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Origin of enclaves and sulfur in the Inada Granite, Tsukuba District, central Japan

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Abstract: Many enclaves with various sizes have been known to occur in granite quarries at Inada where Inada building stones are mined. They are mostly hornfels of fine-grained sandstones with some slates and limestones. None of them appears to be restites but derived from the roof sedimentary rocks at high level of the granitoid emplacement. Thus, they can be called xenolith. Unique pyrrhotite-rich ball enclaves were also found in the Inada granite quarries. They yielded negative $\delta^{34}S$ values around -10 permil, and are considered to be sulfides from not a deep source but shallow source of the pelitic wall rocks, which were originally concentrated in euxenic sediments of the basement accretionary complex. Because of sulfur-rich nature, the enclave became melt in the magma chamber after the entrapment, and thus became spherical forms.

1. Introduction

Many granite quarries are located in an area to the north of Tsukubasan (Mt. Tsukuba) District (Fig. 1). The granite called Inada stone has been mined at Inada, northern edge of the granitic terrane. This locality is well known to produce biotite granite that contains abundantly large blocks of sedimentary rocks, which have been used for gardening (Plate 1A) and sometimes for furniture. In addition, there occur variety of small enclaves including crystalline limestone (Sasada *et al*., 1982), skarn and sulfide ores.

Recently, ball-shaped enclaves containing abundant pyrrhotite were discovered in the open pits of Nakanogumi-Maeyama and Takata-Nishizawa. This kind of enclaves are very rare; only locality so far known in Japan is Manguro in the central part of S-type ilmenite-series Osumi pluton of Miocene age in the Outer Zone of Southwest Japan, where most of the enclaves were interpreted originated in metalliferous shale but some derived from mafic igneous source rock (Ishihara, 1982).

In this short article, we report the mode of occurrence of these enclaves and sulfur isotopic ratios of sulfides occurring in and around the quarries, and consider origin of these enclaves and contained sulfides.

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2. Geologic Outline

The Tsukuba District is located at the eastern end of the ilmenite-series granitic terrain of the Inner Zone of Southwest Japan, and is underlain by late Jurassic-early Cretaceous accretionary complex and late Cretaceous - Paleogene plutonic rocks. The sedimentary rocks were regionally metamorphosed during the Cretaceous time to low P/T type metamorphic rocks in the southern area (Miyazaki *et al*., 1996), but non-metamorphosed in the northern area (Fig. 1).

These sedimentary rocks are classified into Yamizo Supergroup whose proposed subdivisions are summarized by MITI (1986) as Kasama, Kunimi, Takatori and Ayuta Groups from south (lower member) to north (upper member). The Kasama Group occurs in the Iwama area of the studied district, while the Kunimi Group is exposed in the northern-end of the studied district (Fig. 1). The Kasama Group is alternation of sandstone and shale with intercalated chert, limestone and basalt, while the Kunimi Group is rather monotonous alternation of sandstone and shale, but predominantly sandstone.

These sedimentary rocks have been intruded by late Cretaceous gabbroid (75 Ma) and by late Cretaceous-Paleogene granitoids (63-53 Ma, Miyazaki *et al*., 1996). One large cassiterite-bearing wolframite quartz vein deposit and many small showings of tungsten

Keywords: Tsukuba, late Cretaceous, ilmenite-series granite, skarn, xenolith, pyrrhotite, $\delta^{34}S$ value

