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Hydrothermal alteration at Tobaru, Kagoshima prefecture, Kyushu, Japan

Christopher F. Hamilton
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Kyushu, Japan**

Hamilton, Christopher Freeland, M.S.

San Jose State University, 1992

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HYDROTHERMAL ALTERATION AT
TOBARU,
KAGOSHIMA PREFECTURE, KYUSHU, JAPAN

A Thesis

Presented to the
Department of Geology
San Jose State University

In Partial Fulfillment
of the Requirements of the Degree
Master of Science

by
Christopher F. Hamilton
December, 1992

APPROVED FOR THE DEPARTMENT OF GEOLOGY

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ABSTRACT

HYDROTHERMAL ALTERATION AT TOBARU, KAGOSHIMA PREFECTURE, KYUSHU, JAPAN

by Christopher F. Hamilton

In the Tobaru study area, middle Miocene andesitic flows are overlain by a Pleistocene rhyolite. Hydrothermal alteration is evident on the surface by the presence of clay, pyrite, and quartz veins. A zone of silicification, including pockets of advanced argillic alteration, is enveloped by a zone of argillic alteration, which grades into propylitized rock.

X-ray diffraction data indicate the presence of pyrophyllite, tourmaline, and quartz in silicified rocks, illite and smectite in argillized rocks, chlorite in propylitized rocks, and gypsum within fractures. The presence of these minerals may suggest overprinting of an acidic high-temperature hydrothermal system upon an older low-temperature system. Based upon a review of localities with geology and alteration mineralogy similar to that of Tobaru, a magmatic body at depth is inferred to be the source of heat and gases that produced acidic fluids in the hydrothermal system.

Hydrothermal alteration of host rocks may have taken place during the Pleistocene. Propylitic alteration probably took place first and was later overprinted by silicic, argillic, and advanced argillic alteration in some areas. Low-temperature minerals (gypsum and sulfur) were likely deposited last. Today, there is no hydrothermal activity at the surface.

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DEDICATION

This work is dedicated to my parents, Peter and Laurel, and sister, Aleina, who have given me guidance and support throughout my academic career.

TABLE OF CONTENTS

	Page
ABSTRACT	iii
INTRODUCTION	1
Background of Study	1
Purpose of Study	5
Methodology	5
Previous Work	5
GEOLOGY OF SOUTHERN KYUSHU	7
GEOLOGY OF TOBARU	14
Lithology	16
Hokusatsu Andesite	16
Original Rock	16
Altered Rock	20
Yamano Rhyolite	25
Original Rock	25
Altered Rock	28
Discussion of Petrography	28
X-ray Diffraction by Author	36
Sample Preparation	36
Pyrophyllite Method	37
Heavy Liquid Procedure	37
Vein Material Method	38
Obtaining and Interpreting Patterns	38

Distribution of Altered Rocks	46
INTERPRETATIONS	48
General Model of a Hydrothermal System	48
Epithermal Ore Deposits	50
Paradise Peak, Nevada, USA	50
Negros, Philippines	50
Geologic History of Tobaru	52
SUMMARY	56
REFERENCES CITED	57
APPENDIX	61

LIST OF ILLUSTRATIONS

Figure	Page
1. Location Map of Tobaru and Ore Deposits of Southern Kyushu	2
2. General Geology of Southern Kyushu	8
3. General Cross Section of Southern Kyushu	9
4. Generalized Geologic Column of Southern Kyushu	10
5. Ore Deposits, Plate Tectonics, and Volcanic Activity in Kyushu	11
6. General Lithology of Fuke-Tobaru Area	15
7. Grid of Study Area Designated by Mitsui Mining and Smelting Company	17
8. A Nearly Fresh Sample of Hokusatsu Andesite	18
9. Photomicrograph of Incipient Alteration in Porphyritic Hokusatsu Andesite	19
10a. Photomicrograph of Altered Orthopyroxene Phenocryst (Plain Light)	21
10b. Photomicrograph of Altered Orthopyroxene Phenocryst (Crossed Nichols)	22
11. Photomicrograph of Argillized Porphyritic Hokusatsu Andesite	23
12. Photomicrograph of Relict Plagioclase Replaced by Quartz	24
13. Aggregate of Large Grains of Calcite, Quartz, and Pyrite in Hokusatsu Andesite	26
14. Photomicrograph of Argillized Yamano Rhyolite	27
15. Highly Altered Yamano Rhyolite	29

