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K–Ar geochronology of a middle Miocene submarine volcano-plutonic complex in southwest Japan

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Abstract – A volcano-plutonic complex in the Susa area, southwest Japan, consists of the Yamashima andesites, the Koyama gabbros and syn-plutonic porphyrite dykes derived from a common basaltic andesite magma. The complex is closely associated with middle Miocene turbidite deposits. The Yamashima andesites are composed mainly of basaltic andesite feeder dykes, massive submarine lavas with hyaloclastites, and their reworked deposits. The lavas and deposits immediately overlie turbidite deposits, indicating submarine volcanic activity. The Koyama gabbros formed hornfels by contact metamorphism of the surrounding turbidites and andesites. Highly purified clinopyroxene and plagioclase mineral separates from the Yamashima andesites were dated by a K–Ar method using an ultra-low blank K analysis procedure. Ages obtained from duplicate analyses are 16.5 ± 1.5, 15.2 ± 1.4, 15.8 ± 1.7, and 16.5 ± 2.0 Ma for clinopyroxene, and 14.2 ± 0.8, 15.2 ± 0.9, and 15.6 ± 0.9 Ma for plagioclase. The clinopyroxene and plagioclase data define a mineral isochron age of 14.7 ± 0.9 (1σ) Ma with an initial ^{40}Ar/^{36}Ar ratio of 297.3 ± 2.4 (1σ), suggesting that clinopyroxene has no excess argon and can be reliably dated by K–Ar. Most of the groundmass ages are considerably younger (12.1–14.6) than the isochron age, perhaps due to argon loss during alteration. The gabbros give ages of 14.2 ± 0.3 and 14.1 ± 0.3 Ma for biotite, and 13.7 ± 0.3 and 13.7 ± 0.7 Ma for green hornblende. The porphyrite dyke yields an age of 12.5 ± 0.3 Ma for the groundmass, and the pelitic hornfels gives a biotite age of 14.8 ± 0.3 Ma. Our new K–Ar ages, together with previous studies, show that a series of geological events took place in the Susa area between 16 and 13 Ma. Conglomerates and sandstones were deposited in the beginning of marine transgression. Subsequent abrupt deepening led to deposition of a thick black shale unit, turbidite deposits and large-scale submarine channel-fill deposits. Coeval igneous activity formed the volcano-plutonic complex. The magmato-tectonic event was synchronous with the opening of the Japan Sea and the associated clockwise rotation of the southwest Japan arc sliver, recording a unique tectonic setting.

Keywords: gabbro, andesite, K/Ar, geochronology, Miocene, clinopyroxene, Japan Sea.

1. Introduction

Volcano-plutonic complexes have been concisely referred to as ‘plutons that rose into their own ejecta’ (Myers, 1975). They are important geological units which offer views of the roots of volcanoes, and the fossil magma chambers they represent reveal the subterranean structure of volcanic edifices.

Numerous Cretaceous to Tertiary volcano-plutonic complexes occur in the Chugoku district of southwest Japan (e.g. Murakami, 1974; Imaoka, 1986; Imaoka *et al.* 1988; Yuge, Imaoka & Iizumi, 1998). In the Susa area of this district, the Yamashima andesites occur in close association with the Koyama gabbros (Figs 1, 2). This association between effusive andesites and intrusive gabbros is an example of a middle Miocene submarine volcano-plutonic complex (Imaoka & Nakashima, 1996; Imaoka *et al.* 1997), hereafter collectively called the Susa complex. Imaoka *et al.* (1996) attempted to apply K–Ar chronology to both the volcanic and plutonic suites to unravel the formation process of the Susa complex, and to confirm the genetic link between the Koyama gabbros and the Yamashima andesites. However, because the Yamashima andesites are highly altered, it was difficult to determine reliable ages using the techniques then available. The K–Ar ages obtained in that work ranged from 8 to 14 Ma.

Itaya, Doi & Ohira (1996) developed a flame photometric method to precisely determine K contents in minerals and rocks at very low concentrations. They applied the method to K–Ar dating of mineral separates from ophiolite complexes. The method utilizes an ultra-low blank chemical procedure which enables potassium analyses at levels as low as 0.001 wt %. This method can be applied to unaltered phenocrysts separated from the Yamashima andesites, in which the groundmass is commonly altered.

In our present study, K–Ar age determinations were carried out on clinopyroxene and plagioclase phenocryst separates from the Yamashima andesites, and on hornblende and biotite separates from the Koyama

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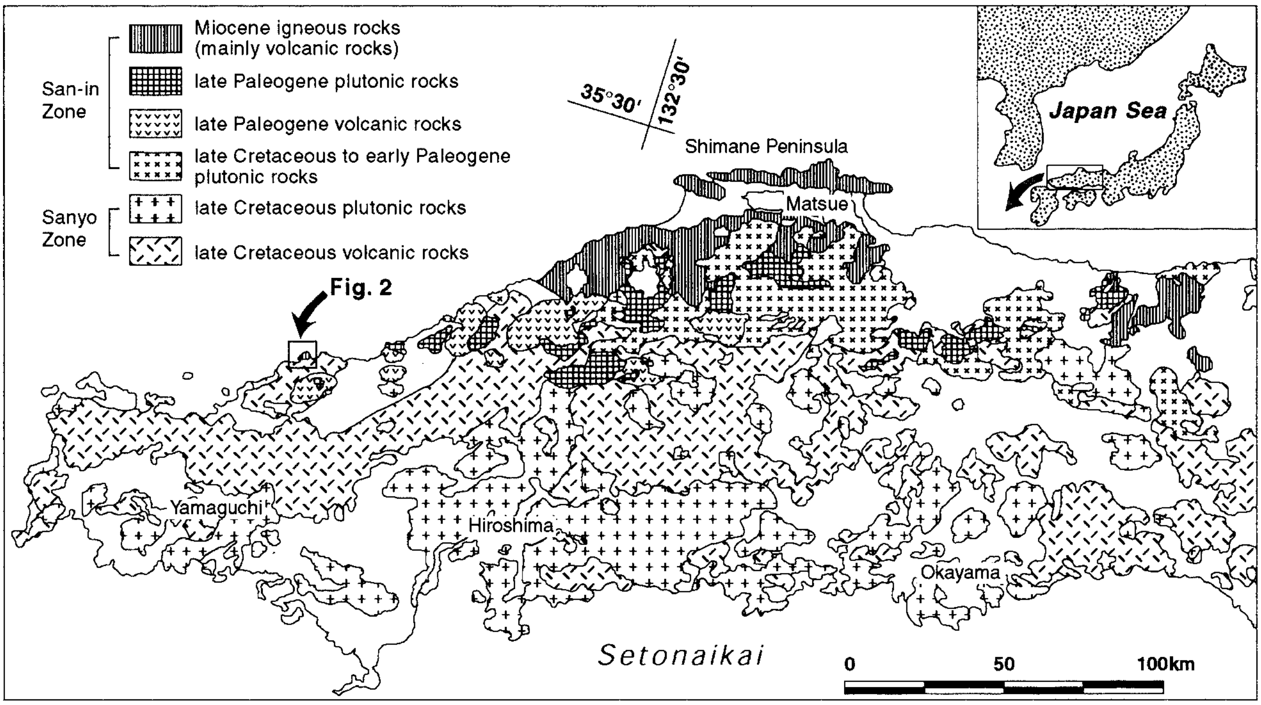

Figure 1. Index map showing the location of the study area and the distribution of Cretaceous to Tertiary igneous rocks in the Chugoku district, southwest Japan (after Geological Survey of Japan, 1992).

gabbros. We discuss the reliability of clinopyroxene K–Ar dating and describe the formation process of the Susa volcano-plutonic complex, based on the ages of volcanic rocks obtained in this study and the ages and lithologies of the host sedimentary rocks reported in the literature.

