Petrogenesis of Granitic Rocks in the Hisakajima Island, Goto Archipelago, Southwestern Japan: A Geochemical Study

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Abstract: Whole-rock chemical compositions including rare earth elements for the granitic rocks from the Hisakajima Island, Goto Archipelago, southwestern Japan were measured in order to constrain their origin and petrogenesis. The granites were divided into two types—a granodioritic group (GD) and a high Fe/Mg ratio granitic group (HFG). The granitic magma was formed by the upwelling of high-temperature mantle material, which might be related to the extension of the Japan Sea around the Middle Miocene. The origin of the GD magma was attributed to the mantle material, while the origin of the HFG magma was attributed to partial melting of the crust by upwelling of the high-temperature mantle. The amount of rare earth elements revealed the secondary addition of light rare earth elements through hydrothermal processes for the granites. Chondrite normalized rare earth element patterns revealed that the HFG rocks were not well differentiated.

Keywords: granite; Goto Islands; southwestern Japan; rare earth elements

1. Introduction

The Japanese island arc was separated from the Asian continent during the middle Miocene, with Northeast Japan rotating counterclockwise (CCW) and Southwest Japan rotating clockwise (CW) to its present position [1,2]. This tectonic event is thought to be related to the expansion of the Japan Sea [3]. During the period associated with the Japan Sea expansion, igneous activity around the Japan Sea became active (e.g., [4]), and intermediate-to-acidic igneous rocks intruded in Southwestern Japan (e.g., [5]). The Tsushima Strait area, where the Tsushima Islands and the Goto Islands are situated, is located at the southern end of the Japan Sea and the western end of Southwestern Japan. The area does not belong to the Southwestern Japan belt from the viewpoint of the Japan Sea expansion, and geomagnetic studies revealed that the area rotated CCW, while the Southwestern Japan belt rotated CW during the Japan Sea extension [6]. Therefore, the Tsushima Strait area is thought to have been under compressional deformation while the Japan Sea Basin expanded. On the Tsushima Islands, igneous activity occurred before this CCW rotation, whereas on the Goto Islands it started afterwards. An age of c. 15 Ma was reported for the igneous rocks from the Tsushima and Goto Islands [6,7], which is almost identical to the age of expansion of the Japan Sea. According to Shin et al. (2009) [8], the Middle Miocene granitoids identified in the Tsushima Islands include leucocratic and gray granites. Leucocratic granite is thought to have originated from felsic magma produced by partial melting of the lower crust, caused by injection of mafic EM I-type magma derived, and gray granites formed by mixing/mingling of this felsic magma with ascending mafic magma in the upper crust. It was also suggested that the formation of these magmas was caused by the ascent of high-temperature mantle materials associated with the expansion of the Japan Sea [8]. Geochronological investigations showed the granitoids in the Goto Islands to be of middle Miocene age [7,9–11]. Their geochemistry, however, is not yet studied in detail. In this study, we provide whole-rock major and trace element (including
Geological Setting and Petrogenetic Information

The Goto islands are located near the Tsushima Strait, which connects the southern end of the Sea of Japan and the northern end of the East China sea, about 100 km west of the Kyushu main island, Southwestern Japan (Figure 1). The archipelago consists of five major islands and more than 200 islets and they are aligned NNE-SSW. The five major islands are named Nakadorijima, Wakamatsujima, Narushima, Hisakajima, and Fukuejima from north to south. Hisakajima and Fukuejima islands consist of Neogene sedimentary rocks of the Goto Group, Neogene igneous rocks and Quaternary volcanic rocks [11] (Figure 2). The geology of this area was studied in detail by Kawada et al. (1994) [11]. The Goto Group is widely exposed in the Goto islands and is divided into three formations—the Lower, the Middle, and the Upper formation at the Fukuejima island [12]. Among these formations, the Middle and Upper Formations are roughly equivalent to the Okuura and Toraku Formations of Sato (1952) [13], respectively [11]. The Okuura Formation is composed mainly of dark gray colored mudstone and fine- to medium-grained sandstone. The lower part of the formation experienced, in part, thermal metamorphism by the Goto granitic rocks. Tuffaceous sandstone and mudstone are intercalated in the upper part of the formation [11]. The Toraku Formation conformably overlies the Okuura Formation, and it consists of sandstone and mudstone, and green tuff intercalations in the sandstone. The sandstone sometimes contains chert conglomerates [11]. The Goto Formation contains early-to-middle Miocene fossils of plants and freshwater and brackish water shells, constraining the depositional age [12,14,15]. It is estimated that there was a fresh-water lake up to 100 km in length [16].

![Location of the study area](image)

**Figure 1.** Location of the study area—(a) Location of the Tsushima and Goto Islands; and (b) enlarged map of the Hisakajima and Fukuejima Island region.