16. Quartz Vugs in Silicified Yamano Rhyolite	30
17. Gypsum Found in Drill Core	31
18. Drill Core Sample Containing Native Sulfur and Gypsum	32
19a. Photomicrograph of Silicified Yamano Rhyolite (Crossed Nichols)	33
19b. Photomicrograph of Silicified Yamano Rhyolite (Plain Light)	34
20. Comparison of X-ray Patterns from Samples Containing Pyrophyllite	42
21. Comparison of X-ray Patterns from Samples Containing Tourmaline	43
22. Comparison of X-ray Pattern from Topaz Standard and a Pattern from a Sample Originally Thought to Contain Topaz	44
23. Comparison of X-ray Pattern from Gypsum Standard and Two Patterns from Samples Containing Gypsum	45
24. General Alteration Map of Tobaru Study Area	47
25. Map of Pacific Rim Showing Locations of Areas Discussed in Text	49
26. A Possible Vertical Relationship Between Advanced Argillic Alteration (High Temperature Minerals) and Intrusive Activity	51
27. Distribution of Alteration Zones at Negros, Philippines	53
28. Hypothetical Geologic History of the Tobaru Area	55

LIST OF TABLES

Table	Page
1. Hydrothermal Alteration Terminology and Characteristic Minerals	4
2. Gold and Silver Production from Deposits in the Hokusatsu and Nansatsu Mining Districts	13
3. Mineralogy Observed in Hand Sample, Thin Section, and X-ray Diffraction	35
4. Results of X-ray Diffraction by C. Hamilton	40

INTRODUCTION

Ore deposits in Japan are of three main types. Deposits of the Kuroko-type, common in northeastern Japan, result from hydrothermal activity in a marine setting (Barnes, 1979). Skarn deposits, created during the contact of magmatic intrusions with carbonate rock (Park and MacDiarmid, 1975; Barnes, 1979), exist in central Honshu; the Kamioka zinc mine west of Nigata is an example. To the south, in Kyushu, are hydrothermal or hot spring gold deposits (Izawa and Urashima, 1989). These deposits are the result of low-temperature (100-260° C) hydrothermal activity in a near-surface environment and are Quaternary in age (Izawa and Urashima, 1989).

The mining areas of southern Kyushu are grouped into two districts: Hokusatsu, meaning "Northern Satsuma" and Nansatsu, meaning "Southern Satsuma." Southern Kyushu was formerly known as the Satsuma Province. This thesis focuses on Tobaru, a small area in the Hokusatsu district near the former gold mine of Fuke (Fig. 1).

Background of Study

From August 1990 to January 1991, the author participated in an internship with BHP-Utah Japan Inc., a mineral exploration firm located in Kagoshima City, Kyushu, Japan. This was the author's first experience working for an exploration company. Duties included rock and soil sample collection and mapping in areas selected to have potential for precious metal deposits. Tobaru was one of these areas.

When investigating a potential area, a common procedure is to define the alteration mineral assemblage present (White, 1990). These minerals

Figure 1. Location map of Tobaru and ore deposits of southern Kyushu. Fuke, Okuchi, Hishikari, and Kagoshima City are referred to throughout text.

reflect temperature and chemistry of the original hydrothermal fluids (Barnes, 1979; White *et al.*, 1990). It is also common practice to classify mineral assemblages into *types* of alteration. (Alteration terminology and associated minerals are listed in Table 1.) Types of alteration and, more specifically, mineral assemblages are useful guides in locating environments favorable for precious metal mineralization (White, 1990). Thus, in order to understand the evolution of a hydrothermal system, an accurate account of the alteration minerals present is essential.

