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# Genesis of the Andalusite-bearing Roseki Ore Deposits in the Abu District, Yamaguchi Prefecture, Japan

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# Abstract

Andalusite-bearing Roseki ore deposits of the Abu district occur mainly in the Kiyodani member of the upper Cretaceous Fukuga formation. The Fukuga formation is characterized by intermediate to acidic volcanism. A lacustrine bed occupies the upper part of the Kiyodani member. This lake sediments accumulated during a quiet time of that volcanism. Biotite adamellite intruded immediately after the accumulation of the Fukuga formation and Roseki ore deposits were formed in the lacustrine bed.

Hydrothermally altered zones, which are composed mainly of andalusite, pyrophyllife, and sericite zones, are mostly distributed in peripheral area of the biotite adamellite stock. Almost all of the andalusite and the pyrophyllite zones have been formed in the lacustrine bed of the Kiyodani member. Both zones are usually surrounded by the sericite zones. The andalusite zone can be divided into three subzones according to main alteration minerals, such as, andalusite-pyrophyllite-kaolinite subzone, quartz-andalusitepyrophyllite subzone, and quartz-andalusite subzone from center to margin of the hydrothermally altered area. Mineral assemblages of andalusite-corundum, andalusitediaspore, andalusite-pyrophyllite, and andalusite-quartz are stable in the early stage of the hydrothermal alteration. Pyrophyllite-diaspore, pyrophyllite-kaolinite, and pyrophyllite-quartz are predominant in the middle stage. The late stage is characterized by kaolinite-muscovite and kaolinite-muscovite-mixed layered minerals. A greisen zone consisting mainly of topaz, augelite, andalusite, muscovite, and quartz is found in the central part of some andalusite zone. The hydrothermal alteration proceeded retrogressively from high-temperature to low-temperature.

The following facts such as volcano-stratigraphic evidences, the zonal distribution of the altered zones, and the K-Ar age determinations of muscovite, suggest that the hydrothermal alteration took place at about the same time as the intrusion of the biotite adamellite. Migration of chemical components is discussed in detail and compared with those of other Roseki ore deposits and some geothermal areas. Depth and temperature of the formation of the main andalusite zones are estimated. It is concluded that the andalusite-bearing Roseki ore deposits of this district were formed in close connection with the post igneous action of the biotite adamellite.

#### 1. Introduction

The Roseki ore is one of the most important industrial materials in Japan. About 1.5 million tons of ores are utilized annually for refractories, paper-clays, pottery and porcelain, tiles, and so forth. Most of the Roseki ore deposits occur in the late Cretaceous rhyolitic tuffs and lacustrine sediments in the Inner zone of Southwest Japan (Figure 1). Well known provinces are Mitsuishi-Yoshinaga of Okayama prefecture and Shōbara of Hiroshima prefecture, followed by Abu district where is famous for a particular type of Roseki ore.

Main ore deposits in this district contain abundant andalusite, which differs from the

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other deposits. In the Mitsuishi and the Shōbara districts, for example, the main constituents are pyrophyllite, kaolinite and quarts. Both districts were studied by many geologists and the genesis of ore deposits has been discussed from various viewpoints (e.g., KINOSAKI, 1963a; MATSUMOTO, 1968; KATAYAMA, 1969). KINOSAKI (1963a) found the Roseki ore deposits in stratiform shape occuring in certain stratigraphic horizon among rhyolitic tuff beds, and considered that they were formed near surface environment. KATAYAMA (1969) has shown a hypothetical scheme on the mechanism of the formation in referring to studies of



Fig. 1 Location of the studied area and distribution of Roseki ore deposits.

hydrothermal systems in recent geothermal areas. These proposals are highly commendable, but detailed information is not adequate to draw a whole genetic history of the formation, therefore a systematic study has begun at the Geological Survey of Japan (FUJII *et al.*, 1971; SHIBATA and FUJII, 1971; KAMITANI *et al.*, 1972; HIRANO *et al.*, 1972).

There are no scientific papers regarding to hydrothermal alteration of the Roseki ore deposits in the Abu district. The present writer began to study these deposits in 1969. Parts of the study have been separately published (KAMITANI, 1974a, b; SHIBATA and KAMITANI, 1974). This paper summarises all the results obtained during the past five years, in-

cluding stratigraphy and structure of the upper Cretaceous system, the mode of occurrences of the hydrothermally altered zones (Roseki ore deposits), mineral assemblages and paragenetic sequence of the alteration products, and loss and gain of chemical components during the alteration. The role of granitic intrusion is considered very important for the genesis of the hydrothermal alteration in the Abu district.

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· 80-	niacian Santonian	Urakawan	Granites	Hiroshim	a granites	Harima granites	Rokko granites	Granites
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100 -	Albian Cenc	1 koan	Kwanmon	group	Kisa g.			
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			North Kyushu	Chu	goku	West	Kinki	Chubu

Table 1 Correlation of the Cretaceous volcanics of Southwest Japan.

After Research Group for Late Mesozoic Igneous Activity of Southwest Japan(1967)

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# 2. Geological Setting

Geology of the Abu is characterized by products of intermediate to acidic igneous activity. The oldest rocks of the studied area are of the Kwanmon group, which is mainly composed of shale, sandstone, and andesitic rocks of the lower Cretaceous. The Shūnan group, which is correlated to the Yahata formation of the northern part of Kyushu, overlies unconformably the Kwanmon group and is unconformably covered by the Fukuga formation of the Abu group of the late Cretaceous. Biotite adamellite intrude them and give a contact metamorphism (Figure 2). The stratigraphic division and correlation to the standard sequences are given in Table 1.

# 2.1 Kwanmon Group

The Kwanmon group is subdivided into the Wakino and Shimonoseki subgroups. The Wakino subgroup is distributed in the southwestern part of this district. It is composed of black shale, sandstone and conglomerate, often accompanied with red shale. This subgroup contacts with the Shimonoseki subgroup by northeast-southwest trending fault. The thickness, though not meassured with accuracy, is about 250 meters. No fossils have been reported from this subgroup in the district.

The Shimonoseki subgroup, which is mainly composed of altered andesite, conglomerate, sandstone, and reddish shale, is distributed in the western part of the studied area. Altered andesite consisting principally of lapilli tuff and tuff breccia is correlated to the Kitahiko-shima formation. It is bluish green to brown in color. This rock is generally massive and its bedding plane is unclear.

Under the microscope, phenocrysts of plagioclase and common hornblende have been decomposed to chlorite, epidote, and carbonate minerals. Accessory minerals are biotite, apatite, hematite, and magnetite. At Tōkōji and Kōda, sulfide ore minerals such as pyrite, chalcopyrite, sphalerite, and galena are disseminated in strongly altered andesitic tuffs. Tōkōji mine had been operated in a small scale for several years since 1940.

The Sujigahama formation composed of conglomerate, sandstone, siltstone, and reddish shale overlies conformably the Kitahikoshima formation. The maximum thickness is estimated about 300 meters.

#### 2. 2 Shūnan Group

The Shūnan group is composed of rhyolitic lapilli tuff, crystal tuff, and lava. It is principally distributed in the southern part of this district (Figure 2). This group overlies unconformably the Kwanmon group and is covered by the Abu group. It is correlated to the Yahata formation and Tadokoyama volcanics of MURAKAMI and HASE (1967).

### 2. 3 Abu Group

Acidic volcanics distributed widely in the Inner zone of Southwest Japan have been grouped several units such as Nöhi rhyolites (KAWADA *et al.*, 1961; YAMADA *et al.*, 1971), Aioi group (KISHIDA and WADATSUMI, 1967), Takata rhyolites (YOSHIDA, 1961), and Abu group (MURAKAMI and HASE, 1967). These igneous activities are considered to have about the same age in the upper Cretaceous (Table 1).

The Abu group has been divided into four formations by MURAKAMI and HASE (1967);







Table 2 Correlation of Cretaceous formations in West Chugoku.

Shinome-Maitani-Efune, Fukuga, Ōmi, and Kumanodake formations. They have been considered to be products of intermediate to acidic volcanisms of about the same age. The Fukuga formation is studied in detail in this paper, because it is most important for the Roseki mineralization. This formation, overlying unconformably the Kwanmon and Shūnan groups, occupies the major portion of the district. The maximum thickness of this formation is estimated to have be attained approximately 1,800 meters. This thick volcanic formation is divided into three members; namely, the Kanai andesite member, Kiyodani member, and Mitsugadake rhyolite member in ascending order (Table 2). According to MURA-KAMI and HASE (1967), the Shirasuyama, Kidokoyama, and Tabira rhyolites corresponding to the Mitsugadake rhyolite member by the writer have about 1,000 meters in total thickness and are covered by the Ōi rhyolite having about 350 meters in thickness.

# 2.3.1 Kanai andesite member

The Kanai andesite member constituting the lowest part of the Fukuga formation is continuously distributed from the northern to the southwestern parts. It is mainly composed of pyroxene andesite and its tuffs. Almost all of these rocks exposed in the northern part are subjected to contact metamorphism due to intrusion of the biotite adamellite. On the contrary, the southern part has remained numetamorphosed, i.e., propylitized andesite. The maximum thickness of this member is approximately 300 meters. The rock is dark blue to

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bluish green in color and generally massive and its bedding plane is obscure.

Microscopically, this rock is generally porphyritic in texture, and pilotaxitic or hyaloophitic in some places. Clino-pyroxene and common hornblende are almost completely altered to chlorite, epidote, and carbonates. Plagioclase phenocryst has been strongly replaced by epidote and carbonates. Resorbed fine-grained quartz is rarely observed as phenocryst. The alteration products of groundmass are chlorite, epidote, carbonate, and opaque minerals.

# 2.3.2 Kiyodani member

This member corresponding to the Iinoura rhyolite and the lake sediment by MURAKAMI and HASE (1967) is mainly distributed at Uku, Kiyodani, Mt. Jingū, and Sasao and their vici-



Fig. 3 Idealized columnar section of the upper part of the Kiyodani member in Uku mine.

nities. The maximum thickness of this member is estimated about 400 meters. The rhyodacitic and rhyolitic tuff beds being about 200 meters thick make up the lower part of the Kiyodani member, and the lacustrine bed, on the other hand, having about 200 meters in maximum thickness constitutes the upper part. Figure 3 illustrates an idealized columnar section of the upper part in Uku mine. The lacustrine bed consists mainly of alternation of tuffaceous sandstone, siltstone, and shale and accompanied with rhyolitic tuffs and tuffaceous conglomerate layers. In general, tuffaceous sandstone predominates in the upper part. Rhyolitic tuffs, contrary to this, are frequently intercalated into the lower part.

Beautiful outcrops of the lacustrine bed are seen at the road-side-cut between the Kyoritsu-Nako mine and the Kiyodani. Here, it consists principally of tuffaceous sandstone, siltstone, and shale, in which graded bedding and cross-lamination can be seen. It is wholly sericitized by later hydrothermal activity, but the texture and the structure of the original rock are seen in most cases. Plant fossil, which was found in the alternation of tuffaceous sandstone and shale of the upper part at Sasao, was identified as *Otozamites* cfr. *bechei* BRONG. by TAKAHASHI (1958).

Microscopically, rhyodacitic and rhyolitic tuffs in the lower part of this member consist mainly of plagioclase, quartz, and potassium feldspars and accompanied with small amounts of biotite and common hornblende. They contain fragments of andesite and shale ("accidental fragments"), rhyolite ("accessory") and pumice ("essential"). Flattened pumice has been completely altered to very fine-grained aggregate of sericite. But the other minerals are generally unaltered. Glassshards and matrix have been devitrified to aggregates of finegrained quartz and potassium feldspar. Weak welding structure is often seen in the lower part of this member. The amount of phenocrysts ranges from 28 to 30 percent by volume (Table 4). Therefore, this rock corresponds petrographically to vitric crystal tuff.

Tuffaceous sandstone is generally inferior to sorting and grading. It is rich in quartz and poor in plagioclase, potassium feldspar, and rock fragments. Biotite and flattened pumice are commonly replaced by fine-grained, felty-shaped sericite.

Tuffaceous sandstone and siltstone of the least altered rocks were collected from the lacustrine bed at Sasao. Their chemical compositions are given in Table 3. They are very similar to those of the Mitsugadake rhyolite member although slightly poor in  $Al_2O_3$  and  $K_2O$ .

# 2.3.3 Mitsugadake rhyolite member

The Mitsugadake rhyolite member is widely distributed in this area. It has the maximum thickness of 1,100 meters around the Mt. Mitsugadake (566.2 m). This member consists of, in ascending order, rhyolite lava, rhyolite tuff, and rhyolite welded tuff, each of which can be correlated to the Shirasuyama rhyolite, Kidokoyama rhyolite, and Tabira rhyolite by MURAKAMI and HASE (1967), respectively.

Rocks which overlie the Mitsugadake rhyolite member are not confirmed in the studied area. The Ōi rhyolite (MURAKAMI and HASE, 1967) constituting the uppermost part of the Fukuga formation is considered to be absent in the area.

**Rhyolite lava;** The rhyolite lava distributed principally in the west of Kiyo and the vicinity of Mt. Shirasu has the maximum thickness of approximately 300 meters. This rock is gray to dark gray in color and has generally clear flow structure (Plate 2-3). It is porphyritic in texture, although phenocrysts consisting mainly of quartz, plagioclase, potassium feldspar, and biotite are small in size and quantity. Plagioclase phenocryst shows commonly pink to yellowish pink in color.

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Sample No.	(1)** 60510	(2)** 60511	(3)* 72616	(4)* 30305	(5)* 22401–A	(6)* 22401 -C	(7)* 110405
SiO <sub>2</sub>	74.81	75.04	72.72	77.94	74.64	79.34	75.42
TiO <sub>2</sub>	0.09	0.08	0.08	0.06	0.18	0.08	0.19
$Al_2O_3$	13.97	14.08	15.08	12.18	13.28	11.61	13.62
$Fe_2O_3$	0.32	0.16	1.00	0.60	0.80	0.52	1.36
FeO	0.97	0.79	0.22	0.18	0.72	0.36	0.29
MgO	0.18	0.14	0.07	0.04	0.27	0.28	0.21
CaO	1.39	1.46	0.46	0.15	0.97	0.15	0.13
$Na_2O$	3.92	3.48	3.78	2. 74	3.50	3.32	3.59
$K_2O$	3.56	3.76	5.46	4.85	3.54	2.68	3.66
S	0.09	0.01		_		-	_
$H_2O^+$	0.41	0.60	0.82	0.67	0.54	1.10	0.92
$H_2O^-$	0.18	0.10	0.30	0.42	0.20	0.24	0.48
Total	99.89	99.72	99.99	99.83	99.64	99.68	99.87
il	0.17	0.21	0.15	0.11		5	
ap	-	-					
mg	0.46	0.23	0.48	0.43			
hm	-		0.67	0.30			
or	21.04	22.22	32.27	28.66			
ab	33.12	29.45	31.99	23.19			
an	6.90	7.24	2.29	0.74			
wo	-						
en	0.45	0.35	0.17	0.10			
fs	1.38	1.14	-				
qz	34.50	36.54	28.74	43.06			
Total	99.16	99.02	98.87	98.74			

 Table 3 Chemical compositions and normative constituents of biotite adamellite and volcanic rocks.

(1), (2): biotite adamellite, (3): rhyolite lava of Mitsugadake rhyolite member, (4): pumice in rhyolite welded tuff of Mitsugadake rhyolite member, (5): tuffaceous sandstone, (6): tuffaceous siltstone, (7): weakly altered tuffaceous sandstone (5, 6, 7: Kiyodani member).

\*: Analyzed by Mr. M. Kawano of Chemistry section, Geological Survey of Japan.

\*\*: Analyzed by Tokyo institute of Coal and Minerals.

Under microscope, this rock shows porphyritic in texture. The phenocrysts are composed of plagioclase, quartz, potassium feldspar, and biotite, in decreasing order of abundance. Plagioclase is euhedral to subhedral crystals and has occasionally finegrained lath-shaped sericite. Potassium feldspar has been hardly replaced by sericite compared with the plagioclase phenocryst. Biotite is perfectly altered to aggregates of fine-grained sericite, epidote, and iron minerals. Content of the phenocrysts is 8 to 9 percent in volume. The groundmass has been perfectly devitrified and changed to an aggregate of fine-grained quartz and potassium feldspar. Spherulite consisting of quartz and potassium feldspar is usually recognized in the groundmass. Alteration products are sericite, chlorite, epidote, and opaque minerals.

Chemical composition of the rhyolite lava is shown in Table 3. This rock is extremely poor in CaO and MgO compared with the Nōhi rhyolite (YAMADA *et al.*, 1971).

**Rhyolite tuff;** This rock overlies conformably the Kiyodani member and the rhyolite lava of the Mitsugadake rhyolite member. But it covers unconformably the Kanai andesite member in the southwestern part where the both members are not present. The thickness of the rhyolite tuff bed is approximately 200 meters. It is composed of rhyolite vitric tuff and often accompanied with a few thin layers consisting of an alternation of tuffaceous sandstone and mudstone. These layers are a few to a several meters thick.

The rhyolite tuff is light gray to bluish gray in color. The phenocryst minerals are plagioclase, quartz, potassium feldspar, and biotite in decreasing order of abundance. Plagioclase of yellowish pink in color is characteristic.

Microscopically, it has crystal fragments of plagioclase quartz, and small quantities of potassium felspar and biotite as phenocryst minerals. Accidental and essential rock fragments such as black shale, andesite, and rhyolite lava are often contained. Matrix is composed of fine-grained quartz and potassium feldspar. Sericite in the matrix is equally abundant or more compared with the rhyolite lava.

**Rhyolite welded tuff;** This rock making up the upper part of the Mitsugadake rhyolite member is widely distributed over the whole area. The maximum thickness is approximately 600 meters measured at the eastern slope of Mt. Mitsugadake. In general, it has been strongly welded and contains many flattened pumices. It shows a distinct eutaxitic texture due to a parallel arrangement of the flattened pumices. According to ONO (1965), a parallel arrangement by essential materials is equivalent to the bedding plane. Hence, it is possible to learn a structure of the rhyolite welded tuff bed by the measurement of that planes.

This bed having about 600 meters in thickness has not been accumulated by a single eruption, but it is considered to have been deposited through several eruptions. Figure 4 is



Fig. 4 Columnar section of the lower part of rhyolite welded tuff bed in the Mitsugadake rhyolite member.