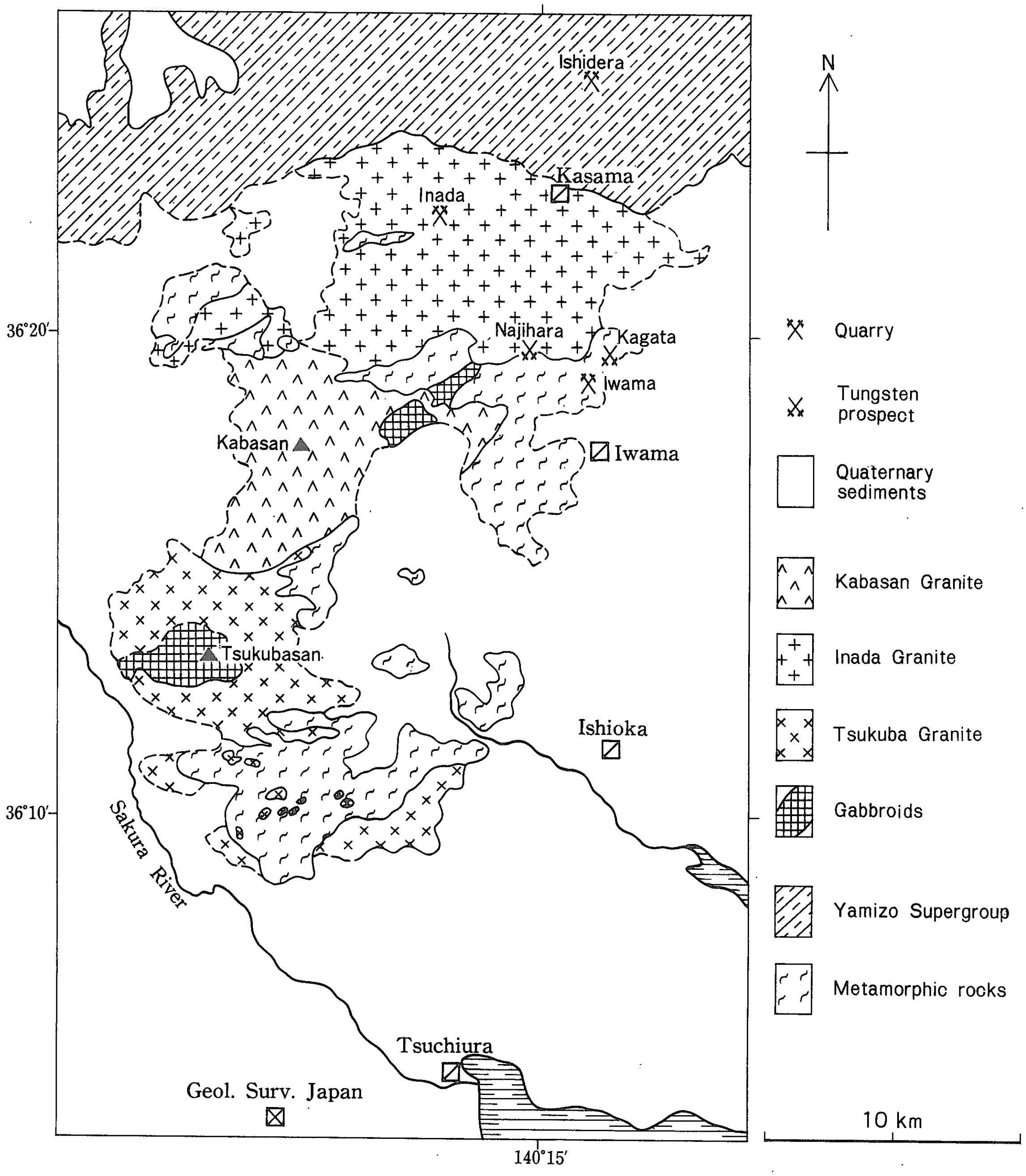

Fig. 1 Distribution of plutonic rocks (Miyazaki et al., 1996) and locality of the studied quarries in the Tsukuba area. Quarry symbol with Inada includes Takata-Iwakura and Takata-Nishizawa pits, and Nakanogumi-Maeyama pit; those with Iwama includes Iwama-Nagasawa and Iwama-Kamigo pits.

have been known related to hidden granitic bodies to the northeast and east of the studied district (e.g., Ogasawara *et al.*, 1993). The granitoids were divided into seven intrusive bodies by Takahashi (1982), but Miyazaki et al. (1996) compiled them three types as Tsukuba, Inada and Kabasan Granites, based upon their intrusive relations and rock facies.

The Tsukuba Granite is exposed in the southern part of the Tsukuba district (Fig. 1). It is either foliated or massive in texture and is concordant partly to the metamorphic wall-rocks, and consists of porphyritic biotite granodiorite, foliated biotite tonalite, muscovite-biotite monzogranite (hereafter abbreviated as granite) and fine-grained granite. Cordierite-bearing orbicular rock, which is designated as Prefectural Monument, is seen in the porphyritic granodiorite.