Our new K–Ar ages suggest that igneous activity was initiated at 16–15 Ma. This coincided with opening of the Japan Sea back-arc basin and clockwise rotation of the southwest Japan arc sliver in which the Susa complex occurs (Otofuji, Matsuda & Nohda, 1985; Otofuji & Matsuda, 1987; Otofuji, Itaya & Matsuda, 1991). The igneous activity took place before, during and after opening of the Japan Sea back-arc basin (Takahashi, 1986; Otofuji, Itaya & Matsuda, 1991; Tamaki *et al.* 1992; Uto *et al.* 1994; Uto, 1996; Kimura *et al.* 2003). Recently, Kimura *et al.* (2003) described the temporal and spatial variations of Late Cenozoic volcanic activity and evolution of the magmato-tectonic belt in the Chugoku district in relation to back-arc basin opening and reinitiation of subduction, based on 442 radiometric ages, including 108 new analyses. The Late Cenozoic magmatic front in southwest Japan moved southward compared with the Paleogene period, and expansion of the volcanic zone into the fore-arc occurred between 20 and 12 Ma (Kimura *et al.* 2003). Expansion of the volcanic zone parallel to the Japan Sea coast and concomitant and abrupt change in magma chemistry may have been related to upwelling of undepleted-mantle asthenosphere induced by opening of the Japan Sea back-arc basin (Nohda *et al.* 1988; Tatsumi *et al.* 1989; Uto *et al.* 1994; Pouclet *et al.* 1995; Kimura *et al.* 2003). The widespread nature of the magmatism has been attributed to rollback of the Heike microplate, which caused passive mantle upwelling (Yamaji & Yoshida, 1998). As such, rifting of the southwest Japan arc and opening of the Japan Sea are believed to have occurred during early to middle Miocene times and geochemical change in igneous rocks of that period to be related to such a tectonic event. However, tectonic syntheses by Tamaki *et al.* (1992), Jolivet & Tamaki (1994), Hall (2002) and others have demonstrated that the timing and mode of opening of the Japan Sea are still matters of debate. In this paper, we also discuss the significance of the activity of the Susa complex in relation to the major tectonic event that occurred along the eastern margin of the evolving Japan Sea.

2. Geological setting

During Cretaceous to Tertiary times, southwest Japan was a typical arc-trench system developed along the continental margin of Asia. The magmatic arc is regarded as paired with the Late Cretaceous to Miocene subduction complex of the Shimanto Belt (Maruyama, 1997; Taira, 2001; Hall, 2002). Cretaceous to early Paleogene felsic to intermediate igneous rocks are widely distributed in the Chugoku district of southwest Japan, and their radiometric ages are concentrated around 85 Ma. Latest Cretaceous to late Paleogene igneous rocks are distributed only in the San-in zone

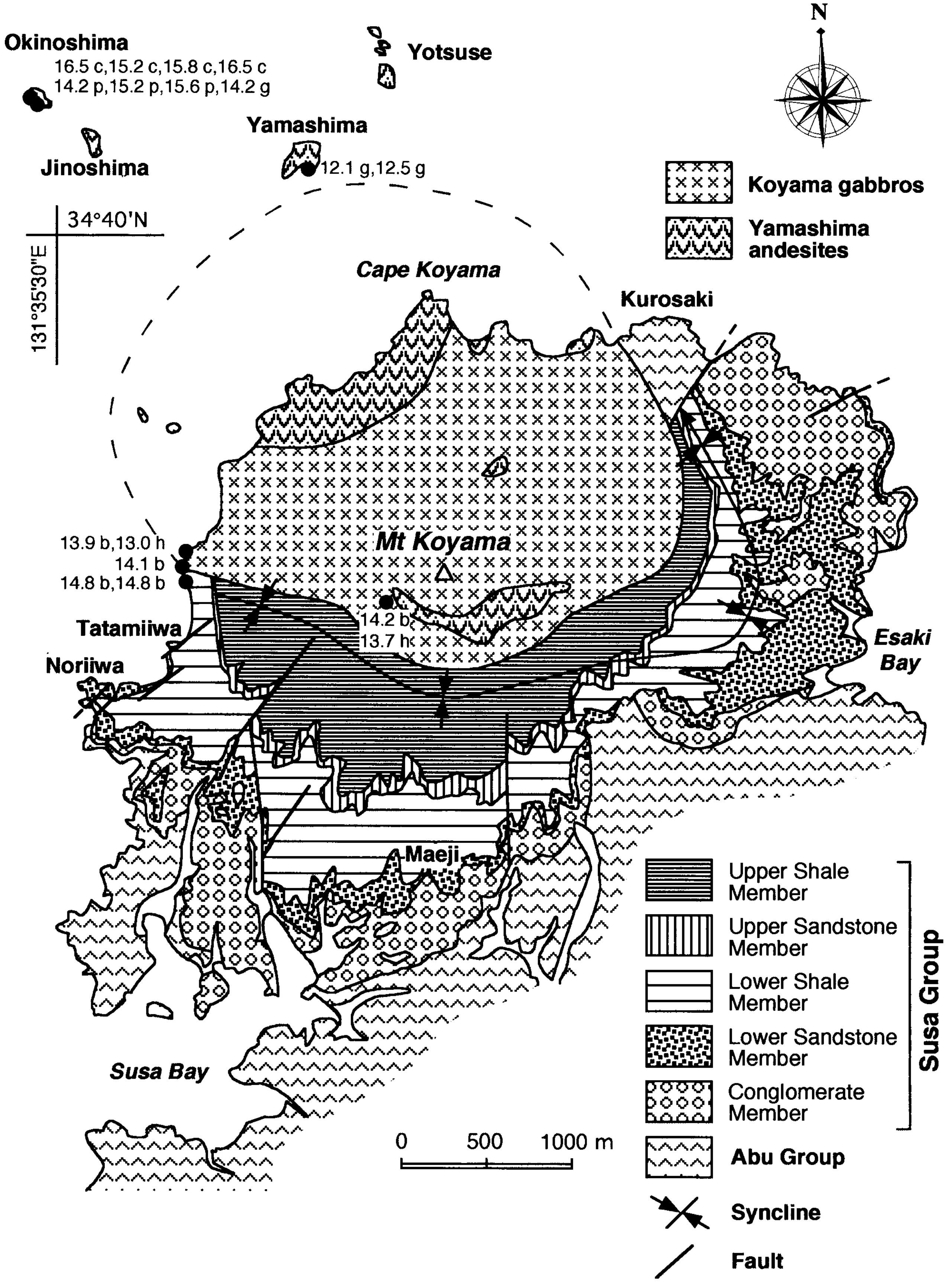

Figure 2. Generalized geological map of the Susa area, Yamaguchi Prefecture, with the locations of the analysed rocks and their K–Ar ages (geological map, compiled from Yamazaki, 1967 and Okamoto *et al.* 1983). Abbreviations: g, groundmass; b, biotite; h, green hornblende; p, plagioclase; c, clinopyroxene.