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an idealized columnar section of the continuous exposure at Nameradani, which corresponds to the lower part of the welded tuff bed. Three compound cooling units are recognized here. The thickness of each compound cooling unit ranges from 50 to 80 meters. In general, the core of each cooling unit is characterized by a distinct eutaxitic texture of wellflattened pumice, and the marginal part has been weakly welded.

This rock is dark gray to chocolate black in color and very hard. Essential materials consisting of pumice and glass shard are commonly lenticular in shape. The maximum size of the lenticular pumice is a few cm by 10 cm. Rhyolite lava, tuff, and welded tuff as accessory rock fragments are contained in the welded tuff. When large lenticular pumices and glass shards do not occur in this bed, it is difficult to distinguish from the rhyolite tuff in a hand specimen.

Under microscope, it shows strongly welded structure (Plate 4–2). The main phenocryst minerals are plagioclase and quartz, while, potassium feldspar and mafic minerals are rare. All of them are angular in shape. Biotite is often bent due to compaction. The matrix has been perfectly devitrified to an aggregate of quartz and potassium feldspar. Spherulite consisting of quartz and potassium feldspar is occasionally seen in the matrix.

Small amounts of the phenocrysts of euhedral plagioclase and resorbed quartz occur within a flattened pumice. The plagioclase and the matrix have often altered to fine-grained feltyshaped sericite. A large pumice was provided for K-Ar age determination.

Sample	Rock type	Modal comp. of phenocryst (%)	Modal composition of phenocryst minerals (%)	Measured points
1) 60402 2) 112206 3) 111301 4) 22601	rhy. w. t. rhy. w. t. rhy. w. t. rhy. w. t. rhy. w. t.	3.5 4.3 4.4 7.5	$pl > qz \gg kf \Rightarrow bt$ $pl \Rightarrow qz > kf \gg bt$ $pl > qz > kf \gg bt$ $qz \Rightarrow pl > kf > bt$	1997 1897 2038 2631
5) 72616	rhy. lava	8.6	pl>qz>kf≫bt	2221
6) 51104	rhy. lava	8.0	pl>qz>kf≑bt	2051
7) 30202	rhy. tuff	28.7	$pl > qz > kf > bt \Rightarrow hb$	2321
8) 111201	rhy. tuff	29.8	$pl \gg qz > kf \Rightarrow bt \Rightarrow hb$	2103

Table 4 Modal composition of phenocrysts in the volcanic rocks.

The phenocryst percentages in the Mitsugadake rhyolite member are extremely low compared with those of the Nöhi rhyolites which includs 30 to 50 percent of phenocryst (YAMA-DA *et al.*, 1971). This difference may suggest that the eruption mechanism was different from that of the Nöhi rhyolites.

Chemical composition of the pumice is shown in Table 3 (sample number 30305). Compared with the rhyolite lava, it is scantily in normative albite and anorthite and rich in normative quartz. Its differentiation index is 94.91 percent. The welded tuff seems to have been solidified from more differentiated magma than the rhyolite lava.

### 2. 4 Intrusive Rocks

Biotite adamellite and various dikes intrude into the Kwanmon, Shūnan, and Fukuga formations of the Abu group.

# 2. 4. 1 Biotite Adamellite

It is exposed as a stock body (30 to 40 km<sup>2</sup>) along the sea shore. This stock has dimen-

sions of about 10 km (northeast-southwest) and 4 km (northwest-southeast) in the maximum breadth.

The main phase of the biotite adamellite is mediumgrained equigranular, but the southern margin is fine-grained aplitic in texture. Between Uku and Modoro situated at the western part of this body, many boulder xenolithes are seen. The xenolith is quartz diorite in composition. In general, quantity of the xenolith is inversely proportional to the thickness of the Kanai andesite member occurring as the roof-pendant. Therefore, it is supposed that the xenolithes have been originated from this member. Pegmatite lenses and veins consisting mainly of quartz, potassium feldspar, and muscovite occur in limited extent in the marginal phase of the adamellite body. They are occasionally accompanied with large crystals of flaky molybdenite.

Under microscope, major constituents of the main phase are plagioclase, quartz, potassium feldspar, and biotite. The plagioclase is euhedral to subhedral crystals. It has composition from albite to oligoclase and shows commonly albite twinning. The potassium feldspar is composed of orthoclase and perthite and is anhedral everywhere. Cuneiform intergrowth of quartz and potassium feldspar is often seen in this phase. The quartz is commonly unhedral but rarely subhedral. Locally, the potassium feldspar is replaced by coarsegrained flaky muscovite. The biotite is euhedral to subhedral and includes often specks of apatite, zircon, and opaque minerals. It has been partially replaced by chlorite. The muscovite is common in the marginal phase. Quantity of muscovite is inversely proportional to that of biotite. It is usually euhedral to subhedral and closely accompanied with potassium

mineral	Pl	Kf	Qz	Bt	Mus	Others	Color index	
72617	34.8	28.8	32.2	3.2	0.0	0.6	3.8	
72814	33.9	25.1	36.1	4.3	0.0	0.5	4.8	Main
72818	39.4	25.4	29.4	5.6	0.0	0.2	5.8	phase
72818B	35.7	23.4	31.0	9.5	0.0	0.4	9.9	
50804A	31.2	33.9	32.1	1.7	1.1	0.0	1.7	
50804C	25.5	35.0	36.4	1.4	1.4	0.2	1.6	Marginal phase
50907	27.0	36.5	30.5	4.8	0.0	0.0	4.8	

Table 5 Modal composition of the biotite adamellite in the Abu district.

feldspar. Based on x-ray diffraction, its polytypism (YODER and EUGSTER, 1955) was identified to  $2M_1$ .

According to a modal analysis (Table 5), the main phase ranges in plagioclase from 34 to 40 percent, potassium feldspar from 25 to 29 percent, quartz from 29 to 36 percent, and biotite from 3 to 10 percent in volume, giving adamellite ratio on three major constituents (Figure 5). In the marginal phase, on the other hand, plagioclase and biotite decrease, and muscovite and potassium feldspar increase as seen in Figures 5, 6, and Table 5.

The biotite adamellite has given a contact metamorphism to part of the Shūnan group and the Fukuga formation. The strike of the contact plane varies from northeast-southwest to east-west and dips gently to south.

Regarding to granitic rocks of Chugoku region, KOJIMA (1964) divided them into three types; Hiroshima granites, Central granite complex, and Inbi granite complex. He 地質調査所月報(第28巻第4号)



Fig. 5 Modal composition of plagioclase, potash feldspar and quartz in the biotite adamellite. Open circle: marginal phase, Solid circle: main phase

Fig. 6 Modal composition of plagioclase, potash feldspar plus quartz and mafic minerals in the biotite adamellite. Simbols same as Fig. 5



pointed out that characteristics of the Hiroshima granites are richer in pegmatites and basic inclusions compared with the Inbi granite complex. According to ISHIHARA (1973), granitic rocks of the Sanyo belt corresponding approximately to the Hiroshima granites of KOJIMA have more muscovite-bearing phase than the Sanin belt, which is roughly equal to the Inbi granite complex. The muscovite-biotite adamellite generally occurs at the marginal parts of given plutons. Available K-Ar age data of the late Cretaceous granitic rocks can be divided into two groups; 70 to 90 m. y. of the Sanyo belt and 35 to 60 m.y. of the Sanin belt (Figure 7). The age difference was confirmed by recent K-Ar determinations (SHIBATA and ISHIHARA, 1974). A distinct difference between two belts beside the age is accessory amounts of Fe-Ti oxides of the granitic rocks (ISHIHARA, 1971; TSUSUE and ISHIHARA, 1974). Magnetite occurs characteristically in rocks of the Sanin belt, and ilmenite-bearing assemblage is common in the Sanyo belt; hence a low magnetic susceptibility in the latter belt (KANAYA and ISHIHARA, 1973).

In the Abu district, muscovite from the pegmatite occurring in the biotite adamellite



yields the K-Ar age of 79.0 m. y. (SHIBATA and KAMITANI, 1974). The muscovite tends to concentrate in the marginal phase and a number of pegmatites occur in this phase, indicating a high vapor pressure condition during solidification of the magma. Magnetic susceptibility was measured very low being  $6 \times 10^{-6}$   $11 \times 10^{-6}$ , and  $33 \times 10^{-6}$  emu/g on three demonstratative specimens (KANAYA, 1974; personal communication). These observations agree with the characteristics of the granitic rocks in the Sanyo belt.

Emplacement of the biotite adamellite seems to be one episode of granitic activities of the Sanyo belt. The adamellite is considered to be solidified at comparatively shallow depth, because of the occurrence of the muscovitebearing phase and pegmatite. Composition of the adamellite is similar to rhyolitic rocks of the Mitsugadake rhyolite member, so that they may have a common source. The time gap between two igneous activities are presumed to be small.

# 2.4.2 Dike Rocks

There are two types of dike rocks; quartz porphyry and porphyrite. Quartz porphyries are principally found in the western part of the area and porphyrites are observed in the central to the western part. They are generally in smallscale. The largest one, a quartz porphyry dike, which is exposed at the southwest slope of the Mt. Tōdake, has a length of about 300 meters and a maximum width of about 10 meters. Almost all of the dikes have a width less than a few meters.

Phenocrysts of quartz porphyry consist mainly of coarse-grained and euhedral to subhedral quartz, potassium feldspar, and plagioclase, and accompanied with small amount of flaky biotite. In the groundmass, an aggregate of auhedral fine-grained quartz and potassium feldspar fills with interspace among the large phenocrysts. Fine-grained sericite is often present in the plagioclase phenocryst and the groundmass.

Many porphyrite dikes invade into the hydrothermally altered zones. Hence, these are altered to an aggregation of meta-halloysite and quartz, or sericite and quartz.

#### 2. 5 Contact Metamorphosed Rocks

Part of the Fukuga formation and the Shūnan group has been converted to contact metamorphosed rocks by intrusion of the biotite adamellite. In this district, a hornblende 地質調查所月報 (第28巻第4号)

hornfels facies or a andalusite hornfels subfacies of MIYASHIRO (1971) is the highest grade, and no pyroxene hornfels facies is recognized.

Mineral assemblage of the hornblende hornfels facies is green hornblende-biotite-plagioclase. This assemblage occurs in the Kanai andesite member which is close to the adamellite. With a distance from the adamellite, epidote-hornblende hornfels and actinollite-epidote hornfels are observed.

Mineral assemblage of andalusite-biotite-muscovite, on the other hand, occurs in the rhyolitic tuff of the Shūnan group and the lower bed of the Kiyodani member near the adamellite. At a distance of above 100 meters from it, those acidic rocks have been converted to a biotite hornfels.

# 3. Structural Geology

Basement rocks of this district consist of the Kwanmon and Shūnan groups in the lower to middle Cretaceous age. The biotite adamellite, which has intruded concordantly into the Shūnan group and Fukuga formation, seems to be comagmatic derivation based on the similarity of chemical composition with the Mitsugadake rhyolite member and the K-Ar age determination. Accumulation of the Fukuga formation was initiated by intermediate volcanism of the Kanai andesite member. Rhyodacitic and rhyolitic tuffs occurring in the lower part of the Kiyodani member have covered conformably the Kanai andesite. A cauldron-like depression took place approximately at the same time as the acidic volcanism. During the rest time of the volcanism, lacustrine sediments having the maximum thickness of about 200 meters deposited in small lakes and swamps.

According to MURAKAMI (1974), many large cauldron-like basin structures having eastwest to northeast-southwest axis are known in rocks of the Abu group. Plutonic activities are associated with the formation of cauldrons.

The Fukuga formation takes on the whole a semi-basin structure opened to east in the studied area. The general strike from the northern to the western parts of this formation is approximately northeast-southwest and the dip is gently towards south. Contrary to this, the strike of the western to the southern parts is generally northwest-southeast and the dip is gently towards north.

Most of large scale faults and fractures in this district are post-hydrothermal alteration. Two groups of the faults are out-standing. One strikes northeast-southwest and the other trends northwest-southeast. In general, the former accompanies a highly fractured zone having width of more than several ten meters and displacement along the fault is estimated for more than 100 meters. These faults are also represented by topographic depressions and steep slopes. The latter has generally minor dislocation and is cut by the northeast-southwest trending faults.

# 4. Hydrothermally Altered Zones

# 4. 1 General Remarks

There are many hydrothermally altered zones in this district. Roseki ore deposits are known in this altered zones of Uku mine, the eastern part of Kiyo, and the western part of Köchi townships. The Roseki ore is mainly composed of andalusite, pyrophyllite, kaolinite,

and quartz, and accompanied with small amounts of muscovite  $(\text{sericite})^{1}$ , diaspore, corundum, and mixed layered minerals. Ores containing pyrite and hematite for more than 0.5 percent are not worth mining as industrial raw materials.

### 4. 2 Distribution of Hydrothermally Altered Zones

Hydrothermally altered zones are principally distributed in the Fukuga formation. The altered area extends approximately 10 kilometers in northeast-southwest direction and 1 to



<sup>1)</sup> A lath-shaped white mica mineral less than about 10 microns in size was definited as "sericite" and coarsegrained flaky-shaped mica mineral as "muscovite". The occurrence is different, being the former one of the main constituent minerals in the sericite zone and the latter occurring in some andalusite zone, greisen zone, and pegmatites. However, they cannot be distinguished by optic and x-ray diffraction methods.

mine	1952	1953	1954	1955	1956	1957	1958	1959	1960	1961	1962	1963	1964
Uku	9, 685	14, 459	15, 020	16, 373	18, 688	15, 681	11, 790	16, 304	9, 270	9, 410	12, 892	13, 607	16, 816
Hinomaru-Nako Gagaradani							2, 410	3, 565	2, 150	3, 805	1,040		1, 243
Torinosu Kiyodani	1, 500*	1, 500*	1, 595	1, 640	2, 140	3, 160							
Kyoritsu-Nako										1,622	2, 566	2, 987	2, 794
Kuwano-Ohira													
Nittai-Kōchi							500*	500*					
Miyazaki-Kōchi							40	190	34	100	425	680	
Miyazaki-Nako						700*	800*						
Nako-Kōchi	456	286	157	224	388								
								<u>`</u>			·		
mine	1965	1966	1967	1968	1969	1970	1971	1972	Tota	1			
Uku	17, 415	16, 134	17, 158	20, 176	19, 663	20, 014	18, 980	16, 299	325, 8	34			
Hinomaru-Nako Gagaradani	3, 018	1, 404	1, 598	1, 502	920				20, 9	47			
Torinosu Kiyodani									11,5	35			
Kyoritsu-Nako	3, 018	3, 500	6, 361	7, 543	5, 443	5, 753	5, 326	3, 878	50,7	91			
Kuwano-Ohira							312	973	1,2	85			
Nittai-Kōchi									1,0	00			
Miyazaki-Kōchi									1,4	69			
Miyazaki-Nako		-							1, 5	00			
Nako-Kôchi									1, 5	11			

Table 6 Production of the Roseki ore in the Abu district.

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3 kilometers in horizontal breadth along southern margin of the biotite adamellite. The hydrothermally altered rocks can be classified into three zones according to critical alteration minerals such as andalusite, pyrophyllite, and sericite. As a whole, the hydrothermally altered zones show roughly a zonal distribution from the peripheral portion of the biotite adamellite stock to the exterior; as andalusite zone, pyrophyllite zone, and sericite zone (Figure 8).

#### Description of the Individual Ore Deposits 4.3

Uku mine: Uku mine producinig about 2,000 tons of ores monthly is a representative Roseki ore deposit in this district. The yearly production in the part is listed in Table 6.

The Kanai andesite member consisting of lavas and tuffs of augite andesite and hornblende-augite andesite is distributed mainly in the western part of the mine area. These rocks have been generally propylitized. Part of this member have been converted to the hornblende-biotite-plagioclase hornfels due to intrusion of the biotite adamellite. The Kiyodani member having the thickness of about 400 meters is composed of rhyodacitic tuff, rhyolitic tuffs, and lacustrine sediments. In the immediate vicinity of the biotite adamellite, acidic rocks such as rhyolitic tuff have been metamorphosed to the andalusite-biotite-muscovite hornfels. The lacustrine sediments have been subjected to intense hydrothermal alteration. The andalusite-bearing Roseki ore deposits of Uku mine occur in this altered zone. The Mitsugadake rhyolite member consisting of rhyolite lava, tuffs and welded tuffs is widely distributed to the west and the south of the mine. This member is, however, released from the hydrothermal alteration.

General bedding plane of the Fukuga formation around the mine strikes approximately east-west and dips steeply towards south.



- Andalusite-biotite-muscovite hornfels 3.
- 4. Hornblende-biotite-plagioclase hornfels
- Biotite adamellite 6. Rhyolite lava
- Rhyolitic tuff 8. Pyroxene andesite 7.



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The biotite adamellite is exposed in small area along sea shore. Many pegmatites and aplites are found in margin of this stock.

Hydrothermally altered zone comprising andalusite zone and sericite zone extends to the range approximately 0.5 kilometer by 1.0 kilometer in the mine area. As a whole, this altered zone is parallel to the contact plane of the country rock and the adamellite. There are three andalusite zones and all of them are surrounded by the sericite zone.

The Roseki ore deposits of this mine are composed of three ore deposits, namely,  $Ch\bar{u}\bar{o}$  (Central), Hokubu (Northern), and Seibu (Western) deposits. The Central ore deposit corresponds roughly to the andalusite zone. The horizontal extension is about 800 meters in length of east-west and 20 to 40 meters in width. The vertical extension has been confirmed down to 80 meters level by underground workings, and it is known that the andalusite zone still continues downwards along the dip. This andalusite zone strikes east-west to eastnortheast-west-southwest and dips  $40^{\circ}$ - $80^{\circ}$  south. This zone is enclosed by the sericite zone (Figure 9).

Hinomaru-Nako mine; There are four Roseki ore deposits in the mine area. Gagaradani ore deposit had been under operation in the past fifteen years by open pit mining. West ore deposit situated at west of the Gagaradani had been prospected in a small scale during a few years. Kiyodani and Torinosu ore deposits are situated at northern part of the Gagaradani. They had been under operation in a small scale during 1950 to 1957 by open and under ground minings.