The Inada Granite occurs in the northern part of the district and intrudes into discordantly metamorphic and non-metamorphic sedimentary rocks, and gives them thermal metamorphism. It is massive in texture, consisting of coarse-grained hornblende-bearing biotite granite, medium-grained hornblende-biotite granodiorite and fine-grained muscovite-biotite granite.

The Kabasan Granite is distributed in the middle part of the district. It intrudes partly into the Tsukuba and Inada Granites but is partly gradual to both the granites (Miyazaki *et al*., 1996). It is composed mainly of medium-grained biotite granite and fine-grained muscovite-bearing biotite granite.

3. Enclaves

Quarries at Inada are unusual among Japanese quarries having large blocks of the sedimentary rocks. These rocks occur as sheet-like body parallel to the roof boundary (Fig. 2), thus can be considered detached roof-pendent. Blocky xenoliths are usually fine-grained sandstone hornfels, and some alternated sandstone and shale hornfels of the Kunimi Group of the Yamizo Supergroup. These sedimentary blocks are angular in shape (Plate 1 B), and have no hybrid phase between the hornfels and host granite. Smaller xenoliths of psammitic (Plate 1 C) and pelitic (Plate 1 D) compositions are also angular in shape. Thus, these enclaves were trapped in the Inada magma chamber at the final, low temperature stage of the emplacement, and can better be called xenolith.

In rare occasion, metamorphic enclaves are found (Plate 1 E). This enclave is intruded by fine-grained granite dikelets (1, Plate 1 E), which has syenitic rim; then the dikelets were deformed. Both rocks were intruded by leucogranite (2, Plate 1 E) and biotite granite (3, Plate 1 E). Thus, this rock is different in the source rocks and may be originated in Wagakunisan metamorphic rocks or other metamorphic rocks hidden at depth.

There occur also calcic xenoliths and sulfide-rich enclaves in the quarries. Carbonate beds are absent in the Kumini Group but some occur in the Kasama Group. These xenoliths may have been originated in the constituents of the Kasama Group. The calcic xenoliths are composed of crystalline calcite with some dolomite and a large cavity which may have formed by loss of the carbonates along cracks after solidification of the host granitic magma (Sasada, 1991).

Some of the calcic xenoliths have been converted to skarn minerals. At one occasion of the Takata-Nishizawa pit, calcite and dolomite are surrounded by diopside and then by phlogopite (Plate 2 A). In rare occasion, carbonate minerals have been completely converted to garnet, epidote and vesuvianite. The host granite around the xenolith was depleted in iron and magnesium, which were probably migrated to the skarn core, thus forming plagioclase and quartz rock (Plate 2 B). These skarn xenoliths are found in fresh granite and have no connection to fractures and aplitic dikes; thus they are considered to have formed by local interaction between the carbonate xenolith and host granitic magmas, when the xenolith was trapped.

Sulfide-ball enclaves occur together with fragmental sedimentary xenoliths (Plate 1 D). They are seen more commonly as aggregates of small balls gathering in a large ball up to 30 cm in diameter (Plate 1 F). The fragments of sedimentary metamorphic rocks can be seen up to 10 volume percent. Sulfide-rich fragments tend to have small ball shapes. The ball-rich part was measured to have a weak magnetic susceptibility up to 3.7×10^{-3} SI st Nishizawa pit, and up to 0.6×10^{-3} SI at the Maeyama pit, while the host fresh biotite granite has $0.06\text{–}0.09 \times 10^{-3}$ SI by KT-5

Fig. 2 Sketch of M. Sasada; the detached roof-pendent at the upper part of Takata-Iwakura pit in 1981 (left). Insert picture taken in 1991 by S. Ishihara; the rock wall was partly coated by black organic materials.

magnetic susceptibility meter.

Under the microscope, the balls are composed of opaque minerals, brown biotite, bluish green amphibole, quartz and feldspars. Feldspars occur in granular texture which appears to be relict texture of original sandstone. They are mostly plagioclase but some have been converted to K-feldspar during the recrystallization. Biotite and amphibole occur everywhere; biotite is replaced by amphibole in some occasions. Coarse-grained portion consists mainly of large crystals of quartz and opaque mineral. The opaque minerals are mostly (ca. 90 vol. %) pyrrhotite and partly (up to10 vol. %) chalcopyrite. Pyrrhotite occurs as irregularly rounded form consisting of finer strongly anisotropic crystals. The mineral is hexagonal variety by x-ray diffraction. Chalcopyrite occurs with mutual boundary with the pyrrhotites and is seen within the crystals as island or margin of the pyrrhotites.