The Neogene igneous rocks consist mainly of the Fukue Rhyolites, Goto Granites, Sazaejima Rhyolites, and Kabashima Volcanic Rocks. The extensional direction of these igneous rocks is NE-SW, almost parallel to the orientation of the Goto Islands. They are intruded by large and small dikes [11].

rare earth element (REE)) chemical compositions of the granitoids from the Hisakajima Island in the Goto Islands.
Figure 2. Geological Map showing the sample localities—(A) Hisakajima and Fukuejima Islands; (B) Hisakajima Island; and (C) northwestern coastal area of Hisakajima Island. The geological map is adopted from Matsui and Kawada (1986) [17]. Solid circles—GD group samples. Open circles—HFG group samples. Cross mark—dike sample. The large circle indicates the distribution of the HFG group rocks. At sites GTA-2, GTC-3, and GTC-4, GD Group rocks occur as enclaves in the HFG Group rocks.

The Fukue Rhyolites intruded into the central part of the Fukuejima Island and consist mainly of rhyolite tuff, tuff breccia, and overlying glassy rhyolite welded tuff. They were intruded by dacite and granophyre of the Goto Granites. The boundary between the Fukue Rhyolites and the Goto Group is usually linear with sub-vertical fault fracture zones, and is often intruded by granophyre-felsite or porphyrite dikes [11]. Whole-rock K-Ar ages of Fukue Rhyolites are 12.4 ± 0.6 Ma [11], 15.4 ± 0.3 Ma, and 15.5 ± 0.4 Ma [10]. Fission-track ages of Fukue Rhyolites are 20.02 ± 1.9 Ma, 19.8 ± 2.5 Ma [9]. Fission-track ages of welded tuff are 19.9 ± 5.0 Ma and 12.5 ± 3.0 Ma [6].

The Goto Granites consist of granodiorite, granodiorite porphyry, and granophyre. These rocks intrude into the sedimentary rocks of the Goto Group and Fukue Rhyolites as stock, dome, or dike, and caused contact metamorphism. Among the Goto Granites, the granodiorite occurs only on Hisakajima Island. It crops out mainly in the northern part of the Hisakajima Island, intruding the Goto Group [11]. Granodiorite sometimes contains light greenish gray colored oval enclaves of hornblende rich, fine-grained quartz diorite, a few to ten centimeters, sometimes several meters in diameter.

The granodiorite porphyry is a coarse-grained porphyritic rock. Large phenocrysts are mainly plagioclase and quartz. Plagioclase phenocrysts are euhedral. The colored minerals consist mainly of clinopyroxene, hornblende, and biotite, and they are significantly altered.
The porphyry intrudes into the Goto Group as stock or dome, and hydrothermal alteration sometimes forms pyrophyllite deposits [11].

The granophyre is a fine-grained rock containing small amounts of quartz and feldspar phenocrysts, and small amounts of biotite. It intrudes into the Goto Group, Fukue Rhyolites, granodiorite, and granodiorite porphyry, and is considered to be the youngest rock of the Goto Granites [11].

Whole-rock K-Ar ages of granodiorite porphyry are 13.2 ± 1.0 Ma [11], 12.1 ± 0.4 Ma, 12.0 ± 0.4 Ma, and 14.9 ± 0.4 Ma [10]. Zircon fission-track ages of Goto Granites are 14.4 ± 1.5 Ma, 14.6 ± 1.6 Ma, and 15.4 ± 2.0 Ma [9], and 16.9 ± 2.0 Ma, 13.7 ± 2.3 Ma, 16.5 ± 2.1 Ma, and 13.2 ± 1.3 Ma [6]. With the exception of the fission-track ages, the reported radiometric ages suggest that the Neogene igneous activity occurred during a relatively short interval [11].

Quaternary volcanic rocks are composed of basaltic lava of olivine basalt and augite olivine basalt, and their related pyroclastic rocks [11]. Whole-rock K-Ar ages of these rocks are 0.73 ± 0.04 Ma and 0.75 ± 0.04 Ma [10].

This study focuses on the granitic rocks of the Hisakajima island, one of the major islands in the southern part of the archipelago. A total of 25 samples of granitic rocks and basic to intermediate rock samples were analyzed.

Granodiorite consists of hornblende, biotite, plagioclase, quartz, and a small amount of potassium feldspar. The hornblende is green to brown, 1 mm or less in length, and euhedral to semi-euhedral. The biotite is greenish brown, 0.5–1 mm in length, and suffers from chloritization. The feldspar is 2 mm or less in length and altered [11]. The quartz is about 0.6–1 mm.

In the northwestern coastal area of Hisakajima Island, quartz diorite was identified in association with granodiorite; their contact was sharp.

Quartz diorite is composed of hornblende, biotite, plagioclase, and quartz. The hornblende is pale green to pale brown, generally 1 mm or less in length, euhedral to semi-euhedral columnar in shape. The biotite is about 0.3–0.5 mm in length. It often coexists with hornblende, and mostly suffered from chloritization [11]. The shape of plagioclase is acicular to prismatic. The quartz is about 0.6–0.8 mm.

The GD group and HFG group rocks, described below, could not be distinguished on the field. Both GD- and HFG-group rocks are composed of similar minerals, but microgranite microstructures were recognized only in the HFG group rocks. Opaque minerals are more common in the HFG group rocks than in the GD group rocks.

The basic-to-intermediate intrusive rocks consist of dolerite and porphyrite, and their intrusions into granitic rocks are mainly identified in the northern coastal area of Hisakajima Island.