The mine area is composed of the Fukuga formation. The Kanai andesite member is distributed to the west of the Gagaradani ore deposit. This rock has been generally propylitized and partly converted to altered rock belonging to an andalusite zone. The Kiyodani member consits of rhyolitic tuff, which predominates in the lower bed, and lacustrine sediment occurring in the upper bed. All of them have been subjected to intense hydrothermal alteration. Accordingly, many andalusite-bearing Roseki ore deposits are formed in this member. The Mitsugadake rhyolite member is widespread in the southern part of this mine area. The rhyolite lava has not been subjected to the hydrothermal alteration, but parts of the rhyolite tuff and the welded tuff have been converted to sericitized rocks in the southern part of the Gagaradani ore deposit. This altered zone is considered to have formed in the same process as the andalusite-bearing Roseki ore deposits. Therefore, it is suggested that the time of the alteration is very close to the accumulation of the rhyolite welded tuff.

The strike of the Fukuga formation ranges from east-west to northwest-southeast and the dip varies from  $10^{\circ}$  to  $30^{\circ}$  south and southwest.

In the Gagaradani ore deposit, the strike of the andalusite zone is approximately northwest-southeast and the dip is gently to south. But the south side of the andalusite zone is cut by a northwest trending fault. The occurrence of the andalusite zone is shown in Figure 10.

There are two old prospecting open-pits (South pits), located at immediate south of the Gagaradani ore deposit. An alunitized rock, which is composed mainly of alunite, kaolinite, dickite, and quartz, is observed in these pits. A small scale and alusite zone occurs at the southeast of the alunite zone. However, the detailed occurrence and the relationship between the two zones are not clear because of insufficiency of the exploration.

The andalusite zone of the West ore deposit extends more than about 300 meters in eastwest direction and 70 to 150 meters across it. The host rock is rhyodacitic tuffs, rhyolitic tuffs, and lacustrine sediment of the Kiyodani member.

The Torinosu and the Kiyodani ore deposits are situated between Kuwano-Ohira mine





Geologic map of the Gagaradani deposit in the Hinomaru-nako mine. Fig. 10



- 1. Sericite zone 2. Silicified zone
- Quartz-andalusite subzone
   Andalusite-pyrophyllite-kaolinite subzone 4. Quartz-andalusite-pyrophyllite subzone
- 6. Porphylite 7. Tuffaceous shale of the Kiyodani member 8. Strike and dip of the Kiyodani member 9. Fault 10. Bore hole
  - Fig. 11 Geologic map of the Kyoritsu-nako mine.

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and Kyoritu-Nako mine. These ore deposits and those vicinities are composed of the Kanai andesite member and the Kiyodani member which have been altered intensely. The great portion of those member have been converted to the altered rocks belonging to the andalusite and the sericite zones.

**Kyoritsu-Nako mine;** Roseki ore deposits of this mine comprise of No. 1, No. 2, and No. 3 ore deposits. All of them have been under operation by open pit mining since 1961. This mine area composed of the Kiyodani and the Mitsugadake rhyolite members. The lacustrine sediments of the Kiyodani member are about 150 meters in thickness. The Mitsugadake rhyolite member overlying the lacustrine bed is composed of rhyolite tuffs and welded tuffs. Porphyrite dikes varying from 1 to 3 meters in width have intruded into them. These members are generally flat lying to gently dipping towards south. Intense hydrothermal alteration is mostly known in the lacustrine sediments, while part of the Mitsugadake rhyolite member has been subjected nothing but weak sericitization. The andalusite zone continues from this mine to Kiyodani ore deposit of Hinomaru-Nako mine. In general, the andalusite zone is enclosed by the sericite zone, but a small scale silicified zone occurs locally between the two zones.

The andalusite zone of No. 1 ore deposit extends northeast-southwest direction and dips gently towards north. From this evidence and many short drilling data, it is supposed that the andalusite zone of No. 1, No. 2, and No. 3 ore deposits is a continuous body. The southeast side of this andalusite zone is cut off by northwest-southeast fault (Figure 11). The extension of the andalusite zone of No. 3 ore deposit is approximately north-south and the dips vary



 1. Wast and debris
 2. Porphyrite
 3. Sericite zone
 4. Quartz-andalusite subzone

 5. Quartz-andalusite-pyrophyllite subzone
 6. White cla vein

Fig. 12 Occurrence of the hydrothermally altered rocks in the No. 3 open pit.

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from  $60^{\circ}$  to  $80^{\circ}$  west (Figure 12).

Kuwano-Ohira mine; This mine situated on the northern slope of Mt. Ohira. The area is composed of the Kanai andesite member and Kiyodani member of the Fukuga formation and the biotite adamellite. The Kanai andesite member near the biotite adamellite has been metamorphosed to hornblende hornfels and part of the member has been altered to chloritized rock. The Kiyodani member comprising the rhyodacitic tuffs and rhyolitic tuffs has been converted to the hydrothermally altered rocks belonging to sericite zone and andalusite zone. The lacustrine sediment is distributed locally around the peak of Mt. Ohira.

General strike of the Fukuga formation is approximately east-west and the dips vary from 20° to 50° south (Figure 13).

The hydrothermally altered zone including that of Kuwano-Ohira mine continues from this mine to Kyoritsu-Nako mine. This altered zone consists mainly of andalusite zone and sericite zone. The chlorite zone is confined to part of the Kanai andesite member.

**Kōchi ore deposits;** They are situated at about 2 kilometers west of Mt. Mitsugadake. There are four pyrophyllite Roseki ore deposits; Nittai-Kōchi, Miyazaki-Kōchi, Miyazaki-Nako, and Nako-Kōchi mines. All of them had been under operation by underground mining from 1950 to 1963. This area is composed principally of the Kanai andesite, Kiyodani, and Mitsugadake rhyolite members. Small scale quartz-porphyry dikes have intruded into them. In general, the Fukuga formation shows a gentle andulation.

Figure 14 is the geologic map of the hydrothermally altered zones around Nittai-Kōchi and Miyazaki-Kōchi mines. The altered zones comprising pyrophyllite zone and sericite zone



Wast and debris
 Sericite zone
 Andalusite zone
 High grade ore (andalusite-kaolinite-muscovite subzone)
 Biotite adamellite
 Open pit
 Strike and dip of Kiodani member
 Fig. 13 Occurrence of the Roseki ore deposits in the Kuwano-ohira mine.

occur mostly in the Kiyodani member and parts of those occur in the Kanai andesite member and the Mitsugadake rhyolite member. However, the pyrophyllite zones are confined to the Kiyodani member. Rhyolite lava and welded tuffs of the Mitsugadake rhyolite member and the quartz-porphyry dikes are free from the alteration.

# 4. 4 Occurrences of Hydrothermally Altered Zones

Altered rocks of andalusite zone are composed principally of andalusite, pyrophyllite, kaolinite, and quartz, and accompanied with small amounts of corundum, diaspore, muscovite, pyrite, and hematite. An appearances of altered rock such as softness, soapy feeling, and appearent specific gravity, are affected remarkably by contents of the major constituent minerals. In Uku mine, Gagaradani ore deposit of Hinomaru-Nako mine, and Kyoritsu-Nako mine, the andalusite zones can be roughly divided into three subzones as follows;

Andalusite zone (Type A)

andalusite-pyrophyllite-kaolinite subzone quartz-andalusite-pyrophyllite subzone quartz-andalusite subzone



Fig. 14 Geologic map of the Kochi Roseki ore deposits.

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Besides, the andalusite zone of Kuwano-Ohira mine, Torinosu, and Kiyodani ore deposits characterized by large amounts of muscovite can be subdivided into two subzones as follows;

Andalusite zone (Type B)

(andalusite-kaolinite-muscovite subzone

quartz-andalusite-muscovite subzone

As the other altered zones, pyrophyllite, greisen, alunite, and silicified zones occur, in a limited extent, in the Fukuga formation.

# 4.4.1 Occurrence of Type A

Occurrence of andalusite zone (Type A) can be observed in detail at open pits of the Central ore deposit in Uku mine (Figure 15).

Andalusite-pyrophyllite-kaolinite subzone; The rocks of this subzone are white in color, compact but soft, and have commonly a soapy feeling. The subzone having width of 2 to 5 meters continues from No. 2 to No. 5 open pits, although it branches frequently. The strikes of this subzone range east-west to eastnortheast-westsouthwest and the dips vary 50° to 70° south. These directions are roughly concordant with the bedding plane of the lacustrine sediment. Figure 16 is an example of the occurrence of this subzone. As seen in the figure, the altered rock which is mainly composed of andalusite, pyrophyllite, corundum, and kaolinite is cut by veinlets comprising assemblages of pyrophyllite-diaspore-kaolinite, pyrophyllite-kaolinite, and pyrophyllite-muscovite. These veinlets are moreover cut by clayey veins. The mineral assemblage of the clayey veins are kaolinite-muscovite and kaolinite-muscovite-mixed layered minerals.

The No. 1 ore deposit of Kyoritsu-Nako mine, having a width from 0.5 to 1.0 meters, has intersected diagonally the bedding plane of the Kiyodani member. The main constituent minerals are similar to those of Uku mine but generally scanty in pyrophyllite and kaolinite. Pyrite has disseminated in this subzone and its surroundings, and the subzone is tainted by its oxidation color.

Quartz-andalusite-pyrophyllite subzone; The altered rocks of this subzone are white in color, compact, and harder than the ores previousely mentioned. This subzone having a width from 5 to 10 meters has been formed around the andalusite-pyrophyllite-kaolinite subzone and changes gradually into the other subzones. The constituent minerals are similar to those of the andalusite-pyrophyllite-kaolinite subzone, however there are remarkable decrease of kaolinite and diaspore, and increase of quartz. Pyrite and hematite also increase slightly.

In Gagaradani ore deposit of Hinomaru-Nako mine, the thickness of this subzone, although not able to measure with accuracy, is more than 10 meters. Muscovite increases locally and while pyrophyllite decreases slightly. Pyrite disseminates into the subzone and it tends to increase towards the lower part.

In the No. 1 and No. 2 ore deposits of Kyoritsu-Nako mine, this subzone has thickness of 10 to 20 meters. At the northern part of the No. 1 ore deposit, three subzones are recognized in the quartz-andalusite subzone. One subzone accompanies with the andalusitepyrophyllite-kaolinite subzone of small scale. On the other hand, quartz-andalusite-pyrophyllite subzone of No. 3 ore deposit has been intruded by two white clay veins having a width up to 1 meter. The altered rocks around the clay veins show commonly white in color. Preexisted disseminated pyrite had been leached out by the invasion of the veins. In this deposit, andalusite-pyrophyllite-kaolinite subzone is not seen.

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1. Wast and debris2. Sericite zone3. Pyrite impregnation4. Hematite impregnation5. White clay vein6. Quartz-andalusite subzone7. Quartz-andalusite-pyrophyllite subzone8. Andalusite-pyrophyllite-kaolinite subzoneFig. 15Geologic map of the Chūō kotai in the Uku mine, showing the zonal distribution of the hydrothermally altered rocks.

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Fig. 16 A sketch of the andalusite-pyrophyllite-kaolinite subzone at the No. 2 open pit in the Uku mine.

Quartz-andalusite subzone; The rocks of this subzone is white, compact, and very hard. The original texture are often observed. In Uku mine this subzone occurs at both sides of the quartz-andalusite-pyrophyllite suzone; namely, the upper quartz-andalusite subzone and the lower one. The thickness of the upper subzone is about 15 meters at maximum and the lower one is about 10 meters. The main constituent minerals are quartz and andalusite, and hydrous minerals such as pyrophyllite, Kaolinite, and diaspore are exceedingly small in amounts (Table 12). Pyrite and hematite occur principally between the quartz-andalusite subzone and the sericite zone. The former is rich in the lower subzone and the latter dominantly occurs in the upper subzone.

In Gagaradani ore deposit, this subzone occurs only at the upper side of the quartzandalusite-pyrophyllite subzone. The maximum thickness of this subzone is about 5 meters. It has often been cut by the pyrophyllite-quartz veins.

In the No. 1 ore deposit of Kyoritsu-Nako mine, occurrence of this subzone is similar to that of Uku mine. Pyrite has remarkably disseminated into the subzone. However, hematite is not observed. The upper subzone changes gradually into silicified zone. The silicified zone comprising only fine-grained mozaic quartz occurs at hanging side of the upper subzone (Figure 11). It is often accompanied with veinlets consisting of andalusite-quartz. An interfinger relationship between this subzone and the quartz-andalusite-pyrophyllite subzone is observed in No. 3 ore deposits (Figure 12). The quartz-andalusite subzone changes gradually into the sericite zone within a few meters. Constituent minerals of this transitional part are andalusite, sericite (muscovite), and quartz.

Two porphyrite dikes have intruded into the quartz-andalusite subzone and the sericite zone, but they have influenced neither thermal nor hydrothermal effects to the peripheral al-

#### tered rocks.

# 4.4.2 Occurrence of Type B

Andalusite-kaolinite-muscovite subzone; The altered rocks of this subzone are light gray to bluish light gray in color and soft, but do not have soapy feeling. This subzone changes gradually into the quartz-andalusite-muscovite subzone. It occurs commonly as small irregular-shaped lens and vein (Figure 13). The largest lens is about 20 meters in length and about 5 meters in width. In general, this subzones occur in a center of the quartz-andalusite-muscovite subzone. Constituent minerals are andalusite, kaolinite, and muscovite and accompanied with very small quantities of quartz and pyrophyllite. Corundum and diaspore cannot be found. A small amount of fine-grained pyrite disseminates, while hematite scarecely occurs in this subzone.

Quartz-andalusite-muscovite subzone; The rocks of this subzone are gray to yellowish white in color, compact, and hard. As shown in Figure 13, there are two types of the occurrences; one following the bedding plane of the original rocks, while the other controlled by fractures. The latter is recognized at the eastern and the western part of Kuwano-Ohira mine. This subzone changes gradually into the sericite zone within a few meters wide. Main constituents are quartz, and alusite, and muscovite and the accessory is kaolinite. Pyrophyllite is rare. Corundum, diaspore, and hematite are not observed. Pyrite disseminates all over this subzone.

Occurrences of Torinosu and Kiyodani ore deposits are generally similar to that of Kuwano-Ohira mine.

# 4.4.3 Pyrophyllite Zone

This zone is distributed only in the western portion of Kōchi township. The altered rocks of this zone are white to pale green in color, compact, and has commonly a soapy feeling. Main constituents are pyrophyllite and quartz, and accompanied minerals are small amounts of kaolinite and quartz. Diaspore and corundum occur often as accessory minerals. Pyrite and hematite disseminate in both the sericite and pyrophyllite zones.

Nittai-Kōchi ore deposit, where details of the occurrence of the pyrophyllite zone can be observed, has been formed replacing the lacustrine sediment (Figure 14). The pyrophyllite zone is 3 to 10 meters in thickness, but lateral extension is limited. In general, this zone is small in scale and lenticular or irregular mass in shape.

## 4.4.4 Greisen Zone

This zone occurs in the central part of the andalusite zone belonging to Type B. It extends more than 100 meters in eastnortheast-westsouthwest direction and has the maximum width of about 30 meters (Figure 17). This zone changes gently into the andalusite zone within a half meter. In the eastern extension of this zone, large amounts of dumortierite occur in the same andalusite zone belonging to Type B. This rock is mainly composed of topaz, augelite, andalusite, quartz, and pyrite and is accompanied with small amounts of muscovite, diaspore, and kaolinite. Accessory minerals are apatite, zirccn, and rutile.

# 4.4.5 Alunite Zone

This zone has been formed in the rhyolite tuff corresponding to the lower bed of the Mitsugadake rhyolite member. However, the relation between this zone and the andalusite zone is rarely observed. A northwest-southeast fault which displacement is estimated more than a hundred meter, separates the andalusite zone of Gagaradani ore deposit from that of the South pits. The alunite zone may have been formed in different geologic and chemical

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Weakly altered rock
 Dumortierite vein
 Topaz-augelite-andalusite-quartz rock (greisen zone)

4. Sericite zone 5. Andalusite zone 6. Dip and strike of host rock

7. Boundary between Kiyodani member and Mitsugadake rhyolite member

Fig. 17 Occurrence of andalusite zone and topaz-augelite-andalusite-quartz rock of the West ore deposit in the Hinomaru-nako mine.

conditions from those of the andalusite zone of Gagaradani ore deposit.

# 4. 5 Structural Control of Hydrothermal Alteration

# 4. 5. 1 Stratigraphic Control

Hydrothermal alteration extends to all members of the Fukuga formation and part of the Shūnan group. Among them, the most intense alteration is confined to the Kiyodani member of the Fukuga formation. The others have been subjected only to sericitization. The lacustrine sediments constituting the upper part of the Kiyodani member has affected by the most intense hydrothermal alteration. In general, the sediments which are located within about 500 meters of net distance from the biotite adamellite, have been converted to the altered rocks belonging to andalusite zone. The sediment cropping out at the northeastern part—northern slope of Mt. Jingu—of this district has been altered to merely sericitized rock, where the net distance between this sediment at Sasao being farther from the adamellite, on the other hand, has been hardly affected by the hydrothermal alteration. The Kanai andesite member around the adamellite has been metamorphosed to hornblende hornfels facies, but pre-existed fractures and those peripheries have been altered to sericitized and chloritized rocks. The Mitsugadake rhyolite member, as a whole, has been slightly altered to sericitized rock along fractures.

In brief, the lacustrine bed of the Fukuga formation close to the biotite adamellite has been subjected to the most intense and the most extensive alteration. Therefore, many and-

alusite-bearing Roseki ore deposits have been formed in this bed.

# 4.5.2 Detailed Structural Control

Occurrences of the main andalusite zones and the pyrophyllite zones can be roughly divided into two types. One is the zones occurring in concordant with a structure of the lacustrine bed, and the other is oblique to the bedding of the original rock.

An example of the former is observed in the andalusite zone of the Central ore deposit, Uku mine. Even though the lacustrine bed altered intensely, the original texture and structure can be observed in many places. Therefore, it is easy to clarify the structure of the host rocks. The andalusite zone extends approximately along the bedding plane of the lacustrine sediment (Figure 15). Similar features are also seen in Gagaradani ore deposit and part of Kuwano-Ohira mine. The hydrothermal alteration terminates rapidly at the tuffaceous shale bed which often occurs at the top of the Kiyodani member. It is supposed that the tuffaceous shale bed behaved as a local cap rock against the hydrothermal solution and the solution infiltrated into the bed having higher permiability than that of the surrounding rocks. The occurrence of the andalusite zone of their ore deposits indicates that the alteration was remarkably controlled by the structure of the lacustrine bed.