A subject of concern at Tobaru is the incomplete and poorly documented information on the alteration mineralogy which, if known, could be significant. Results of an X-ray diffraction analysis performed by the Mitsui Mining and Smelting Company, referred to hereafter as Mitsui, indicated the presence of pyrophyllite and tourmaline on the surface and notations in a log for a core hole drilled in 1981 by Sumitomo Metal and Mining suggested the presence of pyrophyllite and topaz at similar depth. The presence of tourmaline and topaz had not been documented previously in the Hokusatsu District.

Pyrophyllite, tourmaline, and topaz are products of acidic, high-temperature (300-400° C) hydrothermal fluids (Deer *et al.*, 1962; Park and MacDiarmid, 1975; Barnes, 1979; Guilbert and Park, 1986; White, 1990) and are included in the advanced argillic alteration mineral assemblage (Table 1). If advanced argillic alteration exists at Tobaru, it is a new occurrence in a Quaternary, near-surface environment in Kyushu (L. James, written communication, 1991). The hydrothermal system of Tobaru may not match the accepted low-temperature scenario for the region.

Table 1. Hydrothermal alteration terminology and characteristic minerals
(compiled from Park and MacDiarmid, 1975; Barnes, 1979; Jensen and Bateman, 1981; and White, 1990)

Alteration	Characteristic minerals	Notes
Silicic	Quartz veins and vugs	Also called "silicification"
Argillic	Kaolinite, smectite-group (montmorillonite); micas typically not associated	Typically zoned with kaolinite close to veins and montmorillonite farther from veins
Advanced argillic	Pyrophyllite, kaolinite, alunite, dickite, sericite, pyrite, quartz, tourmaline, topaz, zunyite	Minerals commonly in veins; alteration distribution typically irregular
Propylitic	Chlorite, epidote, calcite; minor pyrite, iron oxides, and sericite may be associated	Typically a widespread or regional alteration

Purpose of Study

The purpose of this thesis is to document the alteration minerals present and evaluate the history of hydrothermal activity at Tobaru. The data concerning the presence of advanced argillic alteration, characterized by pyrophyllite, tourmaline, and topaz, are of particular concern.

Methodology

Field work included 14 days of surveying and mapping in the Tobaru area. Soil and rock sample collection was performed by the author and a partner from Mitsui in a study area designated by Mitsui, although the surrounding area including Fuke was also considered. An additional eight to ten days were spent collecting core samples from the Mitsui core yard. Rocks were studied through hand specimen examination, petrographic analysis, and examination of current data on X-ray diffraction of bulk rock samples (performed by Mitsui). An additional X-ray diffraction examination was performed by the author to test for the presence of tourmaline, topaz and pyrophyllite. The samples indicated to contain these minerals by previous workers were used in this examination.

Previous Work

Much unpublished (and inaccessible) data exist with numerous mineral exploration firms that have surveyed the Tobaru area. The total number of surveys that have been done in the area is unknown. A complete, unpublished drill core log from the previously mentioned core hole, which

includes rock types and preliminary alteration mineralogy, was made available to the author.

There is very little published literature on the Hokusatsu district and none specifically on the geology of Tobaru. One article exists concerning Fuke (Fig. 1) in which clay minerals at ore deposit sites were studied and ages of rock units and episodes of alteration were interpreted (Inome *et al.*, 1981). Also, pyrophyllite was reported in an area near Fuke, but no documentation was published on precisely how and where this discovery was made (Izawa *et al.*, 1990).

GEOLOGY OF SOUTHERN KYUSHU

The geology of southern Kyushu consists of upper Miocene to Holocene felsic and andesitic flows over a Cretaceous sedimentary basement (Figs. 2 and 3). A general stratigraphic column of southern Kyushu is illustrated in Figure 4. Deep drilling in the central area of Kyushu indicates that the extrusives are nearly 2 km thick (Kamata, 1985). Lower Miocene felsic intrusives also are present within the basement. All units are fractured, the typical orientation being nearly vertical. Most likely, the fractures served as a "plumbing system" for hydrothermal fluids.

Alteration and ore deposition in this setting are the result of chemical reactions between hydrothermal fluids and adjacent wall rocks (Hayba *et al.*, 1985). Throughout southern Kyushu, various forms of hydrothermal alteration are evident, and ore deposits are hosted in all rock types, most commonly in the andesitic units. Alteration tends to be limited horizontally, but is extensive vertically. Three episodes of ore mineralization are inferred: middle Miocene, early Pliocene, and early Pleistocene (Izawa and Urashima, 1983).