(Fig. 1). They occur there as the San-in batholith (70–55 Ma) and numerous volcano-plutonic complexes that now appear as cauldron clusters (43–30 Ma, Imaoka *et al.* 1996). These cauldrons occur in a linear arrangement roughly parallel to the Japan Sea coastline (Imaoka *et al.* 1988). Progressive narrowing of the volcanic zone due to migration of the volcanic front from the Pacific Ocean side to the Japan Sea side is thus evident (Murakami, 1974; Ishihara, 1981; Iizumi *et al.* 1985). Igneous activity did not occur in the Chugoku district between 30 and 26 Ma (Imaoka *et al.* 1994). This hiatus may be related to the significant reorganization of Cenozoic plate boundaries in southeast Asia which occurred at about 25 Ma (Hall, 2002).

In contrast, abnormally widespread Late Cenozoic igneous activity occurred in all the in-board terrains. These are subdivided into five igneous zones: (1) Outer zone, (2) Setouchi zone, (3) Sanyo zone, (4) San-in zone, and (5) Oki zone, based on geochemical characteristics (Iwamori, 1991, 1992; Kimura *et al.* 2003; Shinjoe, 1997; Tatsumi & Ishizaka, 1982; Tatsumi *et al.* 2001). Among these zones, Miocene fore-arc magmatism occurred in the Outer, Setouchi and San-in zones. In the Outer zone, S-type and I-type granitoids and related volcanic rocks were produced (Nakada & Takahashi, 1979). The Setouchi zone is characterized by occurrence of high magnesian andesite (Tatsumi & Ishizaka, 1982; Tatsumi *et al.* 2001).

A voluminous sequence of mafic to felsic middle Miocene igneous rocks is distributed in the San-in zone, including the Susa area, indicating intense Miocene volcanic activity (Sugimura & Uyeda, 1973). These igneous rocks fill back-arc submarine rift zones or are intruded into the rift-fill sediments (Miyake, 1994; Kano *et al.* 2002). Neogene strata are also extensively developed in the San-in zone. Their stratigraphy, palaeogeography, palaeoenvironment, palaeobathymetry, and subsidence and uplift history have been reviewed and correlated on the basis of biostratigraphic and radiometric age data (Kano & Yoshida, 1984; Kano & Nakano, 1985; Nomura, 1986; Takayasu *et al.* 1992).

The Susa area is located in the northwestern part of the Japan Sea coast of the Chugoku district (Fig. 1). The geology of the Susa area has been described by Nojima (1941), Yamazaki (1967) and Okamoto *et al.* (1983). Figure 2 shows the geological map of this area according to Okamoto *et al.* (1983), together with our new data for the Yamashima andesites and the Koyama gabbros. Late Cretaceous rhyolitic to rhyodacitic pyroclastic rocks of the Abu Group are widely distributed in the study area, and are unconformably overlain by Miocene sedimentary rocks of the Susa Group. The Yamashima andesites are distributed in the northern part of the area. These Miocene volcanic and sedimentary rock sequences were intruded by the plutonic Koyama gabbros, resulting in contact metamorphism. Numerous small-scale andesite dykes are also observed.

2.a. Susa Group

The Susa Group is composed of neritic marine sediments with a total thickness exceeding 450 m. The succession is divided into five members, from base to top, namely the Conglomerate, Lower Sandstone, Lower Shale, Upper Sandstone and Upper Shale members (Fig. 2). Molluscan fossils contained in the Conglomerate and Lower Sandstone members indicate a Langhian (middle Miocene) age (Okamoto *et al.* 1983). The Susa Group forms a half-basin structure opening toward the northwest. The structure has been locally disturbed by intrusion of the Koyama gabbros around the pluton, where the strata have been slightly folded (Fig. 2). At the intrusive contact north of Tatamiiwa, the boundary between the intrusion and the country rock is sharp and almost vertical, and the sediments have been dragged up and tilted steeply outwards from the intrusion. Sparse inclusions and septa of sandstone and shale occur in the gabbros within several tens of metres of the contact plane.

The Susa Group has been thermally metamorphosed by the Koyama gabbros, forming a contact aureole about 700 metres or more in width. Based on mineral associations in the metamorphosed pelitic rocks, the aureole is divided into three mineral zones: biotite zone (200–650 m), cordierite zone (50–200 m) and orthopyroxene zone (0–50 m) in order of decreasing distance from the contact plane (Suzuki & Nishimura, 1983). Reflectance analysis of coalified phytoclasts in the sedimentary rocks shows concentric isoreflectance surfaces in accord with the distance from the gabbros, suggesting that the contact metamorphism affected the entire Susa Group (Chijiwa *et al.* 1993).

2.b. Yamashima andesites

The term ‘Yamashima andesites’ used in this paper is synonymous with the ‘Yamashima volcanic rocks’ as defined by Imaoka *et al.* (1997). The Yamashima andesites consist of basaltic andesite to andesite hyaloclastites and subaqueous massive lavas, closely associated feeder dykes, and their reworked deposits, suggesting submarine eruption. The andesites are sporadically exposed at Cape Koyama and Mt Koyama as roof pendants on the intrusive body of the Koyama gabbros (Fig. 2). Four offshore islets (Yamashima, Chishima, Yotsuse and Okinoshima) are also composed of the Yamashima andesites. Basaltic andesite dykes similar to the Yamashima andesites intrude the Susa Group (Nojima, 1941) and no andesite clasts occur in the Conglomerate Member of the Susa Group (Okamoto *et al.* 1983). The Yamashima andesite activity thus post-dates deposition of the Susa Group.

The Yamashima andesites contain small amounts of pyroxene (< 5 vol. %) and opaque minerals (< 0.5 %), and varying amounts of plagioclase phenocrysts (0.6–32 %). Aphyric andesites are black and show flow structure, whereas porphyritic varieties are dark grey.

Phenocrysts of plagioclase, augite, orthopyroxene pseudomorphs and titanomagnetite are embedded in a hyalopilitic groundmass containing black glass. Phenocrysts occur either as discrete crystals or as polycrystalline aggregates; the latter represent crystal clots or glomerocrysts.

The Yamashima andesites have also undergone contact metamorphism from the Koyama gabbros. Presence of the assemblage orthopyroxene + clinopyroxene ± hornblende + biotite + labradorite + quartz at Cape Koyama and Mt Koyama suggests a metamorphic temperature of 800–860 °C (Suzuki & Nishimura, 1983). Andesites on the offshore islets of Yamashima, Yotsuse and Chishima have a lower temperature mineral assemblage comprising actinolitic hornblende + epidote + biotite + oligoclase + quartz, whereas those on Okinoshima, about 3 km northeast of Cape Koyama, show no contact metamorphic features.