An example of the latter is seen in Kyoritsu-Nako mine and the West pit of Hinomaru-Nako mine. In Kyoritsu-Nako mine the andalusite zone intersects diagonally the bedding plane of the lacustrine sediment, as illustrated in Figures 11 and 12. But sericitization is more extensive in the lacustrine bed than that of the other beds. It is suggested that the hydrothermal solution ascended principally along some pre-existed faults and then infiltrated into the lacustrine bed.

# 5. Minerals

Minerals of the main Roseki ores of this district are listed in Table 18. These minerals were identified principally by optical and x-ray diffraction methods.

## Andalusite $Al_2SiO_5$

Andalusite is one of the most abundant minerals in the six ore deposits. It also occurs in the hornfels near the biotite adamellite stock as a contact metamorphosed mineral. In many cases, andalusite is grayish to colorless and semitransparent in a hand specimen, but in Kuwano-Ohira mine it shows often pale pinkish color and small radiating rossettes. In general, it is difficult to distinguish andalusite from quartz and topaz because of their fine-grained nature being up to 0.5 millimeters in size.

Microscopically, andalusite occurs as short prismatic, radiated, and spongy crystals. In the central part of the andalusite zone, it is commonly subhedral and about 0.5 millimeters in length, but often grows more than 1 millimeters. In the andalusite-pyrophyllite-kaolinite subzone and the quartz-andalusite-pyrophyllite subzone, the mineral has been replaced remarkably by pyrophyllite and kaolinite along the cleavage and the margin of the crystal (Plate 6-1). In the quartz-andalusite subzone, small spongy crystals include many fine-grained quartz (Plate 6-3). Euhedral and radiating crystals remain perfectly in the case of non-clay minerals associated with them (Plate 5-3). It is usually colorless in thin sections. It is distinguished from other minerals by its relatively high refringence and low birefringence. This mineral is biaxial, negative, and 2V is  $82^{\circ}$  to  $85^{\circ}$ . The x-ray diffraction data are shown in Table 7.

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Skinr A	ner, Clark and Appl Am. Jour. Sci., (196	Central ore deposit of the Uku mine.		
D(Å)	I/I <sub>1</sub>	hkl	D(Å)	I/I,
5.54	100	110	5.57	100
4.53	90	101	4.53	70
3.92	70	111	3.93	30
3.52	60	120	3.52	30
3.49	40	210	3.49	30
2.78	20	002		
2.77	90	220	2. 78	70
2.486	20	112	2.486	20
2.482	20	221		
2.466	50	310	2.470	20
2.378	20	031	2. 380	20
2.353	20	301	2.356	10
2.273	40	022	2.276	20
2.255	40	311	2.257	20
2.181	10	230, 122		
2.170	90	320	2.174	40
2.031	10	231		
1.975	20	040	1.975	10
			1	,

Table 7 X-ray powder diffraction data for andalusite.

# **Pyrophyllite** $Al_2Si_4O_{10}(OH)_2$

Pyrophyllite is one of the main constituent minerals in this district except for Kwano-Ohira, Torinosu, and Kiyodani ore deposits. It is commonly pale blue to grayish white in color, soft, and compact, and has a soapy feeling or a waxy touch. Pyrophyllite is difficult to distinguish from kaolinite and muscovite in a hand specimen because of its extreme finegrained nature.

Under microscope, pyrophyllite occurs as an aggregate of fine-grained felty-shaped crystals varying in length from a few to 10 microns. It is distinguished from kaolinite and diaspore by difference of the refringence and the interference color, but the mineral cannot be identified from muscovite by optical method, except for some muscovites occurring as fanshaped aggregation larger than several tenth microns. Pyrophyllite was determined only by x-ray diffraction in this study, using differences of the basal spacings of 9.2 Å and 10 Å.

Pyrophyllite generally occurs with diaspore and kaolinite, so that it is difficult to have a pure pyrophyllite sample. Some separated samples from the andalusite-pyrophyllite-kaolinite subzone of the Central ore deposit in Uku mine were identified to monoclinic pyrophyllite (BRINDLEY and WARDLE, 1970) as shown in Figure 18.

#### Diaspore AlOOH

This mineral is found commonly in all andalusite zones except for Kuwano-Ohira, Torinosu, and Kiyodani ore deposits, but it is less abundant than the previous two minerals. It is extremely fine grains, gray to white in color, and semitransparent in a hand specimen.

Microscopically, diaspore occurs as needle and long pyramidal shaped crystals. Occasionally, it also occurs as coarsegrained crystal which have been replaced commonly by pyrophyllite and kaolinite (Plate 5-2). Diaspore occurs closely associated with pyrophyllite, and also accompanies with andalusite and corundum.

Diaspore, which is characterized by very high refringence and interference color, can be distinguished easily from the other minerals under microscope. This mineral was also confirmed by x-ray diffraction method.

# Corundum Al<sub>2</sub>O<sub>3</sub>

Corundum is found in Uku mine, the Gagaradani ore deposit of Hinomaru-Nako mine. It is also found barely in  $K\bar{o}chi$  ore deposits. In general, it occurs as veinlets or porphyritic aggregates in the andalusite-pyrophyllite-kaolinite subzone and the quartz-andalusite-pyrophyllite subzone. In a hand specimen, it is ultra-marine blue in color and very hard.

Microscopically, corundum is granular to short prismatic crystals and shows often weak pleochroism; that is, O is pale blue and E is colorless. It is characterized by extremely high refringence and low birefringence. Fine-grained short prismatic corundum grows occasionally in and around a coarse-grained andalusite crystal in the andalusite-pyrophyllite-kaolinite subzone (Plate 5-1). It is considered that both minerals were formed in equilibrium at the early stage of the hydrothermal alteration. X-ray diffraction pattern of this mineral is shown in Figure 18.

#### Kaolinite $Al_2Si_2O_5(OH)_4$

Kaolinite is one of the most common minerals in the andalusite and pyrophyllite zones of this district. In a hand specimen, it is white in color and soft. It has usually a soapy feeling.

Under the microscope, it occurs mostly as extremely fine-grained lath-shaped crystals. It has low refringence and birefringence showing first-order gray interference color. Kaolinite has been formed commonly as a decomposition product after andalusite, and it also occurs closely with pyrophyllite in the middle stage of the alteration. In the late stage, kaolinite accompanied with muscovite and mixed layered minerals. Its x-ray diffraction pattern is compared with ASTM 5-0143.

# **Dickite** $Al_2Si_2O_5(OH)_4$

Dickite occurs only in the Gagaradani south pit and the West ore deposit of Hinomaru-Nako mine. It cuts both of the andalusite zone and the alunite zone as veinlets up to a few millimeters in width. Dickite is commonly white in color, semitransparent, and soft. It has also a soapy feeling.

Microscopically, it occurs as small lath-shaped crystals. Its refringence is low and interference color is first-order gray. In general, dickite crystals are somewhat larger than kaolinite. X-ray diffraction pattern of this mineral was compared with the dickite from Shōbara, Hiroshima Prefecture, Japan (TAKESHI, 1958).

# Meta-halloysite Al<sub>2</sub>Si<sub>2</sub>O<sub>5</sub>(OH)<sub>4</sub>

Meta-halloysite occurs as a main constituent mineral of altered dikes in the No. 3 open pit and the road cut of the Kyoritsu-Nako mine. The dikes have been converted to association of meta-halloysite and quartz, accompanied with a small amount of sericite. They are generally yellowish brown in color due to oxidation of pyrite.

# Alunite $(K, No)Al_3(SO_4)_2(OH)_6$

Alunite is found in the Gagaradani South pits. It is one of the main constituent minerals in the alunite zone. In a hand specimen, a pinkish color which is proportional to its contents in the altered rock is diagnostic.

Microscopically, alunite is usually euhedral to subhedral and from 0.1 to 0.5 millimeters

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in length. It has a clear basal cleavage (0001). Its interference color ranges up to straw yellow of the first order.

**Dumortierite**  $4[(Al,Fe)_7BSi_3O_{18}]$ 

Dumortierite occurs as a network in the andalusite zone belonging to Type B on the extension of the West ore deposit. It is generally purple red in color.

Microscopically, dumortierite is mostly needle-shaped crystal, and shows commonly radiated aggregations (Plate 7-3). Its cleavage is parallel to (001) plane. It is characterized by distinct pleochroism; that is, X=purple, Y=Z=colorless. It has parallel extinction in long-itudinal section. In general, dumortierite grows across and alusite and quartz grains, and also forms often radiated aggregates in a core of these minerals. X-ray diffraction pattern of this mineral is given in Figure 19 and Table 8.

**Topaz**  $4[Al_2SiO_4(F,OH)_2]$ 

Topaz is one of the main constituent minerals of "greisen" zone at the West ore deposit. It is commonly gray to colorless and semitransparent in a hand specimen.

Under microscope, topaz is mostly subhedral to anhedral granular but often short prismatic. It differs from andalusite by perfect basal cleavage parallel to (001) plane and negative

	ASTM 7-71		Hinomaru-Nako mine			
D(Å)	I/I1	hkl	D(Å)	$I/I_1$		
10.2 5.89 5.84	10 90	110 200 130	10.27 5.90	10 100		
5.09 5.04 4.26 4.01	90 90 45 50 10	220 040 021 121	5.09 5.01 4.26 4.01	70 10 10		
3.87 3.847 3.827 3.612	50 35 50	$ \begin{array}{r}     031 \\     310 \\     240, 150 \\     211 \end{array} $	3.86	3 10		
3.450 3.399 3.370 3.301	75 50 10	221, 041 330 060	3.453 3.411 3.348 3.324	30 20 30		
3.227 3.066 2.980 2.951	85 55 15	231 051	3.235 3.076 2.947	20 20 30		
2.926 2.891 2.824 2.720	90 70 10		2.938 2.938 2.893 2.820 2.786	50 20 10		
2.667 2.549 2.529	35 100 40		2.780 2.673 2.547 2.533	10 10 40 20		
2.481 2.461 2.421 2.409 2.245	23 25 30 30		2.486 2.460 2.421	10 10 20		
2.343 2.327 2.241 2.226	40 15 20 10		2.348 2.285 2.247 2.225	10 10 10 10		
2.182 2.126 2.100 2.093	30 15 55 30		2.184 2.130 2.097 2.088	10 10 30 40		

Table 8 X-ray powder diffraction data for dumortierite.

elongation (Plate 7-1). Its refringence is lower than that of andalusite and its interference color ranges to straw yellow of the firstorder. Optically this mineral is biaxial and positive. Its 2V ranges from  $51^{\circ}$  to  $53^{\circ}$ . Topaz coexists generally with quartz, andalusite, and diaspore. X-ray diffraction pattern of this mineral is given in Figure 19 and Table 9.

### Augelite $Al_2PO_5(OH)_4$

Augelite occurs in "greisen zone" at the West ore deposit. It coexists commonly with topaz and quartz. In some place, it is present as aggregations of longitudinal crystals of a few millimeters and more in length.

Microscopically, it is colorless. Its birefringence is weak. It is biaxial and with 2V about  $45^{\circ}$ . The large crystal of augelite often encloses many small grains of zircon. X-ray diffraction pattern is given in Table 10.

# Wilkeite $Ca_5[(P,S,Si,C)O_5]_3$ (OH)

Wilkeite was found in the upper open pit of the No. 1 ore deposit, Kyoritsu-Nako mine. It is scattered in the vein consisting mainly of pyrophyllite and kaolinite. It is colorless, hard, and semitransparent in a hand specimen. Almost all of the wilkeite is euhedral and hexagonal short prismatic crystals. The largest crystals is about 1.5 cm along c-axis.

Microscopically, it is colorless and is characterized by very low interference color of the first-order. It has been often replaced by pyrophyllite and kaolinite (Plate 8-1). Wilkeite has

mine	Hinomaru-N		ASTM 12-765	
I/I1	D(Å)	hkl	I/I1	D(Å)
10	4.41	020	6	4.40
10	4.17	002	4	4.19
10	4.09	110	12	4.11
10	3.88	021	6	3.90
80	3.68	111	60	3.69
60	3.18	120	65	3.20
40	3.03	022	35	3.04
40	2.976	121	25	2.986
100	2.928	1.12	100	2.937
30	2.479	130	20	2.4804
40	2.390	103	10	2.3966
30	2.372	131	25	2.3783
80	2.354	023	45	2.3609
30	2.307	200	8	2.3247
10	2.302	113	10	2.3130
10	2.274	210	6	2.2470
10	2.231		-	
10	2,200	040	10	2.1989
10	2.169	211	ĨŽ	2.1711
10	2.125	041	- 8	2.1269
50	2.102	123	45	2.1049
30	2.056	220	25	2.0555
		140	8	1.9872
10	1.975	212	10	1.9816
		$\overline{042}$	4	1.9470
10	1.933	141	6	1.9340
50	1.864	114	25	1.8691
30	1.861	133	25	1.8553
30	1.850			
30	1.812	230	12	1.8212
10	1.793	142	8	1.7969
10	1.777	231	6	1.7796
30	1.669	232	25	1.6706
20	1.654	223	8	1.6561
10	1.630		-	
ĨŎ	1.619	143	12	1.6203
	1.650 1.812 1.793 1.777 1.669 1.654 1.630 1.619	230 142 231 232 223 143	12 8 6 25 8 12	1.8212 1.7969 1.7796 1.6706 1.6561 1.6203

Table 9 X-ray diffraction data for topaz.

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	ASTM 14-38	Hinomaru-Nako mine		
D(Å)	I/I1	hkl	D(Å)	I/I1
6.7	4	110	6.65	10
4.67	60	$001, 20\overline{1}$	4.70	50
4.27	10	111	4.25	20
4.00	80	020	3.99	50
3.963	4		3.690	10
3.604	10	310	3.601	10
3.506	90	$111. \ 31\overline{1}$	3.504	90
3.338	100	220	3.336	100
3.148	16	401	3.145	10
3.038	16	400, 021	3.035	10
2.603	4	130	2.601	10
2.536	10	$20\overline{2}$	2.534	10
2.488	45	221, 311	2.485	20
2 377	8	112 312	2 375	10
2.336	. 4	002, 402	2 345	10
2.22	2	330	2.345	10
	-		2.233	10
2 20	6	331 131	2 199	10
2.20	6	512	2.155	10
1 999	16	040	1 997	10
1 0 3 0	18	421 602	1 0 2 8	20
1 918	10	421, 002 621	1.936	10
1 898	20	240	1 897	10
1.850	50	331	1.865	30
1.007	50	551	1.005	

Table 10 X-ray powder diffraction data for augelite.

been considered to form a solid solution with fluorapatite and elestadite (WINCHELL, 1951). Hence, the chemical analysis of this mineral is necessary for accurate identification. X-ray diffraction pattern of wilkeite is shown in Table 11 and Figure 19.

Muscovite  $4[KAl_2(AlSi_3O_{10})(OH)_2]$ 

Muscovite occurs commonly in the andalusite zone. It increases extremely in the andalusite zone belonging to Type B near the biotite adamellite. It is white to pale green in color and in some places occurs as an aggregation of coarse-grained flaky crystals. The maximum diameter comes up to about 3 cm (Plate 8–2).

Under microscope, it is difficult to distinguish the mineral from pyrophyllite, although in some cases it occus as a large radiated agggregation. Muscovite from andalusite zones and pegmatites in the biotite adamellite in this area were determined  $2M_1$  (YODER and EUGSTER, 1955) by X-ray diffraction method.

Sericite  $4[KAl_2(AlSi_3O_{10})(OH)_2]$ 

Fine-grained mica mineral in the sericite zone is called "sericite" in this report, although x-ray diffraction pattern of both sericite and muscovite in the studied area belong to  $2M_1$  muscovite according to YODER and EUGSTER (1955). In a hand specimen, it is yellowish green to pale green in color, soft, and has a soapy appearance.

Under microscope, sericite occurs as fine-grained lath-shaped crystals up to 10 microns. In the sample consisting of pyrophyllite and sericite, it is difficult to distinguish one from another.

# Mixed layered minerals

Mixed layered minerals were found in the Central ore deposit of Uku mine, the No. 3 ore deposit of Kyoritsu-Nako mine, and Kuwano-Ohira mine. They coexist commonly with kaolinite and muscovite in the clayey veins. This mineral from Uku mine has been iden-

	ASTM 6-0454	Kyoritsu-Nako mine		
D(Å)	I/I1	hkl	D(Å)	· I/I1
8.14	60	100	8.13	20
5.25	5	105	5.25	10
4.63	5	110	4.58	20
4.06	5	200	4.06	10
3.85	10	111	3.87	10
3.45	70	002	3.44	80
3.18	10	102	3.17	30
3.06	30	120	3.06	40
2.80	100	121	2.80	100
			2.77	40
2.70	90	300	2.70	50
2.61	60	202	2.62	30
2.51	20	301	2.52	10
2.24	80	130	2.25	30
2.13	30	131	2.14	10
2.06	10	113	2.06	10
1.99	20	203	1.99	10
1.93	80	222	1.94	30
			1.93	20
1.88	30	132	1.88	. 20
			1.879	10
1.83	80	123	1.84	40
			1.83	30
1.79	30	231	1.798	30
			1.794	20
1.76	50	140	1.771	
			1.767	10
1.74	50	402	1.749	20
	50	102	1.745	10
1.72	50	004	1.721	30
	20		1.717	20
1.630	30	232	1.638	10
1.000			1.634	10
1.603	5	133	1.607	10
1.005	5	100	1.007	10

Table 11 X-ray powder diffraction data for wilkeite.

tified that of montomorillonite and Al-chlorite by MITSUDA (1954). On the other hand, the minerals from the Kyoritsu-Nako mine and the Kuwano-Ohira mine was identified the mixed layered mineral of montmorillonite and mica according to the 25 Å reflection and its subsequent orders.

### Other minerals

Quartz, hematite, and pyrite are common minerals in the hydrothermally altered rocks. Zircon, rutile, apatite, chlorite, and others occur as accessory minerals.

# 6. Textural Relation and Paragenetic Sequence

Paragenetic sequences were estimated from cross-cutting relationship of ores and minerals observed in fields and optical investigation of about 150 thin sections. As shown in Table 13, there are also remarkable differences on the mineral assemblages of the individual ore deposit.