3. Analytical Methods

The whole-rock major and trace element compositions of the 25 samples were determined using a wavelength dispersive X-ray fluorescence spectrometer (WDXRF; XRF-1800, Shimadzu Corporation, Kyoto, Japan), and an inductively coupled plasma mass spectrometer (ICP-MS; 7500 series ICP-MS, Agilent Technologies, Santa Clara, CA, USA). For the WDXRF measurements, a glass bead method was used by employing lithium tetraborate as a flux. The mixing ratio of powdered rock sample and flux were 0.7:6 g for major element (Si, Ti, Al, Fe, Mn, Mg, Ca, Na, K and P) analysis and 2:3 g for trace element (Th, Pb, Ba, Nb, Zr, Y, Sr, Rb, Zn, Cu, Ni, Co, Cr and V) analysis. A Rh target X-ray tube was used. Accelerating voltage and current for X-ray tube were 40 kV and 70 mA for major elements analyses, and 40 kV and 95 mA for trace elements analyses, respectively. Analytical uncertainties were estimated to be better than 2% for SiO$_2$ and Al$_2$O$_3$, and better than 5% for the rest of the major-elements. (e.g., [18]). Details on analytical methods were reported by Nakazaki et al. (2004) [19]. For ICP-MS measurements, the glass beads for trace elements analysis were decomposed in hydrochloric acid, and the REE (La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu) were separated as a group, using a cation exchange resin
(DOWEX™ 50W × 8 200–400 Mesh (H) Cation Exchange Resin). The REE fractions were dissolved in nitric acid for analysis by ICP-MS.

4. Results and Discussion

The granitic rocks in the northern part of Hisakajima Island are divided into two groups. In the first group, where the total Fe for the FeO*/MgO ratio is shown as FeO*, showed little increase with increasing SiO₂ content. In the second group, the FeO*/MgO ratio varied widely but did not correlate with SiO₂ content (Figure 3, Table 1). The relationship between the petrogenesis of granite and the Fe in granite is discussed (e.g., [20]). In this study, rocks with FeO*/MgO ratios less than 10 were classified as granodioritic (GD) group, and rocks with FeO*/MgO ratios greater than 10 were classified as the High FeO*/MgO (HFG) group. Most GD group samples were classified as granodiorite but only GTB-3 was classified as granite. The HFG group samples were all classified as granite. The quartz diorite samples that occur as enclaves in the HFG group rocks, were classified as GD group rocks.

Figure 3. FeO*/MgO-SiO₂ diagram [21] for the rocks in the Hisakajima Island. Solid circles—GD group samples. Open circles—HFG group samples. Cross mark—dike sample. TH—tholeiitic series. CA—calc-alkalic series. Dike samples GTA-2b and GTB-5b fall outside the plotting range. Total Fe as FeO*.

The HFG group samples were distributed in a similar direction to the northeast-southwest extension of the Goto Islands. The Miocene Fukue Rhyolites and the Goto Granites from the Fukuejima and Hisakajima Islands were also distributed in a similar direction (Figure 2). This implied that the HFG group granites also intrude into the granitic pluton of the GD group. The GD samples that were found as enclaves in the HFG pluton were considered to be remnants that escaped assimilation during the intrusion of the HFG magma.

The SiO₂ contents of the GD and HFG group rocks ranged from 63.3–72.4 wt.% and 72.0–75.6 wt.%, respectively. The FeO*/MgO ratios of the GD group rock samples varied from 3.4 to 7.1, increasing slightly with the increasing SiO₂ contents, while the HFG group rock samples showed ratios between 10.2 and 23.5 and showed little variation in SiO₂. Most HFG group samples showed FeO*/MgO ratios in the range of 18–23, although four samples showed FeO*/MgO ratios in the range of 10–15. These four samples cropped out in the northwestern coastal area of the Hisakajima Island, where quartz diorite inclusions and porphyrite dikes occur.
Table 1. Whole-rock chemical composition of rocks in the Hisakajima Island.