2.c. Koyama gabbros

The pluton is about 3 km in diameter and is exposed around Mt Koyama, where it intrudes both the Susa Group and the Yamashima andesites. Intrusive relationships between the Yamashima andesites and the gabbros are observed at Cape Koyama, where the gabbros contain andesite clasts several tens of centimetres in diameter. The pluton is lithologically diverse, but is mainly composed of hypersthene–augite gabbros with minor pyroxenite, anorthosite, tonalite, granodiorite and granite. These varieties are believed to have been formed during *in situ* fractional crystallization of silica-saturated basaltic andesite magma with accumulation of plagioclase and pyroxene (Yamazaki, 1967). The crystallization trend of augite indicates successive elevation of P_{H_2O} of the interstitial residual liquid trapped between the crystallizing phases (Yamaguchi, Tomita & Sawada, 1974). The distribution of a high magnetic anomaly, detected by aeromagnetic prospecting, indicates that the pluton is elongated NW–SE (MITI, 1970). The Koyama gabbros have been described as having a laccolith-like occurrence (Sugi & Kuzuna, 1944), a boss-like profile (Yamazaki, 1967) or as representing a bysmalith-type laccolith (Okamoto *et al.* 1983). The boundary between the pluton and the Susa Group at the shore is very steep, and the boundary between the pluton and the Yamashima andesites gradually changes from nearly flat at the summit of Mt Koyama to nearly vertical at Cape Koyama. Therefore, we conclude that the pluton is a bell-jar intrusion similar to a bysmalith, with the difference that the adjacent strata have a domal structure and are not bounded by a steep fault.

2.d. Dykes

A swarm of porphyrite, basaltic andesite and dolerite dykes several tens of centimetres in width intrude the Susa Group, the Yamashima andesites and the Koyama gabbros. Most of these dykes are radially distributed in and around the Koyama gabbros (Okamoto *et al.* 1983). Some dykes are also cut and thermally metamorphosed by the Koyama gabbros, suggesting at least two stages of dyke emplacement (Suzuki & Nishimura, 1983).

3. Previous chronological data

Matsumoto & Itaya (1986) reported whole rock K–Ar ages of contact metamorphic rocks collected from the Susa area, as being 14 to 17 Ma for the metasedimentary rocks and *c.* 15 Ma for metamorphosed Yamashima andesites. They also gave whole rock ages of the gabbro (*c.* 14 Ma) and of dyke rocks (16, 20 and 23 Ma). Two dyke rocks with extremely low potassium contents had anomalously old ages (20 and 23 Ma) inconsistent with the fossil age of the Susa Group. Uto & Ishizuka (1999) reported an Ar–Ar plateau age of 14.41 ± 0.11 Ma and an inverse isochron age of 14.53 ± 0.14 Ma for the gabbro.

There are two different age estimates for the Yamashima andesites, Paleogene (Murakami, 1968) and Miocene (Okamoto *et al.* 1983), based on lithology. The Yamashima andesites have been dated by K–Ar method, giving 8–14 Ma (Imaoka *et al.* 1996) and 5.5 ± 1.3 Ma (K. Uto, unpub. Ph.D. thesis, Univ. of Tokyo, 1989). The younger ages may be due to significant argon loss during hydrothermal alteration. Imaoka *et al.* (1997) reported K–Ar ages of 13.4 and 14.2 Ma for the groundmass of the Yamashima andesites, 14.1 and 14.2 Ma for biotites from the Koyama gabbros, 14.8 Ma for biotite from Susa Group hornfels, and 12.1 Ma for a porphyrite dyke (Table 1). The results suggest that the Yamashima andesites are middle Miocene in age. The XRD analysis of dated Yamashima samples shows minor chloritization (less than 1 % in volume) of orthopyroxene phenocrysts. Consequently, Imaoka *et al.* (1997) considered that this alteration had an insignificant effect on the ages, because orthopyroxene contains less than 0.05 wt % K, and most chlorites were removed during groundmass separation. However, partial alteration of the groundmass may have caused significant radiogenic ^{40}Ar loss, and the samples dated therefore may not have been ideal. Uto & Ishizuka (1999) also carried out Ar–Ar analyses of the Yamashima andesite, giving a total gas age of 8.2 ± 0.4 Ma, and an apparent plateau age of 12.2 ± 0.4 Ma from high temperature gas fractions. The age spectrum of this sample contains extremely young ages in the lower temperature fractions, suggesting that it was significantly altered. Consequently, we collected fresher samples to acquire more precise and reliable K–Ar age data for the Yamashima andesites. In this process, we chose clinopyroxene phenocrysts because they are generally resistant to alteration.

Table 1. K–Ar age data of rocks and minerals from the Susa area, Yamaguchi Prefecture, southwest Japan

No.	Rock	Analysed sample	Size (mesh)	Potassium (wt %)	Radiogenic ^{40}Ar (10^{-8} ccSTP/g)	K–Ar age (Ma)	Non-radiogenic Ar (%)
OK-001*	Yamashima andesite	Groundmass	150–250	1.61 ± 0.03	84.0 ± 2.0	13.4 ± 0.4	50.0
OK-002	Yamashima andesite	Plagioclase	150–250	0.215 ± 0.011	13.0 ± 0.3	15.6 ± 0.9	79.5
OK-002*	Yamashima andesite	Groundmass	80–100	1.35 ± 0.05	74.9 ± 2.3	14.2 ± 0.5	64.9
OK-005	Yamashima andesite	Clinopyroxene	100–200	0.0186 ± 0.0009	1.19 ± 0.09	16.5 ± 1.5	90.9
OK-005	Yamashima andesite	Clinopyroxene	200–300	0.0162 ± 0.0008	0.96 ± 0.08	15.2 ± 1.4	93.7
OK-005	Yamashima andesite	Plagioclase	100–200	0.206 ± 0.010	11.4 ± 0.4	14.2 ± 0.8	78.6
OK-005	Yamashima andesite	Plagioclase	200–300	0.186 ± 0.009	11.0 ± 0.3	15.2 ± 0.9	79.1
OK-005	Yamashima andesite	Groundmass	80–100	1.28 ± 0.03	72.6 ± 0.9	14.6 ± 0.3	66.9
OK-006	Yamashima andesite	Clinopyroxene	100–200	0.0160 ± 0.0008	0.99 ± 0.10	15.8 ± 1.7	92.5
OK-006	Yamashima andesite	Clinopyroxene	200–300	0.0197 ± 0.0010	1.27 ± 0.14	16.5 ± 2.0	93.2
OK-006	Yamashima andesite	Groundmass	80–100	1.11 ± 0.02	59.5 ± 0.5	13.8 ± 0.3	55.0
KG-100	Koyama quartz gabbro	Biotite	80–145	7.38 ± 0.15	407.2 ± 3.2	14.2 ± 0.3	39.8
KG-100	Koyama quartz gabbro	Green hornblende	80–145	0.52 ± 0.01	27.6 ± 0.3	13.7 ± 0.3	53.0
KG-107	Koyama quartz gabbro	Biotite	145–200	6.02 ± 0.12	331.7 ± 3.2	14.1 ± 0.3	47.5
KG-107	Koyama quartz gabbro	Green hornblende	80–145	0.33 ± 0.02	16.9 ± 0.3	13.0 ± 0.7	58.7
79121-21*	Koyama quartz gabbro	Biotite	48–100	7.10 ± 0.14	391.0 ± 4.6	14.1 ± 0.3	14.1
YM-021	Porphyrite dyke	Groundmass	150–250	2.37 ± 0.05	115.6 ± 0.9	12.5 ± 0.3	36.7
YM-021*	Porphyrite dyke	Groundmass	80–100	2.42 ± 0.04	114.2 ± 3.2	12.1 ± 0.6	67.9
79121-18	Pelitic hornfels	Biotite	150–170	5.98 ± 0.12	343.8 ± 2.7	14.8 ± 0.3	33.3
79121-18*	Pelitic hornfels	Biotite	150–170	5.89 ± 0.09	340.9 ± 9.7	14.8 ± 0.5	84.5

* Data from Imaoka *et al.* (1997).