4. Sulfur Isotopic Ratio

Sulfur isotopic ratio, δ^{34}S CDT (‰), was determined on six selected samples by the methods described by Tazawa and Sasaki (1991). The sample localities are shown in Figure 1 and the results are listed in Table 1. Pyrrhotite contained in a small amount in aplitic dikelet cutting the main facies of the Inada Granite gave −2.4 permil, which is close to −1.1 permil given for the Inada Granite by Tazawa and Sasaki (1991).

Pyrrhotite is the major sulfide mineral in the ball-shaped and gneissic enclaves (Plate 2 C, 2 D). They yielded nearly identical values of −10.6 and −10.0 permil, respectively.

Arsenopyrite-calcite-quartz breccia vein was discovered in the sedimentary rocks of the Kunimi Group in a quarry for crushed stones at Ishidera. This arsenopyrite gave +0.8 permil. The same mineral from arsenopyrite-quartz vein cutting black shale of the Kasama Group at Iwama-Nagasawa pit has similar value of −0.3 permil. Pyrrhotite-quartz lens occurring in skarn with black slate layer of Iwama-Kamigo pit was determined to have −8.0 permil.

There are two tungsten prospects close to the Iwama quarry. Their composite sulfides were identified to have −1.6 permil at Kagata skarn deposit and −3.5 permil at Najihara prospect (Ishihara *et al.*, 1992). At Takatori mine, about 13 km due north of Kasama, wolframite-quartz veins are hosted in the Takatori Group. Their composite sulfides have an average δ^{34}S value of −3.6 permil (Tazawa and Sasaki, 1991).

5. Discussion on the Source of Sulfur

The analysis IN01 (−2.4 ‰) may be representative for the rock δ^{34}S values for the Inada Granite, because this is unaltered aplitic biotite granite and no sedimentary enclaves were seen in and around the sample locality site. Tazawa and Sasaki (1991) reported an average δ^{34}S value of −1.1 permil for the Inada Granite and 0 permil for the sedimentary rocks of the Yamizo Supergroup, and considered sulfur of the granitoid originated in the bsement sedimentary rocks. Thus, the δ^{34}S value around −1 to −2 permil is considered as inherent value for the Inada Granite.

Sulfur isotopic ratios of arsenopyrite and other sulfides occurring with quartz veins and skarn orebodies at Takatori, Kagata, Najihara and Ishidera, are close to or slightly lower than the values for granitoids. Thus, sulfur of these sulfides is considered originated mainly in hidden cupolas of the Inada Granite.

On the other hand, the δ^{34}S values around −10 permil of the pyrrhotite balls are much depleted in ^{34}S and should be originated in different source rocks. Such ^{34}S-depleted rocks are usually found in S-rich euxenic sediments of a closed basin (e.g., −24.8 ‰, an average of claystone and siltstone of the Upper Permian Toyoma Slate, Endo et al., 1973) and also in black shale layers in accretionary terrane (e.g., −17.4 ‰, an average of claystone and siltstone of the Upper Cretaceous Shimanto Supergroup, Sasaki and Ishihara, 1979).

Here in the Tsukuba District, low δ^{34}S values are generally found in rocks of the Kasama Group (Tazawa and Sasaki, 1991). In our study, the lowest δ^{34}S value (−8 ‰) was observed in pyrrhotite lens occurring concordantly with black shale and skarnized fine-grained sandstones at the Iwama-Kamigo quarry (IN07), which is quite different from the values of the Inada Granite. Thus, sulfur of the pyrrhotite lens is considered originated in sedimentary sulfur of the Kasama Group, and concentrated in the pyrrhotite layer or lens during the contact metamorphism.

Microscopic observation also supports this interpre-

Table 1 Sulfur isotopic ratios of sulfides from granitoids and sediments.