| Sample | GD Group Rocks | Dikes |
|--------|----------------|-------|
|        | GTA-2a | GTB-3 | GTB-7a | GTB-7b | GTC-3a | GTC-4a | GTC-9a | GTC-9b | GTC-9c | GTC-9d | GTA-2b | GTB-5b | GTC-7 |
| Lat. N32° |        |       |       |       |       |       |       |       |       |       |       |       |       |
| Lon. E128° |        |       |       |       |       |       |       |       |       |       |       |       |       |
| SiO₂ (wt.%) | 68.14 | 72.35 | 68.30 | 67.52 | 64.45 | 64.45 | 70.55 | 65.30 | 65.09 | 48.93 | 48.81 | 62.40 |        |
| TiO₂ | 0.63 | 0.31 | 0.45 | 0.85 | 0.78 | 0.42 | 0.64 | 0.57 | 0.60 | 1.46 | 1.45 | 0.75 |        |
| Al₂O₃ | 14.05 | 12.89 | 14.44 | 15.01 | 15.33 | 15.19 | 14.40 | 15.42 | 15.43 | 15.09 | 15.69 | 15.58 |        |
| FeO⁺ | 4.09 | 2.90 | 3.28 | 5.80 | 3.26 | 3.73 | 3.79 | 4.14 | 4.14 | 12.70 | 12.69 | 5.97 |        |
| MnO | 0.16 | 0.08 | 0.13 | 0.23 | 0.24 | 0.12 | 0.15 | 0.14 | 0.15 | 0.26 | 0.26 | 0.17 |        |
| MgO | 1.08 | 0.41 | 0.67 | 1.47 | 1.28 | 0.52 | 1.09 | 0.74 | 0.93 | 5.06 | 5.05 | 2.13 |        |
| CaO | 3.28 | 1.98 | 2.68 | 4.21 | 3.50 | 3.20 | 3.18 | 4.19 | 4.34 | 9.21 | 9.22 | 4.28 |        |
| Na₂O | 4.06 | 4.10 | 4.77 | 5.14 | 4.62 | 4.27 | 4.82 | 4.94 | 5.45 | 4.54 | 3.01 | 3.03 | 4.15 |        |
| K₂O | 1.44 | 2.21 | 1.82 | 0.66 | 0.93 | 1.37 | 1.69 | 1.59 | 2.32 | 0.50 | 0.50 | 1.14 |        |
| P₂O₅ | 0.18 | 0.06 | 0.11 | 0.33 | 0.28 | 0.10 | 0.22 | 0.15 | 0.18 | 0.16 | 0.15 | 0.12 |        |
| LOI | 0.86 | 0.78 | 1.03 | 0.94 | 0.87 | 0.38 | 0.77 | 1.01 | 0.56 | 0.73 | 1.93 | 1.56 |        |
| Total | 97.97 | 98.09 | 97.26 | 98.02 | 98.53 | 98.24 | 97.23 | 98.24 | 98.61 | 97.72 | 98.80 | 98.25 |        |
| FeO⁺/MgO | 3.79 | 7.11 | 5.01 | 4.16 | 4.52 | 6.23 | 3.40 | 5.15 | 4.43 | 2.51 | 2.51 | 2.80 |        |
| K₂O+Na₂O | 5.50 | 6.31 | 6.19 | 6.96 | 5.28 | 5.21 | 6.19 | 6.63 | 7.03 | 6.87 | 3.51 | 3.54 | 5.29 |        |
| ASI | 0.99 | 1.01 | 1.01 | 0.96 | 0.96 | 1.02 | 0.97 | 1.02 | 0.95 | 0.71 | 0.70 | 0.98 |        |
| Th (ppm) | 6.51 | 6.59 | 4.46 | 6.35 | 2.49 | 1.45 | 3.94 | 4.20 | 4.51 | 3.94 | — | — | 2.47 |
| Pb | 17.7 | 12.6 | 16.3 | 9.07 | 16.9 | 12.6 | 20.5 | 16.4 | 12.7 | 24.5 | 11.5 | 11.3 | 20.8 |
| Ba | 376 | 451 | 391 | 449 | 215 | 272 | 360 | 414 | 403 | 488 | 109 | 100 | 300 |
| Nb | 3.18 | 2.48 | 2.84 | 2.54 | 1.92 | 1.75 | 1.58 | 3.04 | 3.42 | 2.71 | 1.06 | — | 1.11 |
| Zr | 174 | 176 | 182 | 188 | 143 | 169 | 225 | 137 | 186 | 167 | 71.2 | 54.0 | 131 |
| Y | 48.7 | 41.9 | 36.1 | 40.8 | 34.8 | 37.4 | 26.4 | 47.4 | 39.5 | 45.2 | 17.2 | 25.4 | 36.2 |
| Sr | 157 | 130 | 184 | 203 | 231 | 232 | 181 | 214 | 194 | 197 | 237 | 314 | 213 |
| Rb | 48.7 | 76.8 | 36.3 | 42.6 | 28.0 | 36.5 | 34.5 | 44.9 | 50.7 | 58.1 | 28.36 | 41.77 | 51.86 |
| Zn | 95.3 | 61.0 | 76.2 | 79.8 | 121 | 122 | 87.8 | 74.2 | 63.1 | 96.6 | 108 | 104 | 79.3 |
| Cu | — | 0.19 | 13.0 | — | — | — | — | 12.1 | 16.7 | 3.45 | 20.2 | — | — |
| Ni | 2.13 | 3.81 | 2.42 | 2.57 | — | 0.29 | 2.87 | 1.69 | 2.71 | 1.02 | 1.29 | 19.5 | 11.3 |
| Co | 10.7 | 7.52 | 7.77 | 8.43 | 16.5 | 16.4 | 7.27 | 9.58 | 10.3 | 11.8 | 67.5 | 35.6 | 21.6 |
| Cr | 2.47 | 3.35 | — | — | 1.29 | 0.54 | 0.77 | — | — | — | — | 42.0 | 91.3 |
| V | 38.0 | 20.7 | 13.7 | 12.9 | 7.10 | 4.3 | 16.0 | 18.1 | 18.5 | 30.1 | 435 | 284 | 157 |
| Y + Nb | 51.9 | 49.4 | 38.9 | 43.3 | 36.7 | 39.2 | 28.0 | 50.4 | 42.9 | 48.0 | 18.3 | 25.4 | 37.4 |
| Sample | GD Group Rocks | Dikes |
|--------|----------------|-------|
|       | GTA-2a         | GTA-3 | GTA-7a | GTA-7b | GTA-3a | GTA-4a | GTA-9a | GTA-9b | GTA-9c | GTA-9d | GTA-2b | GTA-5b | GTA-7 |
| Lat. N32°  | Lon. E128° | GTA-2a | GTA-3 | GTA-7a | GTA-7b | GTA-3a | GTA-4a | GTA-9a | GTA-9b | GTA-9c | GTA-9d | GTA-2b | GTA-5b | GTA-7 |
| La (ppm) | GTA-2a         | GTA-3 | GTA-7a | GTA-7b | GTA-3a | GTA-4a | GTA-9a | GTA-9b | GTA-9c | GTA-9d | GTA-2b | GTA-5b | GTA-7 |
| HD Group Rocks | GTA-2c | GTA-3 | GTA-4 | GTA-5a | GTA-6 | GTA-8 | GTA-9 | GTA-3b | GTA-4b | GTA-5 | GTA-6 | GTA-8 | GTA-10 |
| Lat. N32°  | Lon. E128° | GTA-2c | GTA-3 | GTA-4 | GTA-5a | GTA-6 | GTA-8 | GTA-9 | GTA-3b | GTA-4b | GTA-5 | GTA-6 | GTA-8 |
| SiO2 (wt.%) | GTA-2c | GTA-3 | GTA-4 | GTA-5a | GTA-6 | GTA-8 | GTA-9 | GTA-3b | GTA-4b | GTA-5 | GTA-6 | GTA-8 | GTA-10 |
| TiO2 | GTA-2c | GTA-3 | GTA-4 | GTA-5a | GTA-6 | GTA-8 | GTA-9 | GTA-3b | GTA-4b | GTA-5 | GTA-6 | GTA-8 | GTA-10 |
| Al2O3 | GTA-2c | GTA-3 | GTA-4 | GTA-5a | GTA-6 | GTA-8 | GTA-9 | GTA-3b | GTA-4b | GTA-5 | GTA-6 | GTA-8 | GTA-10 |
| FeO* | GTA-2c | GTA-3 | GTA-4 | GTA-5a | GTA-6 | GTA-8 | GTA-9 | GTA-3b | GTA-4b | GTA-5 | GTA-6 | GTA-8 | GTA-10 |
| MnO | GTA-2c | GTA-3 | GTA-4 | GTA-5a | GTA-6 | GTA-8 | GTA-9 | GTA-3b | GTA-4b | GTA-5 | GTA-6 | GTA-8 | GTA-10 |
| MgO | GTA-2c | GTA-3 | GTA-4 | GTA-5a | GTA-6 | GTA-8 | GTA-9 | GTA-3b | GTA-4b | GTA-5 | GTA-6 | GTA-8 | GTA-10 |
| CaO | GTA-2c | GTA-3 | GTA-4 | GTA-5a | GTA-6 | GTA-8 | GTA-9 | GTA-3b | GTA-4b | GTA-5 | GTA-6 | GTA-8 | GTA-10 |
| Na2O | GTA-2c | GTA-3 | GTA-4 | GTA-5a | GTA-6 | GTA-8 | GTA-9 | GTA-3b | GTA-4b | GTA-5 | GTA-6 | GTA-8 | GTA-10 |
## Table 1. Cont.