$\lambda_\beta = 4.962 \times 10^{-10}$/y; $\lambda_e = 0.581 \times 10^{-10}$/y; ^{40}K/K = 0.01167 atm %.

4. K–Ar age determination

For precise K–Ar age determination of the Yamashima andesites, the Koyama gabbros and pelitic hornfels of the Susa Group, we collected samples as fresh and as large as possible. The Yamashima andesites were collected from Okinoshima islet, where no contact metamorphic effects are evident. Seven samples were selected for dating after optical microscope examination. Detailed petrographical descriptions are given in Appendix 1.

The selected samples were crushed in a rotary mill and sieved for mineral separation. The sieved fraction was washed in distilled water in an ultrasonic bath to remove any fine particles on grain surfaces, and then dried in an oven at 80 °C. Magnetite was removed with a hand magnet. The magnetite-free fraction was then passed through an isodynamic magnetic separator. Clinopyroxene and plagioclase phenocrysts were separated from the Yamashima andesites, hornblende and biotite from the Koyama gabbros and biotite from the pelitic hornfels of the Susa Group. Heavy liquids were also used to concentrate the minerals. The process was carried out repeatedly to ensure pure mineral separates. Electrostatic separation or tapping was also used for biotite separation. Clinopyroxenes and hornblendes were dipped into hydrochloric acid in a water bath to decompose any associated impurities. This separation technique resulted in clinopyroxene, plagioclase and biotite separates with better than 99 % purity, and 98 % purity for hornblende. Groundmass was also separated from the Yamashima andesites and porphyrite dykes, to compare the ages with those of mineral separates. These groundmass and mineral separates were used for argon analysis. Separate aliquots were further pulverized in an agate mortar for potassium analysis. We prepared and analysed two size fractions of each mineral to test whether the larger grains were foreign to the magma, that is, xenocrysts containing excess argon.

Potassium concentrations in the biotite, hornblende and groundmass fractions were determined by flame photometry using the method described by Nagao *et al.* (1984), whereas those in clinopyroxene and plagioclase were determined with the low-K analytical method developed by Itaya, Doi & Ohira (1996), using the newly designed and constructed ultra-low blank chemical line mentioned above. Multiple runs of a standard (JP-1, 0.00387 wt %; Itaya, Doi & Ohira, 1996) indicate that the error of the potassium analyses is less than 5 %. However, we assign an error of 5 % to the potassium analyses of clinopyroxene and plagioclase.

Argon was analysed at Okayama University of Science using a 15 cm radius sector type mass spectrometer with a single-collector, utilizing isotope dilution and argon-38 spike methods (Itaya *et al.* 1991). Mass discrimination was checked with atmospheric argon several times each day. Specimens wrapped in Al foil were vacuumed out at 150–200 °C for about 24 hours, and argon was then extracted at 1500 °C in an ultra-high vacuum line. Reactive gases were removed using a Ti–Zr scrubber. The decay constants for ^{40}K to ^{40}Ar and ^{40}Ca, and ^{40}K content in potassium used in the age calculation are 0.581×10^{-10}/y, 4.962×10^{-10}/y and 0.0001167, respectively (Steiger & Jäger, 1977). The errors for the ages are at one sigma confidence level.

Fifteen new K–Ar ages for highly purified mineral separates and groundmass are listed in Table 1, together with the age data reported by Imaoka *et al.* (1997). Coexisting clinopyroxene and plagioclase separates from the Yamashima andesite (OK-005) give identical ages within error, 16.5 ± 1.5 Ma (100–200 mesh) and

15.2 ± 1.4 Ma (200–300 mesh), and 14.2 ± 0.8 Ma (100–200 mesh) and 15.2 ± 0.9 Ma (200–300 mesh), respectively. These ages are the same as those from two other samples, 15.8 ± 1.7 Ma (100–200 mesh) and 16.5 ± 2.0 Ma (200–300 mesh) for clinopyroxene from OK-006, and 15.6 ± 0.9 Ma (150–250 mesh) for plagioclase from OK-002. The groundmass and minerals in each sample have similar ages. Coexisting biotite and hornblende from the Koyama gabbros give similar ages within analytical error, 14.2 ± 0.3 Ma and 13.7 ± 0.3 Ma for KG-100, and 14.1 ± 0.3 Ma and 13.0 ± 0.7 Ma for KG-107, respectively. They are slightly younger than the Yamashima andesites. The porphyrite dyke yields a groundmass age of 12.5 ± 0.3 Ma, which is the same as the porphyrite dyke groundmass age (12.1 ± 0.3 Ma) reported by Imaoka *et al.* (1997). Both ages are significantly younger than the Yamashima andesites. Biotite from the pelitic hornfels gives an age of 14.8 ± 0.3 Ma, identical to the biotite age (14.8 ± 0.4 Ma) determined by lmaoka *et al.* (1997).

5. Discussion

5.a. Implications for K–Ar dating of clinopyroxene

K–Ar age determinations aimed at solving geological and petrological problems have commonly been carried out on K-rich minerals such as biotite, muscovite and hornblende, and rarely on low-K minerals such as pyroxene and plagioclase. Hart (1961) and McDougall (1961, 1963) tried to date pyroxene by the K–Ar method, and showed that pyroxene was suitable for K–Ar dating of igneous rocks emplaced at high levels in the crust, because only very small amounts of excess argon are present and the mineral has excellent argon retention. Clinopyroxene is also chemically resistant to hydrothermal alteration. However, following detection of excess argon in pyroxenes in other studies (Hart & Dodd, 1962; Lovering & Richards, 1964; McDougall & Green, 1964; Allsopp, 1965), the K–Ar method has rarely been applied to pyroxenes. This is due to difficulties in obtaining precise potassium analyses at very low abundances, and in evaluating excess argon content.

The flame photometric method developed by Itaya, Doi & Ohira (1996) overcomes the difficulty of analysing rocks and minerals with very low potassium contents. They applied their method to igneous and metamorphic hornblende from ophiolite complexes, and to coexisting hornblende and clinopyroxene from Cretaceous volcanic rocks. The K–Ar ages they obtained for hornblendes and rocks were compatible with the geology of the host rocks and other chronological data. Their results suggest that conventional K–Ar methodology is also applicable to the geochronology of ophiolite complexes in orogenic belts.