Nos.	Locality	Rock type	Minerals	δ^{34}S(‰)
IN01	Takata-Iwakura pit	Aplite dikelet cutting Inada Granite	Pyrrhotite	-2.4
IN02	Takata-Nishizawa pit	Gneissic xenolith in Inada Granite	ditto	-10.6
IN03	Nakanogumi-Maeyama pit	Ball-shaped enclave in Inada Granite	ditto	-10.0
IN05	Kasama, Ishidera	Sulfides-calcite-quartz filled breccia in sandstone and shale	Arsenopyrite	+0.8
IN06	Iwama-Nagasawa pit	Arsenopyrite-quartz vein in black shale	ditto	-0.3
IN07	Iwama-Kamigo pit	Chalcopyrite-bearing pyrrhotite-biotite-quartz lens in sandstone and shale	Pyrrhotite	-8.0

(A. Sasaki analyst)

tation. Under the microscope, this rock is composed mostly of quartz, biotite and pyrrhotite with small amount of calcic amphibole, feldspars and chalcopyrite; these constituents are very similar to those of the pyrrhotite ball enclaves. Thus, the two rocks appear to have had a common source.

The pyrrhotite ball enclaves may be originally sulfur-enriched sediments, then converted to the biotite-pyrrhotite hornfels by heat provided by hidden granitic cupola, and brecciated and dropped into magma chamber of the Inada Granite at a high level. Because it contained much iron sulfide, it became soon sulfide melt and crystallized in the solidifying crystal mash of the host Inada Granite. Thus, the pyrrhotite is concentrated in an orbicular shape, in contrast to the other clastic sediments kept their fragmental shapes in the magma chamber. The original $\delta^{34}S$ signature is also preserved in the balls.

6. Conclusions

Sulfide-ball enclaves found in the Inada granites of the Takata and Nakanogumi quarries at Inada are considered to have not a deep igneous source but a shallow sedimentary origin. They were originally concentrated in euxenic environment of the Kasama Group, and thermally metamorphosed by intrusion of the Inada granitic magma, then trapped into the granite itself as xenolith.

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日本中央部、筑波地方の稲田花崗岩類に含まれる包有岩類と硫黄の起源

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要　旨

筑波地方の笠間市稲田の稲田石採石場には様々な大きさの包有岩類が分布する．これらは主に細粒砂岩のホルンフェルスであり，少量の粘板岩・石灰岩のホルンフェルスも見られる．これら包有岩類は一般に角礫状であって堆積岩との中間生成物などを伴わず，近傍の八溝層群の堆積岩類が取り込まれた“捕獲岩”であり，稲田花崗岩の固結末期に浅所で取り込まれたものと考えられる．

一方，多量の磁硫鉄鉱と少量の黄銅鉱を伴う球状包有岩が最近発見された．これは，時には堆積岩片と共存する，硫黄同位体比が極めて低い，などの理由から深所の苦鉄質岩に由来するものではなく堆積岩起源である．還元性泥質岩に濃集した堆積性硫黄が続成過程とその後のホルンフェルス化作用で硫化鉱物化し，更に角礫化されて固結しつつあったマグマ溜りに落ち込み，硫化鉱物が溶融した為に球状を呈し，浅所・低温で捕捉されたために初生の低い硫黄同位体比を保持するに至ったものと考えられる．