In terms of plate tectonics, Japan is an active island arc (Uyeda, 1971) with frequent earthquakes and volcanic events occurring both on and offshore. Deep trenches to the east of the islands are also products of subduction. The Nankai Trough, for example, is a trench resulting from the subduction of the Philippine Sea Plate beneath the Eurasian Plate. The Trough is situated 200 km to the east of Kyushu and is parallel to the coast (Fig. 5).

Figure 2. General geology of southern Kyushu (after Izawa and Urashima, 1983, and Yamazaki *et al.*, 1984). Line A-A' refers to cross section shown in Figure 3.

Figure 3. General cross section of southern Kyushu (after Izawa and Urashima, 1983; see Figure 2 for location of A-A').

Figure 4. Generalized geologic column of southern Kyushu (after Izawa and Urashima, 1983).

Figure 5. Ore deposits, plate tectonics, and volcanic activity in Kyushu (after Uyeda, 1971; Izawa and Urashima, 1983; and unpublished data by L. James, 1990; • refers to ore deposit location, triangle refers to site of active volcano).

Kyushu is probably the most volcanically active island of the arc. Sakurajima volcano, located to the east of Kagoshima City, across Kagoshima Bay, is active. Volumes of ash are expelled into the atmosphere and carried by air currents to Kagoshima City and several tens of kilometers beyond. Typically, from 0.5 cm to as much as 3 cm is deposited. These eruptions now occur almost monthly, but in the recent past, eruptions only took place once every six to ten months (M. Taneda, personal communication, 1990). Hydrothermal and volcanic activity is evident 3 km to the north of Sakurajima at Mt. Kirishima, a volcano surrounded by several hot springs. A final example of volcanic activity in Kyushu is the recent eruption of Mt. Unzen near Nagasaki (located in northwest Kyushu, Fig. 5). Steam vent activity in close proximity to Unzen preceded the eruption.

In Kyushu, ore deposits occur in volcanic areas that are the result of subduction (Izawa and Urashima, 1989). The ore deposits occur in a belt adjacent and parallel to a belt including most of the active volcanoes (Fig. 5). Additionally, these features are parallel to the previously mentioned Nankai trough. In Kyushu, subduction probably plays a role in the creation of ore deposits because volcanic activity is a product of subduction and hydrothermal alteration and ore deposition are products of volcanic activity. Information on some of the major gold and silver deposits of southern Kyushu is listed in Table 2.

Table 2. Gold and silver production from deposits in the Hokusatsu and Nansatsu mining districts (based on Izawa and Urashima, 1983, and unpublished data from Mitsui Mining and Smelting Company, 1985)

[Au, gold; Ag, silver; Sil, massive silicified rock; qz, quartz; chl, chlorite; cal, calcite; ad, adularia; kaol, kaolinite; zeo, zeolite; ser, sericite; hall, halloysite; chal qz; chalcadonic quartz; mont, montmorillonite; --, no data available]

Deposit	Production dates	Production (metric tons)			Type	Gangue minerals	Host rock(s)
		Au	Ag	Ag/Au			
Hokusatsu district							
Fuke	1896-1914, 1935-1947	2.4	1.5	0.6	Vein	qz, chl, cal	Andesite
Okuchi	1896-1967, 1976-1980?	17.7	12.0	.7	Vein	qz, cal, clay	Andesite, rhyolite
Hishikari	1983-	11.2	8.7	.8	Vein	qz, ad, kaol, cal, chl	Basement, andesite
Onoyama	1935-1962	1.8	.8	.4	Vein	qz, zeo, cal, ad, chl, clay	Dacite, tuff
Yamagano	1639-1953	28.4	28.3	.7	Vein	qz, cal, ad, ser, zeo, hall	Andesite/shale
Ora	1895-	1.5	8.0	5.3	Vein	chal qz, ad, cal	Andesite
Kushikino	1888-1908, 1960-	55.2	456.2	8.3	Vein	qz, cal, kaol, mont, ser	Andesite
Nansatsu district							
Kuronita	--	--	--	--	Vein	--	Andesite, tuff, rhyolite
Hanakago	--	--	--	8.5	Vein	--	Andesite, tuff
Iiyama	--	--	--	--	Sil	--	Andesite, tuff
Kago	--	--	--	--	Vein	--	Basement, andesite
Akeshi	--	3.7	4.7	3.0	Sil	--	Andesite, tuff
Kasuga	--	5.0	6.7	4.0	Sil	--	Tuff breccia
Iwato	1950-	4.4	6.6	12.7	Sil	--	Andesite, tuff breccia