| Sample | GTA-2c | GTA-3 | GTB-4 | GTB-5a | GTB-6 | GTB-8 | GTB-9 | GTC-3b | GTC-4b | GTC-5 | GTC-6 | GTC-8 |
|--------|--------|-------|-------|--------|-------|-------|-------|--------|--------|-------|-------|-------|
| Lat. N32° | 49°56′ | 49°41′ | 49°49′ | 49°58′ | 50°′ | 49°41′ | 49°56′ | 49°57′ | 49°48′ | 49°39′ |       |       |
| Lon. E128° | 50°54′ | 51°2′ | 52°32′ | 51°57′ | 52°1′ | 51°44′ | 51°38′ | 50°55′ | 50°59′ | 50°57′ | 51°2′ |       |
| K₂O | 2.33 | 1.76 | 1.95 | 2.33 | 1.89 | 1.99 | 2.22 | 2.12 | 2.31 | 2.16 | 2.36 | 2.30 |
| P₂O₅ | 0.03 | 0.05 | 0.05 | 0.02 | 0.04 | 0.04 | 0.04 | 0.03 | 0.04 | 0.04 | 0.04 | 0.04 |
| LOI | 0.62 | 0.62 | 0.85 | 1.31 | 1.25 | 0.81 | 0.88 | 0.67 | 0.58 | 0.48 | 0.68 | 0.62 |
| Total | 98.36 | 98.28 | 99.56 | 98.69 | 99.85 | 98.80 | 97.75 | 98.96 | 98.54 | 98.70 | 98.75 |       |
| FeO*/MgO | 18.6 | 22.7 | 23.5 | 20.0 | 17.9 | 18.5 | 18.5 | 14.8 | 11.8 | 13.4 | 10.2 | 19.5 |
| K₂O+Na₂O | 6.49 | 6.40 | 6.32 | 6.78 | 6.12 | 6.23 | 6.52 | 6.25 | 6.62 | 6.40 | 6.79 | 6.58 |
| ASI | 1.05 | 0.99 | 1.02 | 1.10 | 1.03 | 1.06 | 1.04 | 1.08 | 1.03 | 1.02 | 1.06 | 0.99 |
| Th (ppm) | 5.76 | 5.93 | 6.44 | 7.05 | 7.38 | 6.60 | 7.29 | 7.16 | 4.89 | 6.08 | 6.16 | 6.41 |
| Pb | 14.1 | 19.7 | 13.5 | 11.6 | 19.4 | 18.8 | 12.5 | 16.5 | 16.8 | 16.5 | 14.8 | 15.4 |
| Ba | 568 | 454 | 477 | 570 | 458 | 525 | 525 | 511 | 585 | 473 | 523 | 529 |
| Nb | 4.43 | 3.52 | 3.63 | 4.63 | 3.54 | 3.48 | 3.66 | 4.93 | 3.65 | 3.46 | 3.47 | 3.95 |
| Zr | 219 | 215 | 198 | 253 | 213 | 196 | 221 | 234 | 205 | 216 | 217 | 224 |
| Y | 47.9 | 46.1 | 43.9 | 50.3 | 51.1 | 41.9 | 57.4 | 49.3 | 50.8 | 48.6 | 48.3 | 49.7 |
| Sr | 123 | 125 | 138 | 150 | 127 | 124 | 118 | 106 | 72.4 | 121 | 122 | 117 |
| Rb | 69.4 | 56.7 | 66.9 | 71.3 | 65.5 | 65.9 | 80.1 | 65.8 | 78.4 | 81.2 | 87.0 | 84.5 |
| Zn | 49.0 | 116 | 100 | 30.9 | 83.4 | 85.1 | 66.1 | 62.0 | 44.8 | 123 | 41.6 | 55.1 |
| Cu | 26.1 | 0.08 | — | — | 0.29 | 0.17 | 0.22 | 2.04 | — | 1.12 | 6.32 | — |
| Ni | 4.50 | 1.76 | 3.60 | 5.00 | 2.99 | 4.15 | 4.21 | 4.67 | 4.27 | 3.41 | 4.50 | 3.37 |
| Co | 5.00 | 14.0 | 7.10 | 4.30 | 8.79 | 6.69 | 7.35 | 5.53 | 5.35 | 5.98 | 5.43 | 8.05 |
| Cr | 0.62 | 2.79 | 0.51 | — | 1.50 | 0.61 | 1.60 | 1.07 | 0.57 | 1.84 | 0.45 | 0.25 |
| V | 9.29 | 3.81 | 4.45 | 6.56 | 5.44 | 6.11 | 8.75 | 8.34 | 6.23 | 10.4 | 6.94 | 6.47 |
| Y + Nb | 52.3 | 49.6 | 47.5 | 54.9 | 54.6 | 45.4 | 61.1 | 54.2 | 54.4 | 52.0 | 51.7 | 53.7 |
Table 1. Cont.