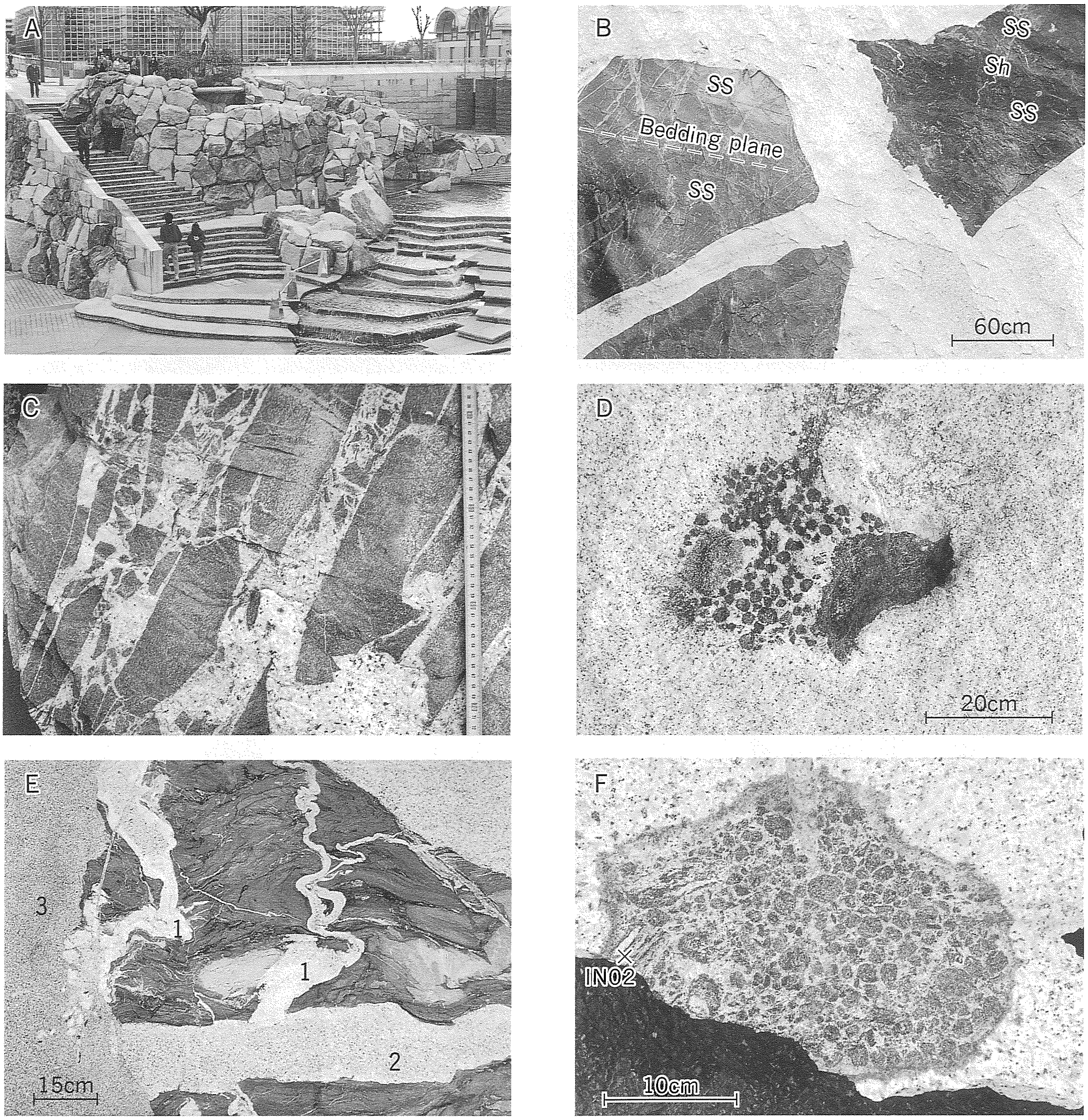

Plate 1　Mode of occurrence of enclaves in quarries at Inada.

A Blocks of sedimentary enclaves and host granite used to decorate the Tsukuba Center park of the Tsukuba Science city.
B Blocks of sandstone (ss) and shale (sh) hornfels of Kunimi Group and hosted granite at Iwakura pit, Takata quarry. Banding of sandstone and shale is seen in the blocks. Note sharp edge of the blocks.
C Fragmental sandstone hornfels intruded by fine-grained aplitic biotite granite at Iwakura pit, Takata quarry.
D Fragmental enclaves of folded sedimentary rocks (large fragments) and sulfide balls in the host granite. Maeyama pit, Nakanogumi quarry.
E Folded hornfels cut by the first-stage syenitic dikelet (1), the second stage biotite granite (2), then finally biotite granite (3). Maeyama pit, Nakanogumi quarry.
F Pyrrhotite-rich enclave (limonite-stained balls) occurring with fragments of sedimentary rocks at Nishizawa pit, Takata Quarry. Sampling site of IN02 is shown.

