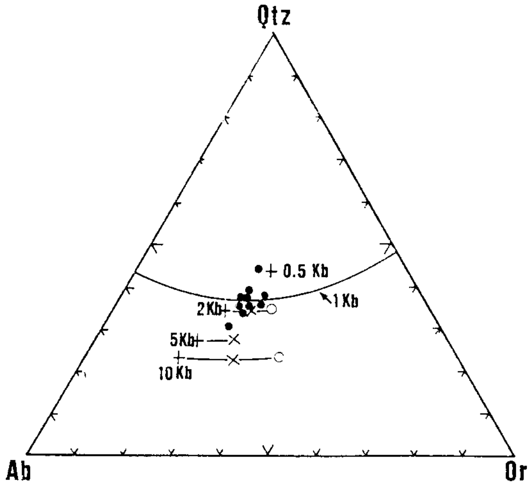

Fig. 9. Phase relations and minimum-melt compositions in the system Quartz-Albite-Orthoclase. CIPW normative compositions of Gadaeri granites are plotted as solid circles. Minimum melt compositions are from Winkler (1974), and Ebadi and Johannes (1991). +, $P_{H_2O}=1$; ×, $P_{H_2O}=0.5$; ○, $P_{H_2O}=0.3$

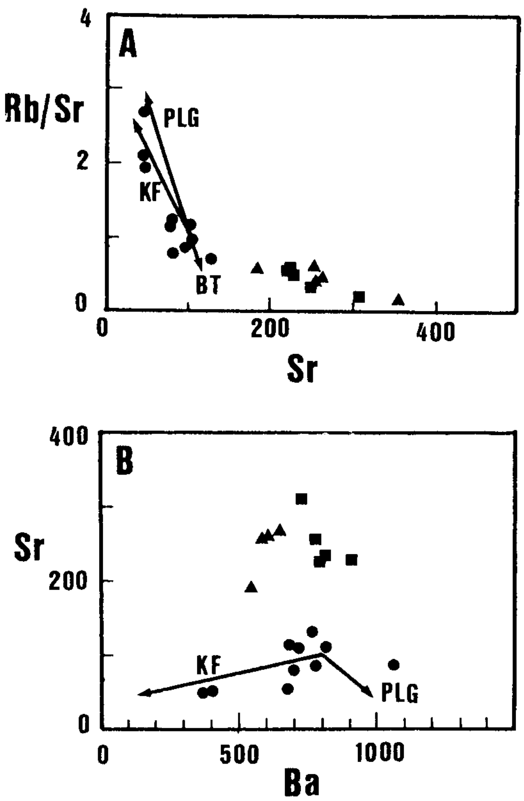

Fig. 10. Rb/Sr vs. Sr, and Sr vs. Ba plots. Bold vectors represent 10% crystallization of mineral phases.

1991) and the temperature difference between magma and wallrock might be large enough to cause efficient fractional crystallization. Considering aforementioned reasons, fractionation model can be applicable in the petrogenesis of the Gadaeri granite. The values of Rb, Ba and Sr as indicators of petrogenesis are illustrated in Fig. 10. Figure 10 demonstrates that the Gadaeri granite samples form a generally linear trend, particularly in the Rb/Sr vs. Sr plot. Fractionation vectors are shown for 10% fractional crystallization of the major phases. The primary trend clearly corresponds to removal of K-feldspar in the liquid. Thus, the evolutionary trend observed in the studied rocks represents that feldspar fractional crystallization has been a major magmatic process at the Gadaeri granitic pluton. Comparison of the geochemical characteristics of the studied rock samples with those of the Masan and Kimhae granites is needed in order to constrain the petrogeı esis of the granitic rocks in the Kyeongsang basin(Lee, 1991). As shown in the major element variation diagrams (Fig. 4), both the Gadaeri granites and the Masan-Kimhae granites represent well defined variation trend. These trend seems to be the magmatic differentiation trend which is controlled by fractional crystallization and indicates the Gadaeri granites are the most evolved phase. However, the evolution of the Gadaeri granites (high Rb/Sr ratio and low Sr content) can not be explained by simple fractional crystallization based on the Rb/Sr vs. Sr plot (Fig. 10) which shows different trends between the Gadaeri granite and the Masan-Kimhae granites. It seems that the difference of the chemical evolutionary trend between the granitic rocks may reflect differences in the parental magma which is mainly due to differences in the source regions or degree of partial melting. Figure 11 shows the variation in ratios K/Rb vs. Rb/Sr of the rocks and shows