| Sample | GTA-2c | GTA-3 | GTB-4 | GTB-5a | GTB-6 | GTB-8 | GTB-9 | GTC-3b | GTC-4b | GTC-5 | GTC-6 | GTC-8 |
|--------|--------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| Lat. N32° | 49°56' | 49°49' | 49°58' | 50°2' | 49°41' | 49°56' | 49°57' | 49°49' | 49°48' | 49°39' |
| Lon. E128° | 50°54' | 51'2'' | 52'32'' | 51'57'' | 52'1'' | 51'44'' | 51'38'' | 50'55'' | 50'55'' | 50'59'' | 50'57'' | 51'2'' |

- **La (ppm)**: 28.1, 23.0, 25.4, 40.6, 28.5, 22.4, 24.5, 26.5, 25.6, 25.7, 28.6, 23.4
- **Ce**: 60.8, 50.3, 55.4, 55.8, 67.9, 46.3, 52.3, 57.5, 55.2, 56.9, 61.4, 52.1
- **Pr**: 7.43, 6.27, 6.86, 10.7, 7.78, 6.23, 6.67, 7.04, 6.77, 7.15, 7.43, 6.51
- **Nd**: 31.1, 27.1, 29.1, 42.3, 33.0, 26.4, 28.8, 29.7, 28.5, 30.5, 31.1, 27.9
- **Sm**: 7.10, 6.59, 6.86, 9.40, 7.91, 6.20, 7.19, 7.14, 6.70, 7.34, 7.22, 6.85
- **Eu**: 1.55, 1.66, 2.06, 2.04, 1.69, 1.61, 1.46, 1.47, 1.70, 1.52, 1.68
- **Gd**: 7.22, 7.00, 7.17, 8.55, 8.60, 6.43, 8.06, 7.45, 7.13, 7.50, 7.30, 7.40
- **Tb**: 1.29, 1.29, 1.32, 1.55, 1.59, 1.21, 1.49, 1.37, 1.31, 1.35, 1.30, 1.36
- **Dy**: 8.29, 8.41, 8.50, 9.49, 10.5, 7.24, 9.85, 8.97, 8.60, 8.66, 8.21, 8.94
- **Ho**: 1.75, 1.80, 1.79, 1.87, 2.23, 1.52, 2.15, 1.92, 1.86, 1.85, 1.75, 1.95
- **Er**: 5.43, 5.57, 5.52, 5.58, 6.96, 4.67, 6.63, 5.91, 5.80, 5.60, 5.28, 6.03
- **Tm**: 0.79, 0.82, 0.80, 0.80, 1.02, 0.68, 0.96, 0.88, 0.84, 0.82, 0.77, 0.87
- **Yb**: 5.33, 5.66, 5.49, 5.57, 6.90, 4.52, 6.37, 6.07, 5.87, 5.67, 5.29, 5.94
- **Lu**: 0.81, 0.86, 0.81, 0.83, 1.02, 0.67, 0.94, 0.89, 0.86, 0.83, 0.80, 0.89