The problem of ‘excess argon’ cannot easily be discounted, although it is easily detected. Recent advances help avoid the problem of excess argon and allow adequate dating of geological materials (Itaya & Okada, 1995). For example, submarine pillow basalts formed on the deep sea floor have extremely high excess argon contents in their quenched rims (e.g. Funkhouser, Fisher & Bonatti, 1968; Dalrymple & Moore, 1968). In this case, the central cores of pillows, which cool slower than the rims, may give reasonable ages. McDougall, Verwoerd & Chevallier (2001) demonstrated that excess argon may even be widespread in subaerially erupted lavas, such as on Marion Island. Significant excess argon has also been detected in mantle xenoliths contained in volcanic rocks (e.g. Nagao & Takahashi, 1993) and in extremely large plagioclase phenocrysts (Damon, Laughlin & Percious, 1967; Dalrymple, 1969; Itaya *et al.* 1984). This shows that excess argon exists in exotic materials in volcanic rocks. The Yamashima andesites examined apparently contain no exotic materials such as xenoliths or xenocrysts, nor extremely large phenocrysts. Electron microprobe analyses of clinopyroxene phenocrysts show little variation in Fe content (see Appendix 1), and they plot on the crystallization trend of cognate Koyama gabbros, as described further in Section 5.b. Limited ranges of Fe contents are common in calc-alkaline volcanic suites (e.g. Fodor, 1971), reflecting clinopyroxene crystallizing at high oxygen fugacity in hydrous magmatic conditions. Therefore, the clinopyroxene phenocrysts examined here were in equilibrium with the magma from which they crystallized (Imaoka & Nakashima, 1996). The age results show that two fractions had the same ages, which were also consistent with those of plagioclase and groundmass in each sample (Table 1).

Although clinopyroxene contains no structural sites suitable for potassium, this element can occur in crystal dislocations and defect structures (Hart & Dodd, 1962) or in small fluid or glass inclusions within crystals (Rama, Hart & Roedder, 1965; McDougall, Polach & Stipp, 1969). If so, clinopyroxene crystallized in equilibrium with a magma should have initial argon trapped in the same site as the potassium, and also have the same initial argon systematics as the magma. If the magma contained significant excess argon, such as in deep submarine pillow basalts, then the trapped argon will have a significant effect on clinopyroxene K–Ar dating because of the extremely low potassium content of that mineral. The fact that coexisting clinopyroxene and plagioclase phenocrysts and the groundmass of each sample have similar ages suggests that Yamashima andesite magma did not contain significant amounts of excess argon. This conclusion is supported by the isochron diagram (Fig. 3), in which there is a good correlation over a relatively wide range of $^{40}K/^{36}Ar$ ratio. The clinopyroxene and plagioclase phenocryst data define an isochron age of 14.7 ± 0.9 (1σ) Ma

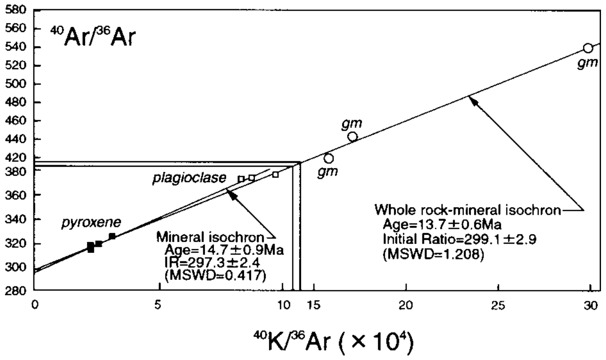

Figure 3. Isochron diagram for the Yamashima andesites. Details of the mathematical treatment of the isochron calculations have been given by York (1966).

with an initial ^{40}Ar/^{36}Ar ratio of 297.3 ± 2.4 (1σ), which coincides with that of atmospheric argon. This isochron has a MSWD of 0.417, which is small enough to ascertain its significance. The arithmetic mean of the mineral ages for the Yamashima andesite is 15.6 ± 0.8 Ma, although the isochron age is 14.7 ± 0.9 (1σ) Ma. This shows the very high sensitivity of the derived age to the estimated isotopic composition of the trapped argon. Combining the clinopyroxene, plagioclase and groundmass data yields a whole rock-mineral isochron age of 13.7 ± 0.6 Ma, slightly younger than that from the clinopyroxene and plagioclase phenocrysts alone (Fig. 3). The initial ^{40}Ar/^{36}Ar ratio of the whole rock-mineral isochron is 299.1 ± 2.9, also slightly larger than that of the clinopyroxene–plagioclase mineral isochron. This slight difference is most likely due to partial argon loss from groundmass materials by alteration, suggesting that the clinopyroxenes provide more reliable ages. This also suggests that the magma of the Yamashima andesites did not have significant amounts of excess argon. Newly obtained and previously reported K–Ar age data and fossil ages of the Susa Group are summarized in Figure 4.

5.b. Middle Miocene submarine volcano-plutonic complex and its tectonic significance

Figure 5 shows our model for the formation of the middle Miocene submarine volcano-plutonic complex