Uku mine; Andalusite-pyrophyllite-kaolinite subzone of this mine consists mainly of andalusite (25.6% in volume), pyrophyllite (28.5%), and kaolinite (17.0%). Accessory minerals are quartz, corundum, and diaspore. In this subzone, grain size of andalusite is usually larger than that of the quartz-andalusite subzone. Almost all of andalusite has been conspicuously replaced by pyrophyllite and kaolinite along cleavage and crack. It is also replaced
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A: Andalusite Au: Augelite K: Kaolinite M: Muscovite Fig. 19 X-ray diffraction patterns of muscovite, dumortierite, topaz, augelite and wilkeite.

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by coarse-grained muscovite showing fan-shaped texture. Corundum is present in a large crystal of andalusite and along grain boundary. It has been commonly replaced by kaolinite, and more rarely by diaspore. Quartz shows resorbed figures, most of which are considered as remnant of the original rocks.

Quartz-andalusite-pyrophyllite subzone is mainly composed of quartz (37.0%), and alusite (24.0%), and pyrophyllite (29.7%) and is accompanied with small amounts of diaspore, kaolinite, and corundum. In this subzone andalusite and corundum have been slightly altered to pyrophyllite and kaolinite. They coexist often with quartz and diaspore. A small amount of pyrite disseminates locally, but hematite is scarecely present.

Quartz-andalusite subzone consists mostly of quartz (59.1% and andalusite (36.4%). Very small amounts of pyrophyllite, kaolinite, diaspore, and corundum are scattered in quartzandalusite mass. Irregular-shaped spongy andalusite, being up to 0.5 mm in diameter, in-

mineral sample	Andalusite	Pyrophyllite	Kaolinite	Diaspore	Corundum	Quartz	Subzone
111806 111906 111907 111911 Avg.	23.8 41.1 33.2 47.4 36.4	$ \begin{array}{r} 2.4 \\ 1.3 \\ 1.5 \\ - \\ 1.3 \end{array} $	$0.4 \\ 1.9 \\ \\ 1.4 \\ 0.9$	4.3  1.5 1.4	$\frac{-}{3.6}$	69.1 55.7 61.7 49.7 59.1	Q-A subzone
111909 111910 12U <sub>7</sub> 10 31115 Avg.	21.9 13.4 36.7 24.0 24.0	39.6 42.7 11.0 25.4 29.7	5.6 2.2 0.5 3.4 2.9	4.3 13.3 2.4 4.9		32.9 37.4 38.5 39.3 37.0	Q-A-P subzone
31101 31103 62U-C 51025 Avg.	35.6 30.0 6.7 30.0 25.6	9.0 23.5 62.7 18.6 28.5	23.4 5.8 12.0 26.7 17.0	1.7 3.0 18.6 14.1 9.4	$     \frac{             1.9}{$	28.4 37.7 9.6 18.9	A-P-K subzone

Table 1	12	Modal	composition	(vol.	%)	of	altered	rocks	of	the	andalusite	zone in	1 the	Uku	mine.
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Q; quartz, A; and alusite, P; pyrophyllite, K; kaolinite.

Table 13 Mineral assemblages of Roseki ore deposits in the Abu district.

												•	
Mineral assemblage Deposit	А	A·P	A·P·K	А∙Р∙К∙М	А∙К∙М	Α·К	Å·M	A·P·M	м·к	м∙к∙р	M · P	Р·К	P
Uku	0	0	۲	0	Δ	Δ	۰	0	0	0	•	0	٥
Kyoritsu-nako	•	٥	۲	۲	0	Δ.	•		0	0			
Ohira	٥	•		Δ	0	Ō	۲	Δ	0	0		0	
Hinomaru-nako	•		Δ	0	۲	Δ.	0		0				
Kochi									Δ.	۲	0	۲	۲
A:Andalusite	ł	K:Kaol	inite	.M: Mu	iscovi	te F	: Pyro	phylli	te				
Predominar	nt	0	Com	mon		ΛPo	or	e Ra	r 0				





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cludes commonly fine-grained quartz with a maximum diameter of about 0.05 mm. Andalusite and original quartz grains have been scarcely replaced by clay minerals such as pyrophyllite and kaolinite. Pyrite disseminates mostly in the lower part of the subzone. The grains contact usually with andalusite and quartz, and rarely with pyrophyllite. Hemetite occurs in the upper part as the same manner as the pyrite.

From mineral associations, textural relations, and field occurrences stated above, hydrothermal action can be divided into three stages (Figure 20).

The early stage is characterized by mineral assemblages such as andalusite-corundum and andalusite-quartz. The former has been principally fromed in the central part of the andalusite zone. The latter is representative in the margin. Small amounts of andalusitediaspore and andalusite-pyrophyllite are also stable in this stage.

The middle stage is represented by assemblages of pyrophyllite-diaspore-kaolinite and pyrophyllite-kaolinite-quartz. The hydrothermal action of this stage is confined to a part of the andalusite zone, and areal extent of the action is much smaller than that of the early stage. These minerals occur frequently as veinlets cutting the altered rocks of the early stage. Where abundant pyrophyllite veinlets have been formed, pre-existed andalusite is perfectly replaced. The decomposition reaction is assumed as follows;

There are also many kaolinite bands of a several millimeters in width in the andalusitepyrophyllite-kaolinite subzone. This narrow kaolinite bands—a sort of reaction rim—are considered to have been formed by reaction between the preexisted andalusite and the solution of the middle stage as follows;

$$\begin{array}{c} Al_2SiO_5 + SiO_2 + 2H_2O = Al_2Si_2O_5(OH)_4 \dots (2) \\ and alusite \\ kaolinite \end{array}$$

The late stage is represented by few veins consisting of kaolinite-muscovite and kaolinitemuscovite-mixed layered mineral. These veins cut the altered rocks of the early and the middle stages. The hydrothermal action is much smaller in scale than that of the middle stage.

**Hinomaru-Nako mine;** In the andalusite zone of Gagaradani ore deposit, assemblages of andalusite-corundum, andalusite-pyrophyllite (-diaspore), and andalusite-quartz are thought to have been formed in the early stage. Corundum and diaspore occur generally in the central part of this andalusite zone. Corundum veinlets and lenses of a several to 10 millimeters in width occur often along some fractures in the quartz-andalusite subzone. Therefore, some corundum may have been locally formed in the middle stage. Microscopic relations of those minerals are similar to those observed at Uku mine.

Pyrophyllite and kaolinite are universally stable phases of the middle stage. Muscovitepyrophyllite-kaolinite, which is locally predominant, is considered to be product of this stage. Pyrophyllite-kaolinite-quartz and pyrophyllite-quartz are very common assemblages, but pyrophyllite-diaspore is not observed.

Mixed layered mineral as found in Uku mine is not recognized in this ore deposit. It is therefore considered that the hydrothermal action of the late stage was conspicuously deficient.

In the South pits, alunite and andalusite zones are cut by dickite veinlets of a few mil-

limeters in width. But crosscutting relationship between the alunite and andalusite zones has not been observed. Euhedral alunite coexists with fine-grained lath-shaped kaolinite and finegrained quartz. Andalusite has been largely decomposed to pyrophyllite-kaolinite (-quartz) assemblage.

In the andalusite zone of the West ore deposit, assemblages of andalusite-quartz and andalusite-muscovite have been formed in the early stage. But most muscovites coexist with kaolinite as replacement products of andalusite. Pyrophyllite was faintly recognized by x-ray diffraction. Paragenetic sequence of muscovite-kaolinite is thought to have been in the middle stage. In greisen, euhedral andalusite, euhedral to subhedral topaz, and mozaic quartz being main constituents seems to have been formed at approximately the same stage according to their textural relations. Small amount of muscovite and diaspore are found as replacement products after andalusite. The greisen zone and andalusite zone have been cut by dickite veinlets having a width of a few millimeters.

**Kyoritsu-Nako mine;** In the andalusite-pyrophyllite-kaolinite subzone of the No. 1 ore deposit, subhedral andalusite being 1.0 by 1.5 millimeters in size and diaspore are commonly replaced by pyrophyllite and kaolinite, and rarely by muscovite. Pyrophyllite and kaolinite occur not only as decomposition products after andalusite but as veinlet-forming minerals. Corundum is seen scarcely. Euhedral wilkeite has been formed in pyrophyllite-kaolinite aggregates. It is usually replaced by pyrophyllite along cracks and rim of the crystal. Assemblages of andalusite-pyrophyllite, andalusite-quartz, and pyrophyllite-kaolinite-quartz are rarely cut by slender kaolinite veinlets.

Microscopic textural relations of the quartz-andalusite-pyrophyllite subzone are similar to those of the andalusite-pyrophyllite-kaolinite subzone. In the quartz-andalusite subzone, fine-grained spongy andalusite includes commonly subhedral fine-grained quartz. Andalusite has been hardly altered to pyrophyllite, kaolinite, and muscovite. Quartz is usually mozaic in texture except for resorbed phenocrystlike crystals. Small amount of diaspore and pyrophyllite occur along grain boundaries between andalusite and quartz.

In the quartz-andalusite-quartz subzone of the No. 3 ore deposit, andalusite has been universally replaced by assemblage of pyrophyllite-kaolinite (-quartz). In many cases, it occurs as irregular-shaped relicts. Corundum and diaspore are not seen everywhere. Where this subzone is cut by kaolinite-muscovite-mixed layered mineral veins, kaolinite and muscovite increase and disseminated pyrite decrease distinctly.

As stated above, assemblages of andalusite quartz and andalusite-diaspore are stable in the early stage of the No. 1 ore deposit. But pyrophyllite-kaolinite in the middle stage is much smaller in amount compared with the No. 3 ore deposit. These indicate that the hydro-thermal solution of lower temperature from the middle to the late stage infiltrated little into the andalusite zone. Pyrophyllite-kaolinite and pyrophyllite-quartz have been formed abundant in the middle stage of the No. 3 ore deposit. In the late stage, kaolinite-mixed layered minerals and kaolinite-muscovite have formed as vein forming minerals in the quartz-andalusite-pyrophyllite subzone.

Kuwano-Ohira mine; In the andalusite-kaolinite-muscovite subzone of this mine, andalusite has been remarkably replaced by kaolinite and often replaced by muscovite. The decomposition reaction of pre-existed andalusite and potassium rich solution is supposed as follows;

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# $3(Al_2SiO_5)+3SiO_2+K_2O+2H_2O=2[KAl_2(AlSi_3O_{10})(OH)_2]....(3)$ and a lusite muscovite

Large fan-shaped muscovite occasionally coexists with euhedral to subhedral andalusite. A small amount of pyrophyllite was confirmed only by x-ray diffraction, but its texture and textural relation with other minerals could not been observed. Quartz having from 0.1 to 0.3 millimeters in size shows commonly a mosaic in texture. No corundum, diaspore, and hematite are found.

In the quartz-andalusite-muscovite subzone, euhedral andalusite is slightly replaced by muscovite and kaolinite. Muscovite occurs mostly at grain boundaries between andalusite and quartz.

Some clayey veins comprising kaolinite-quartz-mixed layered mineral and muscovitequartz-mixed layered mineral have invaded into both subzones.

Stable mineral assemblages of the early st ge are mainly andalusite-quartz and andalusitemuscovite. Some of andalusite-pyrophyllite may have been formed in this stage. Muscovite and kaolinite characterized the middle stage. In the late stage, muscovite-kaolinite(-quartz)mixed layered minerals are formed.

In Torinosu and Kiyodani ore deposits, main mineral assemblages are similar to those of Kuwano-Ohira. The mineral assemblages being rich in muscovite may suggest higher K/H ratio in the solution throughout all stages of the hydrothermal action of these deposits.

Kōchi ore deposits; Main stage of the hydrothermal alteration is represented by assemblage of pyrophyllite-kaolinite-quartz. In some place, very small amounts of andalusite and corundum occur in pyrophyllite-kaolinite aggre ate. Diaspore is also present often as irregular-shaped relic. All of these minerals have been remarkably replaced by pyrophyllite and/or kaolinite.

These textural relationships indicate that and alusite, corundum, and some of diaspore were formed in advance with the hydrothermal action of the main stage.

#### 7. Migration of Chemical Components

Chemical analyses were conducted made on 15 samples in order to know loss and gain of the hydrothermally altered rocks. Three samples of tuffaceous sandstone and siltstone in the lacustrine bed of the Kiyodani member demonstrate the original rocks. All of them contain slightly fine-grained sericite crystals in the matrix, although the most fresh-looking rocks were selected from the surveyed area. The hydrothermally altered rocks were collected from the three andalusite subzones and the sericite zone of the Central ore deposit and its periphery of Uku mine.

Chemical compositions of 15 samples are shown in Table 14, and they are recalculated in Tables 15 and 16. Figure 21 is variation diagrams for the averages of the each subzones and the sericite zone. The loss and gain are graphically represented in Figure 22.

As is evident from the Figure 21, increase of  $H_2O$  and  $Al_2O_3$  and decrease of FeO, CaO, and Na<sub>2</sub>O are remarkable throughout the whole of the hydrothermally altered zone. SiO<sub>2</sub> changes scarcely in the andalusite zone as a whole, while  $K_2O$  decreases markedly in the same zone.

In the sericite zone,  $TiO_2$  and  $Fe_2O_3$  increases about 200 percent compared with the original rocks. Besides, there is conspicuous increase of  $H_2O(122\%)$ ,  $Al_2O_3$  (52%), and  $K_2O$ 

			Table 1	4 Chem	ical comp	position of	hydroth	ermally a	altered ro	cks in the	e Abu di	strict.			
	1**	2**	3**	4**	5**	6*	7*	8*	9**	10*	11*	12*	13*	14*	15**
	31122	31316	111722	111725	111803	111806	111906	111911	111913	111909	111910	31115	31101	31103	31111
SiO <sub>2</sub>	73.36	66.54	66.46	78.39	66.71	81.94	81.96	80.65	83.19	68.02	69.76	63.96	63.38	54.92	53.78
TiO <sub>2</sub>	0.31	0.56	0.65	0.30	0.19	0.16	0.46	0.10	0.15	0.30	0.08	0.14	0.04	0.17	0.16
Al <sub>2</sub> O <sub>3</sub>	14.19	22.66	20.78	13.59	20.56	15.00	15.24	18.40	13.70	26.12	23.90	31.26	31.14	36.69	37.26
Fe <sub>2</sub> O <sub>3</sub>	5.67	0.72	2.79	1.04	3.47	0.32	0.40	0.30	1.20	0.32	0.44	0.47	0.65	0.47	0.16
FeO	0.25	0.07	0.36	0.18	0.07	0.01	0.01	0.01	0.07	0.01	0.01	0.01	0.03	0.01	0.04
MgO	0.14	0.15	0.18	0.29	0.25	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
CaO	0.01	0.07	0.04	0.04	0.01	0.01	0.01	0.01	0.03	0.01	0.01	0.01	0.01	0.01	0.06
Na₂O	1.09	1.26	0.86	0.75	1.04	0.03	0.02	0.02	0.03	0.06	0.38	0.05	0.05	0.04	0.02
K₂O	2.76	4.86	4.55	2.89	4.48	0.23	0.03	0.03	0.27	0.06	1.29	0.23	0.14	0.22	0.18
S	0.01	0.01	0.03	0.01	0.01				0.01						0.03
H₂O+	1.90	2.85	2.77	2.11	2.83	2.14	1.62	0.62	1.11	5.02	3.83	3.64	3.93	6.97	7.95
$H_2O-$	0.02	0.08	0.26	0.18	0.14	0.14	0.04	0.16	0.14	0.12	0.18	0.38	0.16	0.36	0.18
Total	99.70	99.82	99.73	99.76	99.78	99.97	99.78	100.29	99.90	100.03	99.87	100.14	99.92	99.85	99.83

1,2,3,4,5; sericite zone, 6,7,8,9; quartz-andalusite subzone, 10,11,12; quartz-andalusite-pyrophyllite subzone, 13,14,15; andalusite-pyrophyllite-kaolinite subzone.

\*; analyzed by Mr. M. Kawano of Chemistry Section, Geological Survey of Japan.

\*\*; analyzed by Tokyo institute of Coal and Minerals.