Lat.—latitude of the sample site. Lon.—longitude of the sample site. LoI—loss on ignition. ASI—molar $\text{Al}_2\text{O}_3$ / ($\text{Na}_2\text{O} + \text{K}_2\text{O} + \text{CaO}$). —: not detected. Total Fe as FeO*.
FeO*, MnO, TiO₂, CaO, P₂O₅, Al₂O₃, and MgO were negatively correlated with SiO₂, whereas K₂O was positively correlated with SiO₂. On Harker diagrams, the HFG group samples showed a steeper correlation for FeO* and MnO than the GD group samples, whereas Al₂O₃ showed a less steep correlation with SiO₂ (Figure 4). Especially, FeO* and MnO showed distinctly different trends. Regarding trace element compositions, Th, Ba, Nb, Zr, Y, Rb, and Ni increased with increasing SiO₂ content, and Sr, Zn, and Co decreased with increasing SiO₂ content (Figure 5). For Zn and Co, the HFG group samples showed distinctly different trends compared to the GD group samples. These facts suggest that the GD group and HFG group samples have different origins in terms of simple magmatic crystal differentiation processes. On the AFM diagram (Figure 6), which is a triangular diagram with FeO*, MgO, and alkalis (K₂O + Na₂O) as end components, the GD and HFG group samples showed distinctly different trends. The GD group samples showed a decrease in both FeO* and MgO, with increasing alkalis, while the HFG group samples showed a decrease in FeO* with increasing alkali contents, at approximately constant and very low MgO.

In the calc–alkali–tholeiite series classification diagram of Miyashiro (1974) [21], the granitic rocks from the Hisakajima Island were all classified as tholeiitic, with high FeO*/MgO ratios. The conditions under which magmatic rocks followed such a trend were—(1) the magma was reducing, thus minerals such as magnetite were not formed during the initial stage of magma crystallization, and Mg-rich minerals, such as olivine or pyroxene, were selectively crystallized. As crystallization proceeded, the melt became relatively Fe-rich and Mg-poor; (2) Mg became depleted due to the post-crystallization of Mg-containing minerals such as biotite and the relative FeO*/MgO ratio increased rapidly; and (3) the magma source material was Fe-rich and Mg-poor.
Figure 5. Harker diagrams for trace elements. Solid circle—GD group samples. Open circle—HFG group samples. Cross mark—dike sample. The plots of dike samples of GTA-2b and GTB-5b are out of range on this diagram.
In the case of the GD group samples, the FeO*/MgO ratio increased linearly with increasing SiO$_2$ in the range of 63.3–72.4 wt.%, suggesting that Mg was not depleted and that the second process did not occur.

In addition, the AFM diagram showed that the fraction of MgO in the GD group samples decreased with increasing alkali content, suggesting that the GD group samples were depleted in Mg as crystal differentiation proceeded, and that both Fe and Mg were abundant in the early stage of crystal differentiation (Figure 6). Therefore, condition (3) also did not apply to the samples in the GD group, and they were considered to be generated under condition (1). For the samples of the HFG group, the AFM diagram revealed a low and approximately constant Mg fraction, regardless of the degree of crystal differentiation.

The SiO$_2$ content of the HFG samples in this study was limited to the range 72.0–75.9 wt.%, suggesting that the magma was not the product of magmatic differentiation. The microgranite microstructures observed in the HFG group samples suggest that there was not enough time for crystal differentiation before the magma cooled and the rock formed. Therefore, conditions (1) and (2) were not applicable to the formation of the HFG group samples, and it was likely that they were formed under condition (3).

The alumina saturation index (ASI) of the GD group samples ranged from 0.95 to 1.02, while the HFG group samples had a higher ASI between 0.99 and 1.10 (Figure 7). This suggests that the HFG group had an S-type granitoid composition, which is a sedimentary source. A notable feature of the HFG group was that it showed a significantly steeper trend for Fe and Mn in the Harker diagrams than the GD group (Figure 4). This is likely a characteristic of the source.