GEOLOGY OF TOBARU

The study area is about 1 km² and is located between 580 and 740 m above sea level in a mountainous region 10 km to the north of Okuchi City, Kagoshima Prefecture, Kyushu, Japan (Fig. 1). Because much of the area contains logging roads, access is not difficult. A road continues from Okuchi through Fuke to the study area. The road is paved through Fuke but is gravel to Tobaru, which presents hazardous driving conditions during heavy rains. Heavy vegetation cover, the result of the tropical climate of southern Japan, presents difficulties in traversing and interpreting large-scale geologic structure in the area. However, in portions of the area containing groves of tall pine trees, the floor is nearly free of brush, making traversing much easier. Outcrops also are more abundant. Fractures are evident in all outcrops (Fig. 6).

Two types of rock are evident on the surface of the study area: andesite overlain by rhyolite (Fig. 6). The andesite is a member of the Hokusatsu Older Andesites (a unit of the "Older andesites," see Fig. 4). This unit consists of andesitic flows with ages of middle to late Miocene to Pliocene (Izawa and Urashima, 1983). Previous authors mapped the rhyolite as the Yamano Rhyolite (a unit of the "Large-scale pyroclastic deposits" in Fig. 4). It is Pleistocene in age (Izawa and Urashima, 1983; Izawa, 1990; unpublished data from Mitsui, 1985).

Almost all rocks seen on the surface in the study area are hydrothermally altered to some degree. Quartz, gypsum, clay (most notably seen in former feldspar sites), and pyrite commonly are observed in surface and drill core hand samples. A zone of silicification, characterized by quartz veins, is situated in the central portion of the study area and is surrounded by a zone of

Figure 6. General lithology of Fuke-Tobaru area (compiled from unpublished data from Mitsui Mining and Smelting Company, and field work performed by author and R. Otsubo, 1990).

argillic alteration, marked by clays and pyrite. Propylitic alteration, evident by the presence of chlorite and calcite, has affected all other rocks.

Lithology

In the following section, hand specimen and thin section descriptions of rock samples are presented. Surface samples are labeled according to where found on a grid designated by Mitsui (Fig. 7). Samples from drill core are labeled as "T-1 xxx". The drill hole is T-1 (Fig. 6) and the number following is the depth at which the sample was taken. For example, sample T-1 252 was taken from drill hole T-1 at a depth of 252 meters.

Hokusatsu Andesite

Original Rock. In field observation, this unit can easily be mistaken for an aphanitic basalt. Fresh surfaces are dark green to black (Fig. 8). Upon close examination, a porphyritic texture is evident with greenish phenocrysts that could be mistaken for olivine, additionally leading to a false conclusion about the rock. The true rock type is revealed through thin section analysis.

Generally, all samples are similar in texture and composition. In thin section, porphyritic texture also is apparent (Fig. 9). Of the total amount of phenocrysts, about 75-80% are plagioclase, 15-20% are pyroxene, and 5-10% are amphibole. Form is euhedral to subhedral and size ranges from 0.1 mm to 2 mm. The groundmass is composed of the same minerals as the phenocrysts.

The plagioclase composition is andesine by use of the Michel-Lévy method (Moorhouse, 1959; Kerr, 1977). Zoned grains commonly are

Figure 7. Grid of study area designated by Mitsui Mining and Smelting Company. Soil and rock (where possible) were collected at each intersection of grid lines. Sample 0-780 would be from the west corner of the study area, for example.

Figure 8. A nearly fresh sample of Hokusatsu Andesite (sample 0-60). This rock could be mistaken for basalt due to a dark color and obscure porphyritic texture.