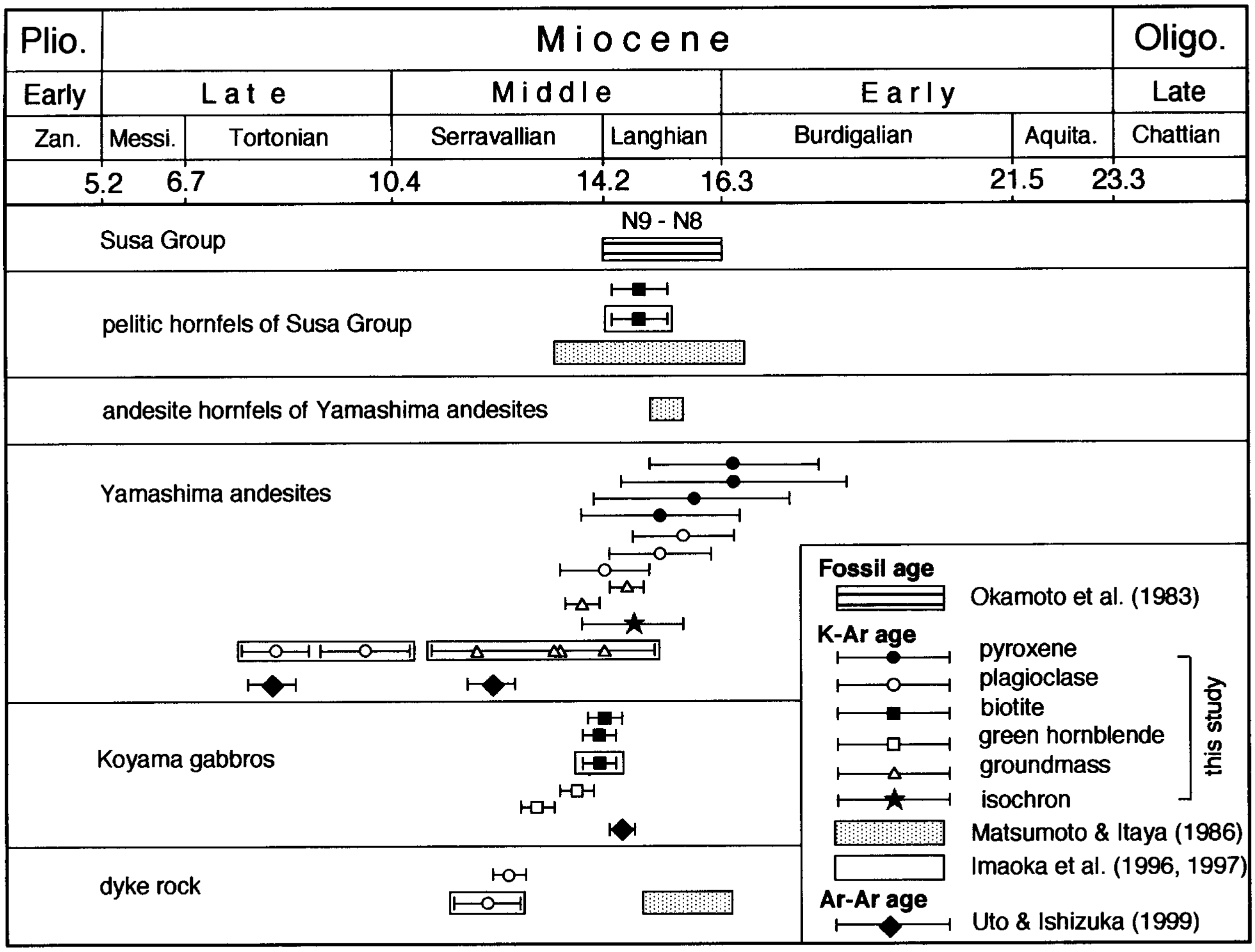

Figure 4. Summary of fossil and K–Ar ages in the Susa area, Yamaguchi Prefecture, southwest Japan. The estimated ages for the stage boundaries are derived from Harland *et al.* (1989).

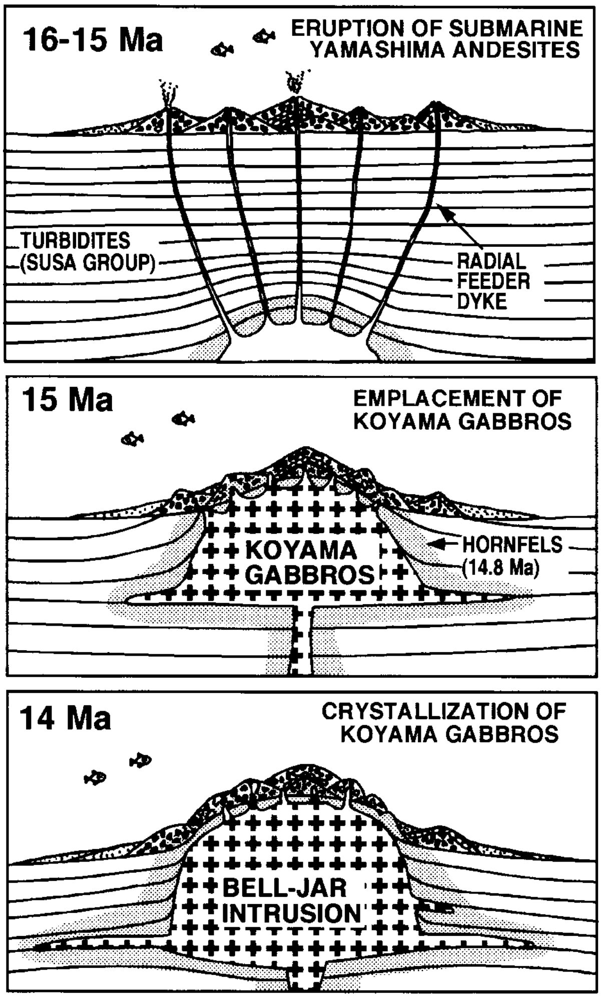

Figure 5. Schematic representation of the formation of the Miocene submarine volcano-plutonic complex in the Susa area, Yamaguchi Prefecture, southwest Japan.

in the Susa area. In middle Miocene time, the Susa area was in a neritic marine environment (Okamoto *et al.* 1983). Sedimentological and palaeontological studies of the Susa Group indicate that the environment changed to tidal or sublittoral conditions in the early Langhian of the middle Miocene (Blow's foraminiferal zone N.8), and conglomerates and sandstones were deposited, marking the beginning of marine transgression. Transgression progressed rapidly in the late Langhian (N.9), leading to deposition of a thick black shale succession, sandstone–mudstone turbidite sequences, and the large-scale channel-fill deposits which are now exposed in the fault-scarp on the southwest coast of Tatamiiwa. This abrupt deepening of the sedimentary basin proceeded together with the submarine volcanism that produced the Yamashima andesites at 15–16 Ma, as suggested by our new K–Ar ages and field evidence. Imaoka *et al.* (1997) reported that the Yamashima andesites are basaltic andesite to andesite hyaloclastites and subaqueous massive lavas, associated feeder dykes, and their reworked deposits. The Koyama gabbros intruded the Susa Group and the Yamashima andesites, and caused contact metamorphism in both. K–Ar biotite ages of the hornfels (14.8 ± 3.0 Ma) and the Koyama gabbros suggest that the gabbros were emplaced just after the Yamashima andesitic volcanism.

As noted in Section 2.c, our field observations suggest that the shape of the Koyama gabbros is a bell-jar intrusion similar in form to a steep-sided laccolith or bysmalith. This dome-shaped structure may be formed when basaltic magma intrudes unconsolidated low-density sediments, as suggested by the observations of Lonsdale & Lawver (1980) and Macdonald *et al.* (1979), who studied the Guaymas Basin and Tamayo transform valley in the Gulf of California, respectively. A high rate of magma supply and progressive increase in viscosity during injection are favourable conditions for the growth of these steep-sided intrusions (Hunt, Averill & Miller, 1953). Field and textural evidence in the Susa area, combined with the K–Ar data, indicates that the pluton was intruded into a high level in the crust at about 15 Ma and cooled down to 300 °C, the closure temperature of biotite (Dodson & McClelland-Brown, 1985), at about 14 Ma. This suggests that after emplacement, cooling and fractional crystallization took about one million years to form the diverse rocks found in the pluton. Small-scale porphyrite dykes were then intruded in the area until about 13 Ma.

With respect to the genetic relationships between the Koyama gabbros and the Yamashima andesites, Nojima (1941) and Oji & Oji (1965) suggested that the andesites were cognate to the gabbros, based on field evidence and similar whole rock chemistry. Imaoka & Nakashima (1996) considered that Al_2O_3 and CaO variations in the Yamashima andesites and Koyama gabbros could be explained by plagioclase accumulation in the upper part of the magma chamber, to which plagioclase phenocrysts had floated (e.g. Marsh, 1988). They thus supported the view of Yamazaki (1967) that plagioclase flotation and settling of olivine and pyroxene were important processes in the Koyama–Yamashima magma chamber. These features indicate that the Yamashima andesites, the Koyama gabbros, and the syn-plutonic dykes are all of cognate origin, and that the Koyama gabbros as presently exposed represent the dissected subvolcanic structure of a volcano-plutonic complex.