MORB-normalized [22] trace element concentrations revealed that the HFG group samples were relatively rich in LIL elements compared to the GD group samples (Figure 8). A geochemical discrimination diagram based on Rb vs. Y + Nb compositions [23] showed that the GD group samples plotted into the volcanic arc granite field, while the HFG group samples plotted close to the boundary between the volcanic arc granite and the within-plate granite fields (Figure 9). This suggests that the GD group granitic rocks in the Hisakajima Island underwent igneous fractionation and that the source material might contain an enriched mantle material [23].
Figure 7. ASI-SiO₂ diagram for the rocks in the Hisakajima Island. A/CNK—molar Al₂O₃/(CaO + Na₂O + K₂O). Solid circles—GD group samples. Open circles—HFG group samples. Cross mark—dike sample. Dike samples GTA-2b and GTB-5b fall outside the plotting range.

Figure 8. Spider diagrams for the rocks in the Hisakajima Island (normalized to average MORB [22]).

Figure 9. Geochemical discrimination diagram [23] for the rocks from the Hisakajima Island. syn-COLG—collisional granites. VAG—volcanic-arc granites. WPG—within-plate granites. ORG—ocean-ridge granites. Solid circles—GD group samples. Open circles—HFG group samples.
These results suggest that the GD Group rocks might have originated from fractional crystallization of a reducing magma derived from enriched mantle material, and the HFG Group rocks might have been generated by partial melting of Fe- and Mn-rich materials derived from sediments in the crust. In order to produce reducing magma, which is the inferred origin of the GD group, it is necessary that the magma was generated without an influence from subduction zones, where O\textsubscript{2} and H\textsubscript{2}O would be supplied from the outside.

In fact, the granites of the Tsushima Islands, in the Tsushima Strait area, were inferred by Shin et al. (2009) [8] to have been generated as a consequence of upwelling of high-temperature mantle material associated with the expansion of the Japan Sea. This model predicts that the magma reaches the crust without being affected by external factors, such as subduction zones. We also adopt such a model for the GD group granitoids. However, the origin of GD group rocks remains an open question. Upwelling of high-temperature material might also be responsible for partial melting of the crust and thus account for the origin of the magma of the HFG group. This suggests that the generation of the granitic rocks of the Hisakajima Island is most likely related to the upwelling of high-temperature mantle material. Models of the influx of high-temperature asthenosphere, such as those of Hirai et al. (2018) [24], also suggest that the extension of the Japan Sea established the conditions for the production of these magmas.

CI-chondrite normalized REE patterns [25] showed that both GD- and HFG-group samples were enriched in light REE, relative to heavy REE (Figure 10). A comparison between the two groups shows that both positive and negative Eu anomalies were observed in the GD group samples, whereas the HFG group showed a Ce anomaly in one sample and negative Eu anomalies in all samples. Among the HFG group samples, sample GTB-5a had the highest light REE concentrations, as well as a large negative Ce anomaly. This is because the original REE composition of the sample GTB-5a is similar to that of other HFG group samples. Due to the presence of dikes, it might have been subjected to secondary effects such as hydrothermal fluids, to impart REE.

![Figure 10. Chondrite-normalized REE diagrams for the Hisakajima Island rocks—(left) GD group samples; and (right) HFG group samples. Normalization values from [25].](image)

The reason for the significantly higher La/Yb ratio of GTB-5a might be that the light REEs were more incompatible than the heavy REEs, and were more abundant in the fluids where they exerted their secondary effects (Figure 11). In addition, Ce can exist stably in the tetravalent state, and Ce (IV) forms precipitates in solution. Therefore, the negative Ce anomaly suggests that the fluid that exerted the secondary effect was oxidizing, and that Ce (IV) was removed from the fluid.
The normalized La contents of the GD group samples ranged from 51.2–170.5, and the normalized La/Yb ratio from ranged from 2.1–4.7, indicating that the incompatible REE were concentrated in the magma, as it crystallized. In contrast, the normalized La content and the normalized La/Yb ratio of the HFG group samples excluding GTB-5a, were in the range of 95.2–122.1 and 2.7–3.8, respectively, and were within the range of the GD group, without showing any clear correlation.

These results supported the model that GD and HFG group samples have different origins in terms of simple crystal differentiation processes, and that the process of HFG group rocks arising from magma took place in such a short time that sufficient crystal differentiation did not occur. Among the HFG group samples such as GTA-2c, GTB-5a, and GTC-6, those with relatively high La contents and La/Yb ratios tend to be associated with nearby dikes of dolerite and porphyrite. As in the case of sample GTB-5a, the high La/Yb ratio is thought to be caused by the preferential addition of light REE, such as La, due to the secondary effects of hydrothermal fluids.

The extension of the Sea of Japan was a favorable event for the upwelling of high-temperature mantle material, which is necessary for the formation of the parent magma of the granitic rocks exposed on Hisakajima Island, Goto Islands, Southwestern Japan.

In addition, the compressive deformation field of CCW rotation in the Tsushima Strait area caused by the Japan Sea extension might have contributed to the uniform directional intrusion of the Fukue Rhyolites and Goto Granites, which might have contributed to the exposure of the GD and HFG group granitic rocks that existed in the basement to the surface. In addition, from the viewpoint of rare earth element composition, the granitic rocks in the Hisakajima Island were subjected to secondary effects, such as hydrothermal alteration and elemental exchange since their formation, leading to their present form.

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