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Sample	1	2	3	4	5		6	7	8	9	
lem.	31122	31316	111722	111725	111803	Avg.	111806	111906	111911	111913	Avg.
Si	35.07	32.10	32.14	37.59	32.21	33.82	39.21	39.05	37.88	39.42	38.89
Ti	0.19	0.35	0.40	0.18	0.12	0.25	0.10	0.28	0.26	0.09	0.18
Al	7.68	12.38	11.38	7.38	11.24	10.01	8.13	8.22	9.79	7.35	8.37
Fe <sup>3+</sup>	4.06	0.52	2.00	0.75	2.51	1.97	0.23	0.29	0.21	0.85	0.40
Fe <sup>2+</sup>	0.14	0.04	0.20	0.10	0.04	0.10	0.01	0.01	0.01	0.04	0.02
Mg	0.09	0.09	0.11	0.66	0.16	0.22	Tr	Tr	Tr	Tr	Tr
Ca	Tr	0.05	0.03	0.03	0.03	0.03	Tr	Tr	Tr	0.02	0.01
Na	0.83	0.96	0.66	0.57	0.80	0.76	0.02	0.02	0.01	0.02	0.02
K	2.34	4.16	3.91	2.46	3.60	3.29	0.20	0.03	0.03	0.23	0.12
0	49.60	49.35	50.83	49.12	49.29	49.83	52.10	52.10	52.01	51.98	52.05
										100.00	100.00
Total	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00
Total Sample	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00
Total Sample	100.00 10 111909	100.00 11 111910	100.00 12 31115	100.00 Avg.	100.00 13 31101	100.00 14 31103	100.00 15 31111	100.00 Avg.	100.00	* .	Average of
Total Sample Iem.	100.00 10 111909 33.51	100.00 11 111910 34.02	100.00 12 31115 31.10	100.00 Avg. 32.88	100.00 13 31101 30.92	100.00 14 31103 27.15	100.00 15 31111 27.13	100.00 Avg. 28.70	100.00	100.00	Average of original rocks
Total Sample lem. Si Ti	100.00 10 111909 33.51 0.20	100.00 11 111910 34.02 0.05	100.00 12 31115 31.10 0.09	100.00 Avg. 32.88 0.11	100.00 13 31101 30.92 0.28	100.00 14 31103 27.15 0.11	100.00 15 31111 27.13 0.10	100.00 Avg. 28.70 0.16	100.00 16* 35.74 0.09	<u>100.00</u>	Average of original rocks
Total Sample lem. Si Ti Al	100.00 10 111909 33.51 0.20 14.57	100.00 11 111910 34.02 0.05 13.20	100.00 12 31115 31.10 0.09 17.21	100.00 Avg. 32.88 0.11 14.99	100.00 13 31101 30.92 0.28 17.20	100.00 14 31103 27.15 0.11 20.99	100.00 15 31111 27.13 0.10 21.52	Avg. 28.70 0.16 19.90	100.00 16* 35.74 0.09 6.80	<u>100.00</u>	Average of original rocks
Total Sample lem. Si Ti Al Fe <sup>3+</sup>	100.00 10 111909 33.51 0.20 14.57 0.24	100.00 11 111910 34.02 0.05 13.20 0.32	100.00 12 31115 31.10 0.09 17.21 0.34	Avg. 32.88 0.11 14.99 0.30	100.00 13 31101 30.92 0.28 17.20 0.47	100.00 14 31103 27.15 0.11 20.99 0.36	100.00 15 31111 27.13 0.10 21.52 0.10	Avg. 28.70 0.16 19.90 0.31	100.00 16* 35.74 0.09 6.80 0.62	<u>100.00</u>	Average of original rocks
Total Sample Iem. Si Ti Al Fe <sup>3+</sup> Fe <sup>2+</sup>	100.00 10 111909 33.51 0.20 14.57 0.24 0.01	100.00 11 111910 34.02 0.05 13.20 0.32 0.01	100.00 12 31115 31.10 0.09 17.21 0.34 0.01	Avg. 32.88 0.11 14.99 0.30 0.01	100.00 13 31101 30.92 0.28 17.20 0.47 0.02	100.00 14 31103 27.15 0.11 20.99 0.36 0.01	100.00 15 31111 27.13 0.10 21.52 0.10 0.02	Avg. 28.70 0.16 19.90 0.31 0.02	100.00 16* 35.74 0.09 6.80 0.62 0.25	<u> </u>	Average of original rocks
Total Sample Iem. Si Ti Al Fe <sup>3+</sup> Fe <sup>2+</sup> Mg	100.00 10 111909 33.51 0.20 14.57 0.24 0.01 Tr	100.00 11 111910 34.02 0.05 13.20 0.32 0.01 Tr	100.00 12 31115 31.10 0.09 17.21 0.34 0.01 Tr	Avg. 32.88 0.11 14.99 0.30 0.01 Tr	100.00 13 31101 30.92 0.28 17.20 0.47 0.02 Tr	100.00 14 31103 27.15 0.11 20.99 0.36 0.01 Tr	100.00 15 31111 27.13 0.10 21.52 0.10 0.02 Tr	Avg. 28.70 0.16 19.90 0.31 0.02 Tr	100.00 16* 35.74 0.09 6.80 0.62 0.25 0.15	<u> </u>	Average of original rocks
Total Sample Ilem. Si Ti Al Fe <sup>3+</sup> Fe <sup>2+</sup> Mg Ca	100.00 10 111909 33.51 0.20 14.57 0.24 0.01 Tr Tr	100.00 11 111910 34.02 0.05 13.20 0.32 0.01 Tr Tr	100.00 12 31115 31.10 0.09 17.21 0.34 0.01 Tr Tr Tr	Avg. 32.88 0.11 14.99 0.30 0.01 Tr Tr	100.00 13 31101 30.92 0.28 17.20 0.47 0.02 Tr Tr Tr	100.00 14 31103 27.15 0.11 20.99 0.36 0.01 Tr Tr	100.00 15 31111 27.13 0.10 21.52 0.10 0.02 Tr 0.05	Avg. 28.70 0.16 19.90 0.31 0.02 Tr 0.02	100.00 16* 35.74 0.09 6.80 0.62 0.25 0.15 0.30	<u> </u>	Average of original rocks
Total Sample Ilem. Si Ti Al Fe <sup>3+</sup> Fe <sup>2+</sup> Mg Ca Na	100.00 10 111909 33.51 0.20 14.57 0.24 0.01 Tr Tr 0.05	100.00 11 111910 34.02 0.05 13.20 0.32 0.01 Tr Tr 0.29	100.00 12 31115 31.10 0.09 17.21 0.34 0.01 Tr Tr 0.24	Avg. 32.88 0.11 14.99 0.30 0.01 Tr Tr 0.13	100.00 13 31101 30.92 0.28 17.20 0.47 0.02 Tr Tr 0.04	100.00 14 31103 27.15 0.11 20.99 0.36 0.01 Tr Tr 0.03	100.00 15 31111 27.13 0.10 21.52 0.10 0.02 Tr 0.05 0.02	Avg. 28.70 0.16 19.90 0.31 0.02 Tr 0.02 0.03	100.00 16* 35.74 0.09 6.80 0.62 0.25 0.15 0.30 2.57	100.00	Average of original rock
Total Sample Ilem. Si Ti Al Fe <sup>3+</sup> Fe <sup>2+</sup> Mg Ca Na K	100.00 10 111909 33.51 0.20 14.57 0.24 0.01 Tr Tr 0.05 0.05	100.00 11 111910 34.02 0.05 13.20 0.32 0.01 Tr Tr 0.29 1.12	100.00 12 31115 31.10 0.09 17.21 0.34 0.01 Tr Tr 0.24 0.20	Avg. 32.88 0.11 14.99 0.30 0.01 Tr Tr 0.13 0.46	100.00 13 31101 30.92 0.28 17.20 0.47 0.02 Tr Tr 0.04 0.12	100.00 14 31103 27.15 0.11 20.99 0.36 0.01 Tr Tr 0.03 0.02	100.00 15 31111 27.13 0.10 21.52 0.10 0.02 Tr 0.05 0.02 0.16	Avg. 28.70 0.16 19.90 0.31 0.02 Tr 0.02 0.03 0.10	100.00 16* 35.74 0.09 6.80 0.62 0.25 0.15 0.30 2.57 2.73	100.00	Average of original rock
Total Sample Iem. Si Ti Al Fe <sup>3+</sup> Fe <sup>2+</sup> Mg Ca Na K O	100.00 10 111909 33.51 0.20 14.57 0.24 0.01 Tr Tr 0.05 0.05 51.37	100.00 11 111910 34.02 0.05 13.20 0.32 0.01 Tr Tr 0.29 1.12 50.99	100.00 12 31115 31.10 0.09 17.21 0.34 0.01 Tr Tr 0.24 0.20 51.10	Avg. 32.88 0.11 14.99 0.30 0.01 Tr Tr 0.13 0.46 51.15	100.00 13 31101 30.92 0.28 17.20 0.47 0.02 Tr Tr 0.04 0.12 50.95	100.00 14 31103 27.15 0.11 20.99 0.36 0.01 Tr Tr 0.03 0.02 50.55	100.00 15 31111 27.13 0.10 21.52 0.10 0.02 Tr 0.05 0.02 0.16 50.60	Avg. 28.70 0.16 19.90 0.31 0.02 Tr 0.02 0.03 0.10 50.70	100.00 16* 35.74 0.09 6.80 0.62 0.25 0.15 0.30 2.57 2.73 50.75	100.00	Average of original rocks

Table 15 Reculculated chemical composition of original rocks and hydrothermally altered rocks.

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sample			S	Sericite zo	one (mg/c	c)			Q-A subzone (mg/cc)						
comp.	31122	31316	111722	111725	111803	Avg.	$\pm$ mg/cc	(%)	111806	111911	111906	111913	Avg.	$\pm$ mg/cc	(%)
SiO <sub>2</sub>	2049.6	1818.8	1749.7	2094.7	1805.3	1903.6	+ 49.0	- 2.4	2206.5	2228.0	2247.0	2265.9	2236.9	+290.2	+ 14.9
TiO <sub>2</sub>	8.6	15.3	17.1	8.0	5.1	10.8	+ 7.0	+184.2	4.3	2.8	12.6	4.1	6.0	+ 2.2	+ 57.9
Al <sub>2</sub> O <sub>3</sub>	394.3	619.4	547.1	363.2	556.4	496.1	+169.2	+ 51.8	403.9	508.3	417.8	373.2	425.8	+ 98.9	+ 30.3
Fe <sub>2</sub> O <sub>3</sub>	157.6	19.7	72.7	27.8	93.9	74.3	+ 51.6	+227.3	8.6	8.3	11.0	32.7	15.2	- 7.5	- 33.0
FeO	6.9	1.9	9.5	4.8	1.9	5.0	- 6.7	- 57.3	0.3	0.3	0.3	1.9	0.7	- 11.0	— 94.0
MgO	3.9	4.1	4.7	7.7	6.8	5.4	- 0.9	- 14.3	Tr	Tr	Tr	0.3	0.1	- 6.2	- 98.4
CaO	0.3	1.9	1.1	1.1	1.1	1.1	- 9.6	— 89.7	Tr	Tr	Tr	0.8	0.2	- 10.5	- 98.1
Na <sub>2</sub> O	30.3	34.4	22.6	20.0	28.1	27.1	- 61.2	69.3	0.8	0.6	0.5	0.8	0.7	— 87.6	- 99.2
K₂O	76.7	132.8	119.8	77.2	121.2	105.5	+ 21.7	+ 25.9	6.2	0.8	0.8	7.4	3.8	- 80.0	- 95.5
H <sub>2</sub> O+	52.8	77.9	72.9	56.4	76.6	67.3	+ 37.0	+122.1	57.6	17.1	44.4	30.2	37.3	+ 7.0	+ 23.1
H₂O−	0.6	2.2	6.8	4.8	3.8	3.6	- 4.3	- 54.4	3.8	4.4	1.1	3.8	3.3	- 4.6	- 58.2
Ap. G.	2.78	2.73	2.63	2.67	2.71	2.70	)		2.69	2.76	2.74	2.72	2.7	3	

Table 16 Loss and gain of elements of hydrothermally altered rocks in the Uku mine.

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sample		Q –	A - P sub	ozone (m	g/cc)		A - P - K subzone (mg/cc)						
comp.	111909	111910	31115	Avg.	± mg/cc	(%)	31101	31111	31103	Avg.	$\pm$ mg/cc	(%)	mg/cc
SiO <sub>2</sub>	1878.2	1905.5	1786.9	1856.9	- 89.8	- 4.6	1759.4	1495.9	1435.6	1563.6	-383.1	- 19.7	1946.7
TiO₂	8.3	2.2	3.9	. 4.8	+ 1.0	+ 26.3	12.2	4.5	4.4	7.0	+ 3.2	+ 84.2	3.8
$Al_2O_3$	721.2	652.8	873.3	749.1	+422.2	+129.2	864.4	1036.4	959.1	953.3	+626.4	+191.6	326.9
$Fe_2O_3$	8.8	12.0	13.1	11.3	- 11.4	- 50.2	18.0	4.5	12.3	11.6	- 11.1	- 48.9	22.7
FeO	0.3	0.3	0.3	0.3	- 6.0	— 51.3	0.8	1.1	0.3	0.7	— 10.0	85.5	11.7
MgO	Tr	Tr	Tr	Tr	- 6.3	-100.0	Tr	0.3	Tr	0.1	- 6.2	- 98.4	6.3
CaO	Tr	Tr	Tr	Tr	— 10.7	100.0	Tr	1.7	Tr	0.6	- 10.1	94.4	10.7
Na <sub>2</sub> O	1.7	10.4	1.4	4.5	- 83.8	— 94.9	1.4	0.6	1.0	1.0	- 87.3	- 98.9	88.3
K₂O	1.7	35.2	6.4	14.4	- 69.4	- 82.8	3.9	5.0	5.8	4.9	— 78.9	- 94.2	83.8
H₂O+	138.6	104.6	101.7	115.0	+ 84.7	+229.5	109.1	221.1	182.2	170.8	+140.5	+463.7	30.3
H₂O−	3.3	4.9	10.6	6.3	— 1.6	- 20.3	4.4	5.0	9.4	6.3	- 1.6	- 20.3	7.9
Ap. G.	2.74	2.73	2.79	2.75			2.78	2.78	2.62	2.73	3		2.54

Genesis of the Andalusite-bearing Roseki Ore Deposits (Masaharu KAMITANI)

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(26%). Decrease of CaO, Na<sub>2</sub>O, and FeO is -10%, -31%, and -43%, respectively (Figure 22). Almost all of SiO<sub>2</sub> and MgO seem to have remained in the sericite zone.

In the quartz-andalusite subzone,  $TiO_2$ ,  $Al_2O_3$ ,  $H_2O_3$ ,  $H_2O_2$  increase in small quantities. Almost all of FeO, MgO, CaO, Na<sub>2</sub>O, and K<sub>2</sub>O decrease. Therefore, it is thought that these components were leached out from the original rocks for this subzone.

There is not so much difference on the chemical compositions between the quartz-andalusite-pyrophyllite subzone and the andalusite-pyrophyllite-kaolinite subzone. MgO, CaO, Na<sub>2</sub>O, and K<sub>2</sub>O of the original rocks have been perfectly removed in both subzone. However, addition of  $Al_2O_3$  and  $H_2O$  to the andalusite-pyrophyllite-kaolinite subzone is extremely remarkable.

The mineral assemblages and the compositional variations of Gagaradani ore deposit of Hinomaru-Nako mine and the No. 1 and No. 3 ore deposits of Kyoritsu-Nako mine are similar to those of Uku mine. The andalusite zones of Type B of Kuwano-Ohira mine, Torinosu, and Kiyodani ore deposits are characterized by large quantity of potassium. Except for  $K_2O$ , the compositional variations of these deposits resemble to those of the altered rocks of Uku mine. It is assumed that the potassium was flooded throughout all stages of the hydrothermal alteration and thus formed the muscovite dominant andalusite zones of Type B.

The compositional variations in Abu district are similar to the deeper part of Ōtake





Fig. 22 Loss and gain of hydrothermally altered rocks in the Uku mine.

geothermal area, Kyushu (HAYASHI, 1973), and Nishidani Roseki ore deposit, Gotō mine (MINATO, 1965), but not similar to Ugusu silica deposit (IWAO, 1963) and Bonten-yama Roseki ore deposit (FUJII, 1963), both of which were formed near the surface by reaction between strong acid solution and wall rocks.

# 8. Potassium Argon Ages of Roseki Ore Deposit and Related Igneous Rocks

Isotopic age determination concerning on Roseki ore deposits exclusive of Yagi mine in Mitsuishi district, Okayama Prefecture (SHIBATA and FUJII, 1971), has not yet been carried out, because of difficulty of obtaining suitable materials.

The age of formation has been estimated by stratigraphy of the host volcano-sedimentary rocks, although even plant fossils are very rare. Some predicted the age by comparison of chemical compositions of volcanic rocks (SHIBATA *et al.*, 1967).

Unlike other Roseki ore deposits, tabular muscovites are common in Roseki ore deposits of this district. Muscovites for the K-Ar age determination were collected from No. 3–1 open pit and No. 2 open pit in the Central ore deposit of Uku mine (Figures 8 and 15, Sample numbers; U-31203A, U-31207). This muscovite occurs in pyrophyllite-bearing kaolinite-muscovite vein that cut through the central part of the andalusite zone. The analysed samples demonstrate the late stage of the hydrothermal alteration.

Another sample was collected from pegmatite occurring as a small lenticular mass in the biotite adamellite which is exposed along the northern coast of Uku mine (UP-1, Figure 8). Muscovite is pale blue in color, coarse-grained and has tabular, habit. The pegmatite represents the latest stage of consolidation of the biotite adamellite.

From the host volcanic rocks, a suitable material of pumiceous lens was found in the rhyolite welded tuff of the Mitsugadake rhyolite member, the Fukuga formation, and was provided for the whole rock determination to make clear the time of the accumulation. The rhyolite welded tuff, constituting the upper part of the Mitsugadake rhyolite member, has been strongly welded and contains large quantity of pumice fragments. The pumice consists principally of quartz and plagioclase phenocrysts and its matrix has perfectly devitrified. Fine-grained lath-shaped sericite scatters in the plagioclase phenocryst and matrix. The sample was collected from road cut along Yamanokuchi river situated at about 1,200 meters southeast of Mt. Mitsugadake (Sample number; 7330305, Figure 8).

Sample	Locality	Rock type	Mineral	K2O (%)	Atm. <sup>40</sup> Ar (%)	Age (m.y.)
1) U-31203A	Central ore deposit of the Uku mine	Hydrothermally altered rock	Muscovite	50.5	17.1	82.4±2.7
2) U-31207	Central ore deposit of the Uku mine	Hydrothermally altered rock	Muscovite	9.77	4.8	81.9±2.6
3) 7330305	Southeast of Mt. Mitsugadake (566.2 m)	Rhyolite welded tuff	Whole rock (pumice)	4.70	23.8	65.6±2.2
4) UP-1	Northern part of the Uku mine	Pegmatite in b:otite adamel- lite	Muscovite	10.33	11.7	79.0±2.6

Table 17 K-Ar ages of the andalusite-bearing Roseki ore deposits and related igneous rocks.

After Shibata and Kamitani (1974b)

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The results of four determinations are listed in Table 17. Muscovites from the pegmatite (79.0 m. y.) and the Roseki ore deposit (81.9 and 82.4 m. y.) give almost identical age, indicating similar age of intrusion and hydrothermal alteration, which are correlated to the Santonian of the Upper Cretaceous. Whole rock age of 65.6 m. y. on the pumice, on the other hand, is younger than the stratigraphically estimated age. The Abu group including the Fukuga formation has been correlated to Nöhi, Aioi, and Takata rhyolites (95–85 m. y.) by RESEARCH GROUP FOR THE LATE MESOZOIC IGNEOUS ACTIVITY IN THE INNER ZONE OF SOUTH-WEST JAPAN (1967). Moreover, the Fuguga formation is clearly intruded by the biotite adamellite (79.0 m. y.) in this district, as mentioned previously. Therefore, any given age for the Fukuga formation should be older than that of the adamellite. The younger age of about 66 m. y. may be originated in devitrification and weak sericitization of the rhyolite welded tuff. However, definite explanation cannot be given until more chronological data are available.