In summary, geological events in the Susa area occurred in a short period between 16 and 13 Ma. Initial deposits of the Susa Group, shallow-marine conglomerates and sandstones, coincided with initiation of a marine transgression. Subsequent deposition of thick black shales, sandstone–mudstone turbidite sequences, and large-scale channel-fill deposits indicate abrupt deepening in the area at 16–15 Ma, probably due to rapid subsidence and marine transgression culminating in and around the Japan Sea (Yamaji, 1990; Ingle, 1992; Kano *et al.* 2002). Coeval igneous activity formed the submarine volcano-plutonic complex. These events of rapid tectonic subsidence and andesitic magmatism were synchronous with the opening of the Japan Sea and the associated clockwise rotation of the southwest Japan arc sliver. The complex was likely produced in association with rifting of the Yamato Basin and the adjacent area (Yamaji, 1990; Miyake, 1994). The volcano-plutonic complex formed in the Susa area thus records a unique tectonic situation. Similar settings were widespread along the present Japan Sea coast in the Japan Arc, that is, the Shimane Peninsula (Miyake, 1994; Kano, 1996), Tango Peninsula (Furuyama *et al.* 1997), Noto Peninsula (Kano *et al.* 2002) and Sakhalin Island, Russia (Kano *et al.* 2000). This tectono-magmatism is correlated with that of the Yamato and Japan basins of the Japan Sea, and it characterizes back-arc spreading. Our K–Ar results further confirm the exact timing and character of a specific tectonic event in the western part of the southwest Japan arc.

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Appendix 1. Description of dated rock specimens

OK-002 Yamashima andesites (131°35′25″ E, 34°40′27″ N)

Basaltic andesite from Okinoshima islet. The rock is dark grey, is unaffected by contact metamorphism, and no xenoliths or xenocrysts are observed. It shows distinct porphyritic texture, with phenocrysts of plagioclase, clinopyroxene, orthopyroxene (pseudomorphs) and titanomagnetite embedded in a hyalopilitic groundmass with black glass. Plagioclase phenocrysts (core An_{82-87}, rim An_{60-72}) up to 1.6 mm in length are euhedral, lath-shaped, and show glomeroporphyritic texture. Groundmass plagioclase is invariably slightly more sodic (An_{46-65}) than phenocrystic plagioclase. Clinopyroxene ($Ca_{39}Mg_{42}Fe_{19}$–$Ca_{36}Mg_{44}Fe_{20}$) is up to 1.2 mm in length and is euhedral to anhedral. Orthopyroxene (0.9 vol.%) is completely replaced by chlorite, although all other minerals are very fresh. Euhedral to subhedral titanomagnetite microphenocrysts (16–29 mol.% ulvospinel mol.) up to 0.6 mm occur as discrete grains. Some magnetites have ilmenite lamellae or show granular intergrowth with ilmenite. Peripheral parts of magnetites are often altered to maghemite. In the groundmass, magnetite usually occurs as euhedral grains a few to several tens of microns in diameter.

OK-005, OK-006 Yamashima andesites (131°35′25″ E, 34°40′26″ N)

Basaltic andesite from Okinoshima islet. No xenoliths or xenocrysts are observed. Petrographic characters strongly resemble the above sample (OK-002).

KG-100 Koyama quartz gabbro (131°36′45″ E, 34°38′45″ N)

Biotite and green hornblende (80–145 mesh) were separated from this quartz gabbro, collected from the southwestern slopes of Mt Koyama. Biotite separation was perfect, but we could not remove all minute grains of clinopyroxene attached to the hornblende. The rock is mainly composed of plagioclase (71.1 vol. %), quartz (10.1 %), clinopyroxene (10.0 %), orthopyroxene (0.8 %), inverted pigeonite (1.1 %), green hornblende (2.9 %), biotite (3.0 %) and small amounts of Fe–Ti oxide minerals (0.8 %) and K-feldspar (0.2 %). Plagioclase (1.0–2.0 mm) shows oscillatory zoning, and albite and Carlsbad twins. Quartz (0.1–1.3 mm) and K-feldspar ($<$ 0.3 mm) occur as interstitial fillings. Clinopyroxene (0.1–2.0 mm) and orthopyroxene occur as subhedral grains. Inverted pigeonite is often enclosed by clinopyroxene (augite) and contains herring bone-like augite lamellae. Green hornblende (0.1–0.3 mm) occurs as anhedral vermicular crystals which replace clinopyroxene or form reaction rims on that mineral. Biotite (0.1–0.3 mm) encloses corroded magnetite and ilmenite, and occurs as reaction rims around clinopyroxene and orthopyroxene, and as discrete subhedral to euhedral grains. Apatite and zircon occur as accessory minerals.

KG-107 Koyama quartz gabbro (131°36′02″ E, 34°39′00″ N)

Biotite (145–200 mesh size) and green hornblende (48–145 mesh size) were separated from the quartz gabbro collected north of Tatamiiwa (Fig. 2). Separation of the green hornblende was perfect, but we could not remove all grains of green hornblende attached to biotite. The rock is mainly composed of plagioclase (55.1 vol. %), quartz (5.5 %), clinopyroxene (16.0 %), orthopyroxene (7.3 %), green hornblende (15.8 %), biotite (0.2 %), Fe–Ti oxide minerals (0.1 %), and very small amounts of anhedral K-feldspar. Plagioclase (0.4–1.2 mm) shows oscillatory zoning and albite twin and Carlsbad twins. Quartz crystals up to 0.4 mm across occur as interstitial fillings. Clinopyroxene (0.2–1.7 mm) and orthopyroxene (0.4–1.2 mm) occur as subhedral to anhedral grains. Green hornblende (0.2–1.0 mm) occurs as reaction rims or patchy or vermicular replacements of pyroxenes, and as interstitial grains. It also occurs as anhedral interstitial grains among plagioclase. Biotite (0.05–0.3 mm) occurs as reaction rims around green hornblende and as anhedral interstitial grains. Apatite and zircon occur as accessory minerals.

YM-021 Porphyrite dyke (131°36′27″ E, 34°40′14″ N)

This grey porphyrite dyke intrudes Yamashima andesites, and was collected from Yamashima islet. Texture is porphyritic, with a well-crystallized groundmass. No sign of contact metamorphism is observed under the microscope, and no xenoliths or xenocrysts are present. All minerals except orthopyroxene are very fresh. The phenocrysts are plagioclase, clinopyroxene, orthopyroxene (pseudomorphs) and titanomagnetite. Euhedral to subhedral plagioclase phenocrysts (core An_{71-82}) 0.4 to 1.5 mm in length often show glomeroporphyritic texture, as does euhedral–subhedral 0.3 to 1.5 mm clinopyroxene ($Ca_{42}Mg_{43}Fe_{15}$–$Ca_{41}Mg_{41}Fe_{18}$). The cpx is slightly richer in Ca and poorer in Fe than that in the basaltic andesites. Orthopyroxene is partially replaced by chlorite. Anhedral titanomagnetite microphenocrysts are up to 0.7 mm in diameter, and contain 25–26 mol. % ulvospinel. Groundmass plagioclase (An_{16-39}) is euhedral.

79121–18 Pelitic hornfels (131°35′59″ E, 34°38′55″ N)

Biotite (150–170 mesh) was separated from orthopyroxene zone pelitic hornfels collected north of Tatamiiwa, 20 cm from the contact between the Koyama gabbros and the Susa Group. The rock shows distinct granoblastic texture under the microscope. Average grain size of quartz and plagioclase is 0.1 mm, and that of biotite is 0.2 mm. The hornfels is mainly composed of fine-grained recrystallized orthopyroxene, cordierite, andalusite, fibrolite, biotite, K-feldspar, plagioclase, quartz, and small amounts of apatite and tourmaline.