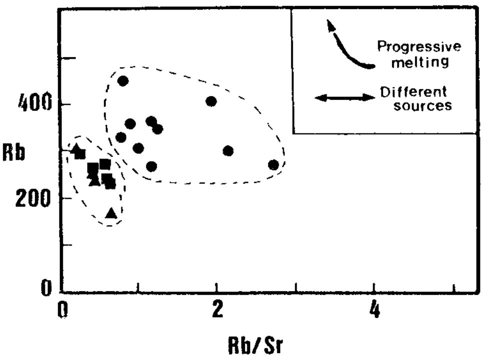

Fig. 11. K/Rb vs. Rb/Sr plot of the Gadaeri, Masan, and Kimhae granites. Symbols are same as in Fig. 3.

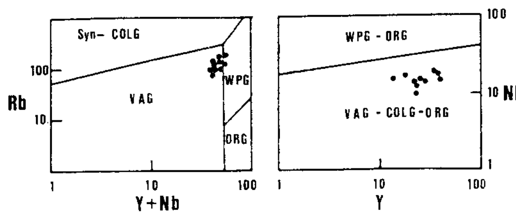

Fig. 12. Discrimination diagrams of Rb vs. Y+Nb, and Nb vs. Y for the Gadaeri granites (Pearce et al., 1984). COLG : collision granite, WPG : within plate granite, ORG : oceanic ridge granite. VAG: volcanic arc granite

trends that might result either from different degrees of partial melting of a fixed source composition or from the different source composition (after Strong and Hanmer, 1981). In Fig. 11, the Gadaeri granite and the Masan-Kimhae granites cluster in two separate groups. It represents that these rock groups were generated from different parental materials and the Gadaeri granites might not be the final products of fractional crystallization from same parental magma. In order to constrain the genetic environment of the Gadaeri granites, analyzed rocks were plotted on the Rb vs. (Y+Nb) and Nb vs. Y discrimination diagrams (Fig. 12). These rocks plot within volcanic arc granite field which is for subduction related magma suite formed along destructive plate margin.

CONCLUSIONS

Geochemical and petrographic data have provided useful information to constrain the chemical evolution of the Gadaeri granitic rocks. Geochemical and petrographical study of the rocks from the study area allow the following conclusions:

1. Geochemical data indicate that the Gadaeri granitic rocks have calc-alkaline and I-type characteristics. The rocks have high aluminum content and normative corundum indicating a high degree of peraluminosity.

2. The Gadaeri granitic rocks generally show the micrographic texture which represent the shallow depth emplacement. The granitic rocks are well plotted within 1 to 0.5 kbar ternary minimum point under water saturated conditions. Consideration of minimum-melt compositions suggests that the depth at the time of the crystallization of bulk magma was very shallow.

3. The evolutionary trend observed in the studied rocks represent that feldspar fractional crystallization has been a major magmatic process at the Gadaeri granitic pluton. Different chemical characteristics between the Gadaeri and the Masan-Kimhae granites can not be explained by fractional crystallization or different degrees of partial melting and it reflects that the magma source for Gadaeri granites was different from that of the Masan and Kimhae granites.

4. Based on all the geochemical characteristics and tectonic discrimination diagrams, the Gadaeri granites represent subduction related magma suite formed along destructive plate margin.

ACKNOWLEDGEMENT

Financial research grant for this study was provided by the Center for Mineral Resources Research sponsored by the Korea Science and Engineering Foundation which is greatly acknowledged. Helpful and constructive reviews by Prof. S.T. Kwon is greatly appreciated.

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Manuscript received 1994

경상남도 울산지역 가대리화강암에 대한 암석화학적 연구

최선규* · 위수민**

요 약: 울산지역에 분포하는 가대리 화강암체는 울산 철 · 텅그스텐 광화작용의 관계화성암으로서 전형적인 미문상조직을 보여주며, 칼크-알칼리 계열과 I-type화강암류의 지화학적 특성을 나타내고 있다. 가대리화강암은 물이 포화된 상태에서 0.5~2.0 kbar의 압력조건하에서 생성된 것으로 이는 천부에서의 분별결정에 기인된 것으로 화학성분의 분화는 대부분 알칼리 장석의 분별결정작용으로 이루어졌다. 경상분지 최남단에 분포하며 철광화작용과 관련된 미문상화강암체인 마산-김해 화강암체와 가대리화강암체는 미량성분의 화학조성을 비교해볼때 전혀다른 모마그마에서 분화되었음을 시사해준다.