# 9. Genesis of Roseki Ore Deposit

# 9.1 Geologic Setting and Depth of Hydrothermally Altered Zone

Occurrences of main Roseki ore deposits and those altered zones have been stated previously in detail; that is, the andalusite zone of Uku mine occurs concordantly with the structure of the Kiyodani member and has more or less stratiform in shape. The andalusite zones of Hinomaru-Nako mine, Kyoritsu-Nako mine, Kuwano-Ohira mine, and the others, in many cases, occur in oblique to the bedding planes of the host rocks.

The original rock of the andalusite zones is entirely the Kiyodani member of the Fukuga formation. Furthermore, almost all of the andalusite zones except Kuwano-Ohira mine are located to the upper bed—the lacustrine sediment—of the Kiyodani member.

In general, the andalusite zone and pyrophyllite zone are surrounded by sericite zones. Weak sericitization is widespread in rhyolitic rocks of various units.

The andalusite zone, where the hydrothermal alteration was most intense, is confined to the Kiyodani member near the biotite adamellite stock. Net distances from the adamellite to each andalusite zone can be estimated concerning stratigraphy and geologic structure. The andalusite zone of the Central ore deposit of Uku mine is located 400 to 500 meters from the adamellite and Gagaradani ore deposit of Hinomaru-Nako mine and Kyoritsu-Nako mine have similar distances, 400 to 600 meters. The West ore deposit of Hinomaru-Nako mine is situated within 300 meters. In Kuwano-Ohira mine, some andalusite zones have been formed at 20 to 30 meters from the adamellite. Rocks of the Kiyodani member around Mt. Jingu, situated at a distance more than 1,000 meters from the adamellite, have been merely subjected to weak sericitization. Generally speaking, no andalusite zone has been observed at a distance more than 700 meters from the adamellite stock.

Depth of the hydrothermally altered zones at the time of formation can be estimated assuming i) the related igneous activity is of the adamellite for the very close spatial relationship mentioned above and ii) the adamellite is comagmatic with the Mitsugadake rhyolite member of the Fukuga formation for their similarity in bulk composition.

This estimation may give us a minimum depth, since the studied area may have been covered once by the Ōi rhyolite, which is distributed to the southwest of this area (MURA-KAMI and HASE, 1967). The maximum thickness of the Fukuga formation is about 1,800 meters (Table 2). Approximate depth of the andalusite zone can be estimated by removing

the thickness of the Kanai andesite member and the lower bed of the Kiyodani member because the andalusite zone occurs mostly in the upper bed of the Kiyodani member. It is also necessary to consider variable thickness of the rhyolite lava belonging to the Mitsugadake rhyolite member.

In Uku mine and its vicinity, the rhyolite lava having a thickness of about 100 meters covers the Kiyodani member. Consequently, the andalusite zone of this mine can be estimated to have been formed at the depth of 900 to 1,100 meters from the surface. In Gagaradani ore deposit of Hinomaru-Nako mine and No. 1, 2, and 3 ore deposits of Kyoritsu-Nako mine, the andalusite zones are supposed to have been formed at 800 to 1,000 meters in depth. They may have been slightly shallower in depth than that of Uku mine because the rhyolite lava occurs as very thin layer. The depths of the andalusite zones of Kuwano-Ohira and the West ore deposits are estimated within 1,200 meters on account of the rhyolite lava which is not existed and the andalusite zones occurring mainly in the lower bed of the Kiyodani member.

# 9. 2 Characteristics of Hydrothermal Alteration

There are clear differences on chemical compositions between the hydrothermally altered rocks and the original rocks.

In the quartz-andalusite subzone, FeO, MgO, CaO, Na<sub>2</sub>O, and K<sub>2</sub>O decrease strikingly. SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and H<sub>2</sub>O, on the otherhand, have a tendency to increase. Immobile components in the early stage are SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> (Figure 22). Increase of SiO<sub>2</sub> in this stage is also clear from assemblage of andalusite-quartz and silicified rocks which have been locally formed. While, alkali metal and alkaline earth metal oxides are remarkably leached from the original rocks. Consequently, it can be considered that chemical potential of SiO<sub>2</sub> in the hydrothermal solution was generally high and pH condition was parhaps acid to weak acid.

In the sericite zone,  $SiO_2$ ,  $Al_2O_3$ ,  $K_2O$ ,  $Fe_2O_3$ , and  $TiO_2$  can be regarded as immobile components. All the components except  $SiO_2$  have increased. As the result, sericite, hematite, pyrite, and rutile formed as alteration minerals in this zone. Almost all of CaO, MgO, and Na<sub>2</sub>O have been migrated to outside of the sericite zone. Parhaps, they had been discharged into surface throughout some conduits of solution.

Following the formation of the andalusite zone, pyrophyllite and kaolinite were formed in the middle stage of the alteration. It was revealed through the studies of HEMLEY (1959) and HEMLEY and JONES (1964) that the stability relations between pyrophyllite, kaolinite, and muscovite depend on temperature and K/H ratio. In this stage of the alteration of Uku mine, Gagaradani ore deposit of Hinomaru-Nako mine, and Kyoritsu-Nako mine, pyrophyllite and kaolinite are predominantly formed in the andalusite zone. While, in Kuwano-Ohira mine, Torinosu, Kiyodani, and West ore deposits are scanty in pyrophyllite. In these deposits, which are situated near the biotite adamellite, muscovite is stable phase rather than pyrophyllite under the same temperature and pressure. In the andalusite zones which were formed at a distance from the adamellite, pyrophyllite and kaolinite may have been stable on account of low K/H ratio.

Alunite zone is found in the South pit of Gagaradani ore deposit. Often alunite is formed around the surface (e. g. IWAO, 1963). However, in Matsukawa geothermal area, alunite zone has been developed at the depth of about 600 meters from the surface (SUMI, 1968). The rhyolite tuff bed, which is original rock of the alunite zone, must had been already

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covered by the rhyolite welded tuff of about several hundred meters thick when the alteration took place. Oxidation of solution might be activited through mixing of ascending solution and descending meteoric water from surface. Accordingly, the alunite zone seems to have been formed at some depth from the surface by sulfuric acid solution.

Pyrite and hematite formed during the early to the middle stage of the alteration. Pyrite is one of the most common minerals and only sulfide. It disseminates abundantly in the andalusite zones, which are formed close to the adamellite. Hematite is locally seen in the upper part of the andalusite zone of the Uku, Gagaradani, and Kōchi ore deposits. According to MAYER and HEMLEY (1967) stability relation of pyrite, chalcopyrite, magnetite, and hematite depends on fugacities of sulfur and oxygen. From the occurrences a fore mentioned, it is supposed that fugacity of sulfur was high close to the adamellite and oxygen, on the other hand, was sufficiently active at a distance from the adamellite.

#### 9. 3 Formation Temperature of Roseki Ore Deposits

# 9.3.1 Andalusite

The mineral assemblages in the early stage of the hydrothermal alteration are andalusite-corundum, andalusite-diaspore, andalusite-pyrophyllite, and andalusite-quartz.

There are many studies concerning synthesis and stability relation of andalusite and pyrophyllite since 1954 (Roy, 1954; Roy and Osborn, 1955; HEMLEY, 1959; KERRIC, 1968; ALTHAUS, 1969; etc.). The results are summarized in Figure 23.

The formation depth of the Roseki ore deposits (andalusite zones) in Abu district have been setimated within 1,200 meters from the surface, therefore, the calculated total pressures are 0.2 to 0.3 kilobar. The Vapor pressure is thought to be still lower than the total pressure. In general, hydrothermal syntheses have been made under the limited conditions such as total pressure equals to vapor pressure and its solution contains only a few kinds of





50-(250)

bases and ions. Also the reaction rate is very fast compared with that of the natural solution. Hence, the results from these experiments are inapplicable directly to natural minerals, although they are available for understanding of mutual relations between each mineral.

HEMLEY (1959) found that andalusite, pyrophyllite, and muscovite are in equilibrium at  $450^{\circ}$ C, 15,000 p. s. i. Based on the single crystal method, KERRIC (1968) estimated the phase boundary of pyrophyllite  $\rightleftharpoons$  andalusite+quartz passed through the points of  $430^{\circ}$ C, 3.9 kilobars and  $410^{\circ}$ C, 1.8 kilobars. According to ALTHAUS (1969), the equilibrium curve of andalusite and pyrophyllite is set on higher-temperature side than that of Kerric's curve (Figure 23). If Kerric's curve is extraporated to the low vapor pressure side and is applied for the estimation of formation temperature of the andalusite zone in Abu district, the temperature turns out below  $400^{\circ}$ C. The equilibrium relations of andalusite-pyrophyllite and andalusite-diaspore have been also confirmed by microscopic observation.

#### 9.3.2 Pyrophyllite

Pyrophyllite, diaspore, and kaolinite are representative minerals of the middle stage of the hydrothermal alteration. The assemblages are pyrophyllite-diaspore, pyrophyllite-kaolinite, and pyrophyllite-diaspore-kaolinite. They occur mainly as replacement minerals after andalusite.

There are many studies on hydrothermal syntheses and phase relation including pyrophyllite (Roy and OSBORN, 1955; HEMLEY, 1959; KENNEDY, 1959; ALTHAUS, 1969; THOMPSOH, 1971) and stability fields of pyrophyllite, kaolinite, and muscovite have been established. The phase relation between pyrophyllite and kaolinite was studied especially by Hemley, Althaus, and Thompson. According to Hemley's study, the stable relation of kaolinite-pyrophyllite-muscovite-potassium feldspar is controlled principally by K/H ratio. HAYASHI (1973), on the other hand, reported an occurrence of monoclinic pyrophyllite-diaspore-kaolinite association from Ōtake geothermal area, Ōita Prefecture, and extraporated Althaus's equilibrium curve of pyrophyllite-kaolinite into low-temperature and low pressure domain based on a thermodynamic calculation.

In Abu district, the formation depth of pyrophyllite is the same as that of andalusite. Hence, the total pressure is less than 0.3 kilobar and vapor pressure is perhaps far less than the total pressure. Thompson's curve (Figure 24), therefore, can be extraporated into the low-temperature domain, and may be applicable for the pyrophyllite-kaolinite assemblage of Abu district.

The occurrence of pyrophyllite, which was identified monoclinic pyrophyllite based on BRINDLEY and WARDLE (1971), and the association with other minerals in this district are similar to those reported from the other areas, such as Shobara (MATSUMOTO, 1968), Goto (IWAO *et al.*, 1953; MINATO, 1965), and Ōtake areas (HAYASHI, 1973). They may have been formed under the similar physical-chemical conditions.

# 9. 4 Related Igneous Activity to the Hydrothermal Alteration

In most of other Roseki ore deposits, genetically related igneous source is considered volcanic to subvolcanic activities. As is evident in the foregoing descriptions, however, the biotite adamellite is thought to be the heat source for the hydrothermal alteration in Abu district, because of the following geological and mineralogical evidences, as well as the K-Ar mineral ages of the adamellite and ore deposits, which are identical within the analytical error.

The intimate relation between the hydrothermal alteration and the biotite adamellite is



Fig. 24 Equilibirum curves of kaolinite and pyrophyllite according to various authers.

considered by the following evidences;

1) The Kiyodani member, which is close to the adamellite stock, has undergone intense hydrothermal alteration. Andalusite zones have been formed in such portion. The alteration becomes weak with the distance from the adamellite and it becomes also gradually small in scale. Generally speaking, the hydrothermally altered zones distributed around the adamellite stock show roughly a zonal arrangement from the inner side to outwards; andalusite zone, pyrophyllite zone, and sericite zone.

2) In the early stage of the alteration, assemblages of andalusite-corundum, andalusitediaspore, andalusite-pyrophyllite, and andalusite-quartz are formed. Pyrophyllite-diaspore (-kaolinite), pyrophyllite-kaolinite, and pyrophyllite-quartz characterize the middle stage. In the late stage kaolinite-muscovite and kaolinite-muscovite-mixed layered minerals are locally formed. It is suggested that the alteration in this district transfered retrogressively from hightemperature to low-temperature.

3) Andalusite has been formed during the early stage of the alteration and in many cases it is replaced by pyrophyllite, kaolinite, and muscovite in the middle to the late stage. These simple retrogressive patterns of 2) and 3) may be characteristics of plutonic type of cooling history rather than subvolcanic or volcanic type.

4) The mineral assemblage of kaolinite-muscovite-mixed layered minerals has been formed under lower temperature. However, this assemblage is still stable even though it had been formed close to the adamellite. Evidence that this assemblage was subjected to contact metamorphism by the adamellite has not been found anywhere.

5) Pyrite disseminates universally into the altered zone. But any evidence of thermal effect of the adamellite; e. g. pyrite converted to pyrrhotite, has not been found. The above 4) and 5) would reject the possibility of a syngenetic formation of the ore deposits with the

host volcanic rocks.

6) The greisen zone occurs in the central part of the andalusite zone of the West ore deposit. On the extension of the greisen zone dumortierite and wilkeite are formed in the same andalusite zone. The greisen zone is characteristic of granitic activities of the Sanyo belt.

Based on the evidences as stated above, the relationship between the hydrothermal alteration and the biotite adamellite can be concluded as follows;

The biotite adamellite intruded into the Fukuga formation of the Abu group. The top of the adamellite seems to have been consolidated at shallower depth of 1,500 to 1,800 meters from surface. Part of volatile components ( $H_2O$ ,  $H_2S$ , S,  $SO_2$ , F, P, B, etc.) separated from the adamellite magma ascended along some fractures and formed the greisen zone. In the andalusite zones near the adamellite, assemblage of andalusite-muscovite is predominant compared with other andalusite zones. Potassium fixed into muscovite was perhaps supplied from the adamellite. It is considered that the biotite adamellite played an important role as a heat-source and as supplier of volatiles to some extent for the hydrothermal alteration.

#### 10. Comparison with Other Hydrothermally Altered Zones

There are many, more than one hundred of, Roseki ore deposits in Japan. The Mitsuishi and Shobara districts of late Cretaceous in age are the most well-known Roseki provinces. Some ore deposits in Nagano Prefecture (FUJII and INOUE, 1970) and Toya, Hokkaido (NARI-TA *et al*, 1968) occur in andesitic rocks of Miocene age.

In South Korea, many pyrophyllite deposits have been recognized in upper Cretaceous acidic pyroclastic sediments (KINOSAKI, 1963a). Moreover, pyrophyllite deposits are distributed in North Carolina, U.S.A. (ZEN, 1961) and Coromandel, New Zealand (SWINDALE and HUGHES, 1968).

On the Roseki ore deposits in Chugoku, KINOSAKI (1963a) pointed out that the deposits were stratiform in shape and formed in the same rhyolitic bed simultaneously with the accumulation of host rock. Regarding to Mitsuishi-Daiyama, KATAYAMA (1969) proposed a model in which the deposit was formed at 500 to 1,000 meters under the surface through a convection of acidic solution. Iwao (1962, 63) explained the formation mechanism of the Ugusu silica deposit generated through the reaction between andesitic rocks and strongly acidic solution. Bontenyama Roseki ore deposit formed at surface was investigated by FUJII (1969).

Recently, interesting papers appeared in connection with geothermal areas, among which studies of Matsukawa (SUMI, 1968, 72) and Ōtake (HAYASHI, 1973) are most valuable.

#### **10. 1** Occurrences of Roseki Ore Deposits and Hydrothermally Altered Zones

Exposed areas of hydrothermally altered zones of main Roseki ore deposits and geothermal areas are from a few to several squre kilometers. Alteration zones can be roughly classified into two types, although they show striking variation. One is stratiform type that follows distinct stratigraphic horizon such as lacustrine sediment, and the other is irregularshaped type controled principally by faults and fractures.

The Roseki ore deposits of Mitsuishi and Shobara districts have been thought to be stratiform-shape by KINOSAKI (1963a),  $\overline{O}MORI$  (1965), and MATSUMOTO (1968). In Mitsu-

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ishi, however, it has been confirmed that porous silicified zones were formed oblique to the bedding plane of rhyolitic tuff (FUJII et al., 1971; KAMITANI and FUJII, 1972). Moreover, in Kamagamine mine, Shobara district, a pyrophyllite zone intersects bedding plane of original rock (KAMITANI, 1970). In many cases, main passage way of hydrothermal solution is controlled by some weakness such as fault and fracture.

In Ugusu silica deposit, Iwao (1972) inferred dikes and quartz veins in the host rocks as the passage-ways of the hydrothermal solution. The altered zone of Ōtake geothermal area is controlled by northwest-southeast trending faults (HAYASHI, 1973). Matsukawa geothermal area is vertically elongated because solution was confined to vertically narrow channels in the andesitic rocks (SUMI, 1968).

Andalusite-bearing Roseki ore deposits and hydrothermally altered zones of Abu district occur mainly in the Kiyodani member. Andalusite zones, as mentioned before, are mostly confined to the lacustrine sediment. In general, porosity of the lacustrine sediment is larger than that of other rocks. Therefore, connate water contents of this bed may be much abundant compared with the other rocks. Circulatoin and infiltration of meteoric water should be more probable in this sediment. Small faults and fractures seem to be the channel way for the andalusite-bearing Roseki ore deposits in Abu district.

#### 10. 2 Zonal Arrangement of Hydrothermally Altered Rocks

There are two types of the zonal arrangement of altered rocks. One is rich in  $Al_2O_3$ and the other is characterized by porous silicified rocks (Figure 25). Shobara district (MA-TSUMOTO, 1968), Goto mine (MINATO, 1965), and Abu district belong to the former and the



Fig. 25 Schematic comparison of the principal areas of hydrothermal alteration in Japan.

latter is seen in Mitsuishi-Daiyama (FUJII et al., 1971; KAMITANI and FUJII, 1972), Bontenyama (FUJII, 1967), and recent geothermal areas.

In Shobara and Goto, the altered zones have arranged from the center to the margin; diaspore-pyrophyllite zone, pyrophyllite zone, compact silicified zone, and sericite zone. The zonal arrangement of the main ore deposits in Abu district is similar to the above example. However, there are distinct differences in constituent minerals, i. e., in this district, andalusite, pyrophyllite, diaspore, and kaolinite occur in the center of the alteration, and topaz and augelite are often seen in the same part. Sericite zone occupies the most outside of the altered zone. The change in chemical composition, as a whole, is similar to that of Shobara and Goto.