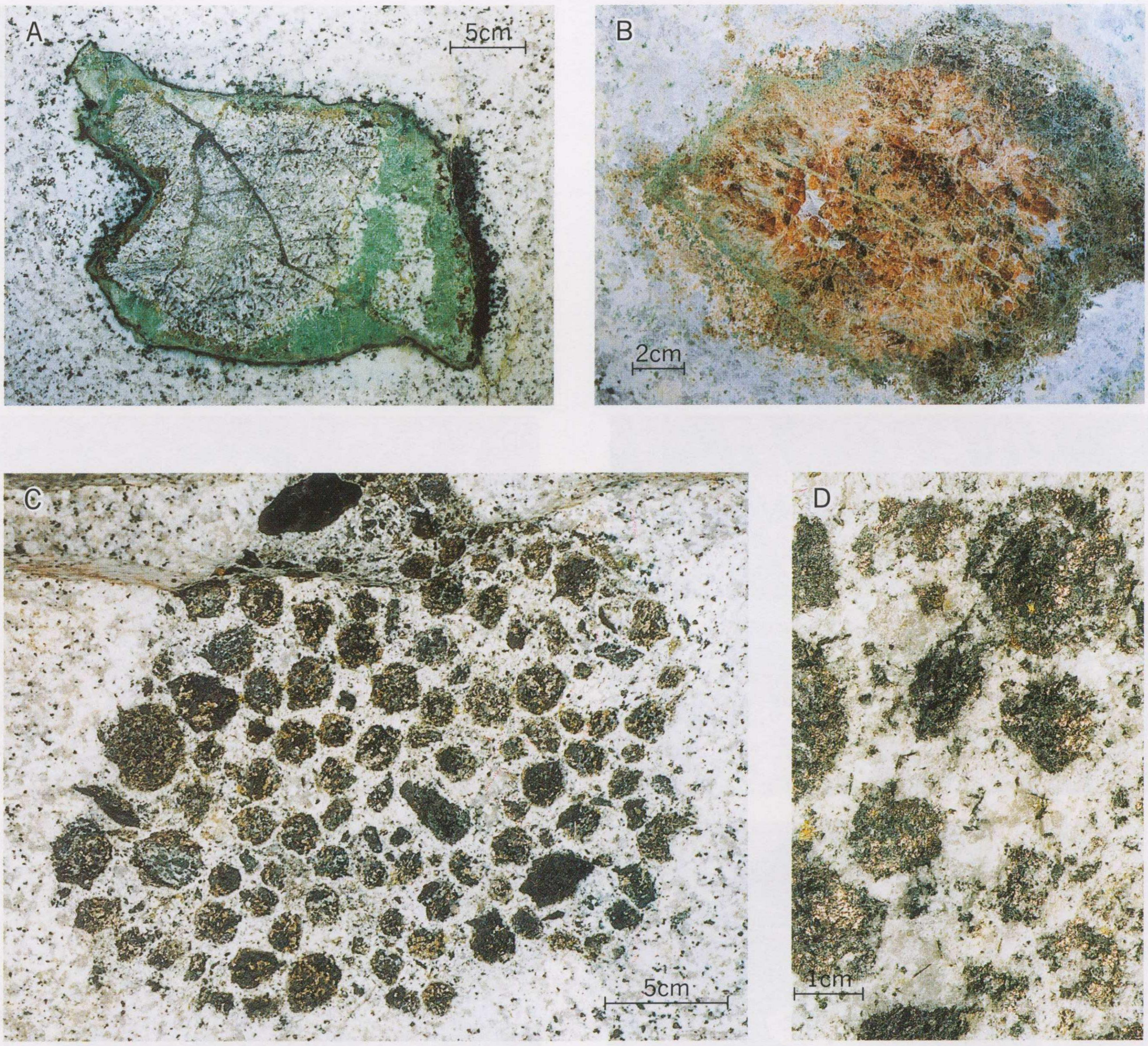

Plate 2 Mode of occurrence of skarn and ball enclaves.

A Calcite and dolomite (white in the center) replaced by diopside (green at margin) and phlogopite (dark green) along the contact with the host granite. Nishizawa pit, Takata quarry. Collection of Takata Centennial Museum.
B Garnet skarn with calcite in cavity, surrounded by green skarn (vesuvianite and epidote). Nishizawa pit, Takata quarry. Collection of Takata Centennial Museum.
C Ball enclaves occurring with some fragments of sedimentary rocks.
D Close-up of the ball enclaves. Sulfides are mostly pyrrhotite. Both C and D from Maeyama pit, Nakanogumi. Collection of Takata Centennial Museum.