A porous silicified rock is considered to have formed through a reaction between strong acid solution and host rock. Therefore, the formation of this rock is confined around the surface. Ugusu silica deposit, which is one of the types, was studied in detail by IwAO (1962, 63). The alteration zoning pattern in this deposit is porous silicified zone, alunite zone, montmorillonite zone from the center to the margin.

Generally, rising hydrothermal solution changes sulfate acid rapidly as it aproachs the surface due to oxidation from  $H_2S$  and/or  $SO_2$ . It was pointed out by IwASAKI (1970) that the major part of the volcanic hot springs in Japan are extremely abundant with  $SO_4$  anion. Near the surface, the components except  $SiO_2$  are mostly leached out from the host rock by strongly acid solution. In an extreme case, even  $Al_2O_3$  can be migrated, which is one of the most resistant components. Chemical potential of  $SiO_2$  is generally low under low-temperature, so that addition of  $SiO_2$  from the solution occurs little. Porous silicified rock originated from a residual enrichment will be formed around the surface. In the circumference of the silicified rock, pyrophyllite, kaolinite, diaspore, and alunite are formed in corresponding to each physical-chemical condition. Aluminosilicate minerals such as sericite, chlorite, and montmorillonite containning in some quantities of  $K_2O$ ,  $Na_2O$ , and MgO are stable phases in the most exterior portion of the altered zone. If the original rock is andesitic in composition, chlorite and montmorillonite will be stable phases. While, sericite is more predominant where the original rock is rhyolitic in composition.

In the altered zone of the Roseki ore deposit ocurring close to or within an intrusive body, porous silicified rock cannot be formed. For instances, in Goto and Shinyo mines, the zonal arrangement shows pyrophyllite-diaspore, pyrophyllite, and sericite zones from the center to the margin. The central part of the altered zone is characterized by remarkable increase of  $Al_2O_3$  and  $H_2O$ . Recently, pyrophyllite-diaspore zone was confirmed in the center of the alteration in Ōtake geothermal area by numerous drillings at several 100 meters bellow the surface (HAYASHI, 1973).

# 10. 3 Constituent Minerals of Hydrothermally Altered Rocks

Altered minerals from main Roseki ore deposits and geothermal areas are shown in Table 18. Main constituent minerals are pyrophyllite, kaolinite, sericite, and quartz. Pyrite and hematite occur commonly. Andalusite, topaz, dumortierite, and zunyite are very rare. Andalusite is found a little as accessory mineral in Yakuno, Hörö (YAMAMOTO, 1965), Myöközan, Kumano (KINOSAKI, 1963a), and Goto mines (IWAO *et al.*, 1943; HAMACHI, 1943). Recently, it was also found in the Shōwa-Shōkōzan (KAMITANI, 1974c). SUMI (1968) reported andalusite accompanied with pyrophyllite in Matsukawa geothermal area. But it occurs as ac-

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AREA MINERAL	MITSU- ISHI	SHOBA- RA	ABU	GOTO	ικυνο	UGUSU	OTAKE	MATSU- KAWA
ANDALUSITE			0	×				×
CORUNDUM	X	$\triangle$	$\triangle$	$\triangle$				
PYROPHYLLITE	0	Ø	Ø	Ø	Ø	Δ	Ø	$\triangle$
DIASPORE		0	Δ	0			$\triangle$	X
BOEHMITE	$\triangle$	Δ			×			
KAOLINITE	0	0	0	0	0	Ø	0	0
DICKITE		Δ	X		×		X	
MUSCOVITE (SERICITE)	0	Δ	0	1	Δ		Δ	
ALUNITE	×	Δ				0	0	0
DUMORTIERITE			0	0				
TOPAZ			0					
ZUNYITE				0				0
APATITE			0					
🔘 Abunda	nt (	) Comm	on 🛆	Poor	×Rare		Accesso	v minera

Table 18 Mineral assemblages of Roseki ore and hydrothermally altered rocks in Japan.

cessory mineral.

In Abu district, andalusite is one of the main constituent minerals in all andalusite zones. Topaz, augelite, dumortierite, and wilkeite, most of which are considered generally "pneumatolytic minerals", often concentrate in the central part of some andalusite zones. They also coexist closely with andalusite, quartz, diaspore, and muscovite. In Kanakura mine (IwAo and ŌSHIMA, 1951), Yonago mine (ŌTA and KATADA, 1955), and the others, Nagano Prefecture, tourmaline, zunyite, and apatite have been found. The occurrences suggest existence of a high-temperature stage in advance of the formations of pyrophyllite, diaspore, and kaolinite. From the study on the thermal history of Matsukawa area, SUMI (1972) concluded that pyrophyllite zone containing a small amount of diaspore, zunyite, andalusite was formed throughout a high-temperature stage of the geothermal activity.

#### 11. Summary

The major part of Abu district is occupied by the Fukuga formation, which is mainly composed of the Kanai andesite member, Kiyodani member, and Mitsugadake rhyolite member. It has about 1,800 meters in maximum thickness. The lacustrine sediment of the Kiyodani member accumulated in small-scale of lakes during a break of the volcanism of intermediate to acidic in compositions. It consists mainly of the alternation of tuffaceous sandstone, siltstone, shale, and rhyolitic tuffs. After the sedimentation, a large scale of volcanism took place, involving eruption of lava, pyroclastic fall and flow of rhyolitic in composition. The maximum thickness of this member is approximately 1,100 meters. Among them, the rhyolite welded tuff bed is the thickest, being more than 600 meters. It has several more compound cooling units and the central part of each shows commonly a distinct eutaxitic texture.

Immediately after the accumulation of the Fukuga formation, the biotite adamellite intruded into these rocks. This adamellite is stock in shape extending approximately northeastsouthwest direction. A small scale of pegmatite and muscovite-bearing phase is dominant

in the southern margin of this stock. Accordingly, it is thought that the adamellite consolidated under the condition of high-vapor pressure. Based on the stratigraphic sequence and its structure of the Fukuga formation, it is assumed that the stock intruded up to about 2,000 meters below the surface.

Andalusite-bearing Roseki ore deposits occur in the intensely altered zones. The hydrothermally altered zones consisting mainly of andalusite zone, pyrophyllite zone, and sericite zone are mostly distributed in the periphery of the adamellite stock. The zonal distribution can be recognized such as andalusite zone, pyrophyllite zone, and sericite zone from the adamellite stock side to outside. The distribution and the occurrence of the andalusite zones suggest that the hydrothermal alteration is closely related to intrusion of the biotite adamellite. This assumption has been supported by the K-Ar age determinations of muscovites from the adamellite and the andalusite-bearing Roseki ore deposit at Uku mine.

The Kiyodani member is conformably covered by the Mitsugadake rhyolite member. The upper bed of the Kiyodani member has a greater permeability than those of the other members because it is a lacustrine sediment. This physical character may be suitable for circulation and infiltration of meteoric water and heat convection. WHITE *et al.* (1971) explained that a vapor dominated meteoric hydrothermal system would circulate down to several kilometers under surface. In Ōtake geothermal area, clastic sediments of the Tertiary age lying about 1 kilometer under the present surface have been considered predominant reservoir of hydorthermal liquid (HAYASHI, 1973). Almost all of the solutions related to hydrothermal alteration were considered to be meteoric in origin according to studies of oxygen isotopic ratio  $(O^{18}/O^{16})$  and hydrogen isotopic ratio (D/H) (e. g. TAYLOR, 1967).

Intrusion of the biotite adamellite brought rise of temperature of the Fukuga formation in a regional extent. Circulating water in the lacustrine bed should have been changed to hightemperature hydrothermal solution by a conductive heat-transfer from the adamellite. On the other hand, high-temperature fluid separated from the adamellite magma ascended directly along fractures. Some volatiles (H<sub>2</sub>O, H<sub>2</sub>S, S, SO<sub>2</sub>, F, P, B, etc.) have been fixed as topaz, augelite, dumortierite, wilkeite, and pyrite with andalusite, muscovite, and so forth, thus formed "greisen" zone at the center of some andalusite zones. The West ore deposit of Hinomaru-Nako mine is one of the examples. Hydrothermal solution responsible for pyrophyllite zones has been generally thought to be acid to weak acid conditions. Sulfate acid solution gives rise to easily through oxidation of H<sub>2</sub>S and/or SO<sub>2</sub>-bearing solution. The acid hydrothermal solution percolated the lacustrine bed (Figure 26). Consequently, the hydrothermally altered zone of the stratiform type was formed. Part of the acidic hydrothermal solution reached to the Mitsugadake rhyolite member, or even flowed out the surface. Porous silicified zone and alunite zone may have formed around the surface by reaction between the host rock and the strong acid solution. The silicified zone is not seen at present in this district. It is, however, suggested that the alunite zone at the South pit of Gagaradani ore deposit may possibly be the lower extension of the porous silicified zone.

The andalusite zone was formed by subtraction of MgO, CaO, Na<sub>2</sub>O, and K<sub>2</sub>O and addition of  $Al_2O_3$  and  $H_2O$  through the reaction between the acid solution and the host rock under high-temperature. Generally,  $Al_2O_3$  is hardly mobile with weak alkaline to weak acid solution, but it becomes soluble in proportion to increase of hydrogen ion. Parts of alkali metals and alkaline earth metals, which had been perfectly leached out from the original rock of the andalusite zone, were fixed into the sericite zone. Among these metals, MgO and CaO



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Fig. 26 Schematic drawing of relationship between stratigraphis sequence and hydrothermal alteration.

were transfered further into the peripheral parts.

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Andalusite, corundum, pyrophyllite, diaspore, and quartz were formed in the early stage of the hydrothermal action. Soon after, the hydrothermal action began to decline. Pyrophyllite, diaspore, and kaolinite of the middle stage, therefore, were formed within part of the andalusite zone in rather small scale compared with the andalusite zone. In the same stage of Kuwano-Ohira mine, on the other hand, muscovite became more stable phase by increase of K/H ratio in the solution. In the late stage, kaolinite, muscovite, and mixed layered minerals were locally formed as small scale veins. The hydrothermal action of this stage declined much as compared with that of the middle stage.

Based on stratigraphy and geologic structure, changes of the chemical composition, and isotopic dating, the Fukuga formation and the biotite adamellite are considered as volcanoplutonic association. The adamellite is an ultimate source for the hydrothermal alteration. The hydrothermal solution changing temperature from high to low retrogressively reacted mainly with the lacustrine sediments of the Kiyodani member. For the formation of large-scale Roseki ore deposit, it is necessary to have highly permeable stratum such as the lacustrine sediment close to heat-source.

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# 山口県阿武地区含紅柱石ろう石鉱床の成因

# 神谷雅晴

わが国には多数のろう石鉱床が知られているが,そのほとんどは中生代白亜紀の酸性火山岩層および新第 三紀の中性-酸性火山岩層を母岩として生成する. これらはいずれもパイロフィライトを主とし,若干のカ オリン鉱物およびセリサイトを伴う.

阿武地区のろう石鉱床の多くは紅柱石・パイロフィライト・カオリナイトを主成分鉱物とし、白雲母・コ ランダム・ダイアスポアなどを随伴する.ろう石鉱床の母岩である中生代白亜紀の阿武層群福賀累層は濃飛 ・相生・高田流紋岩類とほぼ同時期の活動になると考えられている.

福賀累層は下位より上位に向かって金井安山岩層・木与谷層・三ケ岳流紋岩層と累重しており、その最大 層厚は約1,800mである.金井安山岩層は輝石安山岩・同凝灰岩を主とする.木与谷層は、下部で流紋岩質 凝灰岩が優勢であるが、上部では凝灰質砂岩・シルト岩・頁岩を主とし凝灰質礫岩および流紋岩質凝灰岩を 挾有する.上部層は層厚約200mの陸水堆積層である.三ケ岳流紋岩層は流紋岩溶岩・凝灰岩・溶結凝灰岩 からなる.なかでも溶結凝灰岩層は厚さ50m以上の数枚の Cooling unit からなり、最大層厚 600m+と考 えられる.

福賀累層は関門層群および周南層群を基盤として堆積し,全体として東に開いた半盆状構造を示す. 黒雲 母アダメロ岩は火山活動の末期あるいはその直後に福賀累層の北翼部の構造におおむね調和して貫入・固結 した.

含紅柱石ろう石鉱床の形成に関与した熱水変質作用によって、紅柱石帯・パイロフィライト帯・セリサイ

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ト帯が生じているが,それらは黒雲母アダメロ岩体の南縁部から外側に向かって紅柱石帯・パイロフィライ ト帯・セリサイト帯という帯状分布を示している.同時に,それらの熱水変質帯の規模もアダメロ岩体から 離れるにつれて小さくなる.

熱水変質作用は福賀累層木与谷層において強く,また広範囲に亘っている.とくに,木与谷層上部層(陸 水堆積層)中においてきわめて顕著である.したがって大半のろう石鉱床が本層中に形成されている.

紅柱石帯の形成深度は火山層序・構造および黒雲母アダメロ岩との関係から地表下 800-1,200m と推定された. 紅柱石帯はアダメロ岩体に近接した木与谷層中に生じており,アダメロ岩体から 700m 以上距った場合にはわずかにセリサイト化を蒙っているにすぎない.

宇久中央・日ノ丸奈古ガガラ谷および共立奈古の各鉱床では、熱水変質作用の初期に紅柱石-コランダム. 紅柱石-ダイアスポア、紅柱石-パイロフィライト、紅柱石-石英という鉱物組合せが安定である. 変質作用 の中期を代表する鉱物組合せはパイロフィライト-ダイアスポア、パイロフィライト-カオリナイト-石英、 パイロフィライト-石英である. その後期を特徴づけるものはカオリナイト-白雲母 あるいは カオリナイト-白雲母-混合層鉱物である. 熱水変質作用の初期において紅柱帯が生じ、中期では比較的限定された範囲に おいてのみ変質作用を認める. 後期はさらに劣勢となり、細脈状あるいは網状の変質を示す.

これに対して,桑野大平・鳥ノ巣・木与谷など黒雲母アダメロ岩体にきわめて近接して形成された各鉱床では,熱水変質作用全般を通じて白雲母が安定である.しかし,パイロフィライトはほとんど生成しない. これは熱水溶液中の K/H 比の相違を反映したものと考えられる.

日ノ丸奈古西部鉱床では紅柱石・白雲母・石英を主とする紅柱石帯のほぼ中心部にトパズ-アウゲライト-紅柱石-白雲母-黄鉄鉱-石英からなるグライゼン帯が確認された.この産状と,熱水変質帯の配列,火成岩 とろう石鉱床のK-Ar 年代,産出鉱物および鉱物組合せなどから,阿武地区紅柱石ろう石鉱床の成因について以下のように結論した.

阿武層群福賀累層と黒雲母アダメロ岩とは一つの Volcano-plutonic complex と考えられる. 黒雲母アダ メロ岩の貫入によって福賀累層の温度は全般的に上昇した. 木与谷層上部層はその物理性から, 地表水の浸 透・循環が容易であって, アダメロ岩の貫入により熱水溶液を生じたであろう. 同時に, アダメロ岩から分 離した揮発性成分に富む高温流体は母岩の割目に沿って上昇し, グライゼン帯を形成した. 熱水変質作用は その初期において高温かつ優勢であったが, 中期から後期にかけては温度の下降とともに急激に衰微した. 比較的大規模な層状のろう石鉱床の形成には, 陸水堆積層の存在と熱源となるべき酸性マグマの貫入が重要 な役割を果している.

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- 1 Open pit of Central ore deposit of the Uku mine.
- 2 No. 1 open pit the Kyoritsu-Nako mine.A: Andalusite zone, S: Sericite zone, F. Fault
- 3 Flow structure of the rhyolite lava in the Mitsugadake rhyolite member at Kabutozaka.





e c



- 1 Rhyolite welded tuff of the Mitsugadake rhyolite member at Yamanokuchi river.
- 2 Hydrothermally altered rock derived from the lacustrine sediment (tuffaceous sandstone and siltstone) of the Kiyodani member at the Gagaradani ore deposit.
- 3 Altered taffaceous sandstone of the Kiyodani member.
  - Q: Quarts, P: Pyrophyllite (Crossed nicols)

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Plate



- 1 Weakly sericitized tuffaceous siltstone of the Kiyodani member (Crossed nicols).
- 2 Rhyolite welded tuff with well-developed welded texture of the Mitsugadake rhyolite member at Yamanokuchi river.
   Kf: Potassium feldspar, P: Plagioclase, Q: Quartz (Single nicol)
- 3 Biotite adamellite.

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B: Biotite, P: Plagioclase, Kf: Potassium feldspar, Q: Quartz (Single nicol)



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0.5 m m

0.5 m m



1 Paragenesis of andalusite and corundum in the andalusite-pyrophyllitekaolinite subzone of the Uku mine (Single nicol)

3

- 2 Roseki ore from the Miyazaki-Köchi mine. Diaspore is remarkably replaced by pyrophyllite.
  - D: Diaspore, P: Pyrophyllite, Q: Quartz (Crossed nicols)
- 3 Euhedral and alusite in the and alusite zone belonging to Type B at the Hinomaru-Nako mine.
  - A: Andalusite, M: Muscovite (Crossed nicols)

Plate 5





1 Coarse grained andalusite from the Uku mine. Andalusite is replaced by kaolinite and pyrophyllite.

K: Kaolinite, P: Pyrophyllite, A: Andalusite (Crossed nicols)

- 2 General appearance of Roseki ore of quartz-andalusite-pyrophyllite subzone.
  - A: Andalusite, P: Pyrophyllite, Q: Quartz (Single nicol)
- 3 General appearance of Roseki ore of quartz-andalusite subzone. A: Andalusite, Q: Quartz (Single nicol)



Plate 6





 Topaz in the greisen zone of the West ore deposit of the Hinomaru-Nako mine.
 T: Topaz, P: Pyrite, Q: Quartz (Single nicol)

- 2 Augelite in the greizen zone of the West ore deposit.
  - Augelite, Q: Quartz (Crossed nicols)
- 3 Radiating dumortierite in the andalusite zone belonging to Type B.D: Dumortierite, A: Andalusite (Single nicol).

Plate 7



Wilkeite from the No. 1 ore deposit of the Kyöritsu-Nako mine.
 W: Wilkeite, P: Pyrophyllite (Single nicol)



2 Coarse-grained muscovite from the Central ore deposit of the Uku mine.