Figure 9. Photomicrograph of porphyritic Hokusatsu Andesite (sample 100-0, crossed nicols). Area covered is approximately 1.75 cm x 1.2 cm.

subhedral and contain minor fine-grained opaques in the core area. Albite twinning is common and Carlsbad twinning is present to a lesser degree.

In all samples, pyroxene and amphibole are similar in size and are in close proximity to one another. Identification is possible by noting typical prismatic shape and cleavage traces. Sample 200-630A contains a few relatively unaltered phenocrysts of orthopyroxene (Figs. 10a and 10b).

Altered Rock. Argillic alteration and silicification are evident in the altered samples of the Hokusatsu Andesite. The original texture may be interpreted by the relict shapes of phenocrysts.

In hand specimen, the samples of argillized rock are soft and crumbly. In some samples, relict euhedral plagioclase sites range in size from 1 to 4 mm in length. These sites contain sericite, smectite, and/or kaolinite (according to the results of the Mitsui X-ray diffraction analysis). A relict porphyritic texture is evident in thin section (Fig. 11) and is further apparent through a comparison with fresh rock (Fig. 9). The opaques in Figure 11 may be pyrite.

In thin sections of silicified rock, euhedral voids (0.09-0.18 mm in length), having a shape similar to Carlsbad-twinned feldspars, are filled with quartz (Fig. 12). The vacancies are most likely relict plagioclase sites. The quartz crystals appear similar to those in a geode; growth direction was from the wall towards the center of the vacancy. Aggregates of similar-sized grains are found in the central area of some vacancies. Other vacancies are completely filled with round, inclusion-free quartz grains (0.06 mm). The

Figure 10a. Photomicrograph of partly altered orthopyroxene phenocryst (center of photo) in Hokusatsu Andesite (sample 200-630A, plane light). Area covered is approximately 8.7 mm x 6 mm.

Figure 10b. Photomicrograph of partly altered orthopyroxene phenocryst (center of photo) in Hokusatsu Andesite (sample 200-630A, crossed nicols). Area covered is approximately 8.7 mm x 6 mm.

Figure 11. Photomicrograph of argillized porphyritic Hokusatsu Andesite (sample 900-780, crossed nicols). Area covered is approximately 1.75 cm x 1.2 cm.

Figure 12. Photomicrograph of relict plagioclase phenocryst replaced by quartz (sample 700-120, crossed nicols). Area covered is approximately 1.75 cm x 1.2 cm.

groundmass is composed of fine-grained quartz, possibly some sericite, and opaques (which may be pyrite and/or anatase).

Chlorite is a common alteration mineral in propylitized rocks. It is found as very fine-grained, greenish-brown masses completely replacing pyroxene or as fibrous masses making up veins. Calcite is associated with chlorite, appearing as fine-grained (<0.05 mm) patches adjacent to veins or aggregates of quartz. Fine-grained epidote commonly is found in the patches of calcite, but in minor amounts. Calcite also exists as large (0.8-1 mm) subhedral crystals commonly displaying typical cleavage (Fig. 13), or as small patches of fine grains associated with relict pyroxene and plagioclase phenocrysts. The aggregate of coarse minerals depicted in Figure 13 may be a xenolith from the underlying sedimentary unit (the Shimanto Supergroup).

Yamano Rhyolite

Original Rock. The Yamano Rhyolite is moderately to intensely altered in the study area. A fresh sample was not obtained. However, relict textural features suggest this rock was felsic and porphyritic in texture with feldspar phenocrysts in a fine-grained, possibly glassy, matrix. These features are most apparent in thin section. The samples appear as a mosaic of alteration minerals. Large, wavy, fine-grained patches (possibly sericite) are inferred to be relict plagioclase sites because of a characteristic rectangular shape (Fig. 14). Devitrified glass is evident in some samples which suggests the groundmass was originally fine-grained and glassy.

Figure 13. Aggregate of large grains of calcite, quartz, and pyrite (right-hand portion of photo) in Hokusatsu Andesite (sample T-1 499, crossed nicols). The aggregate may be a xenolith from the Shimanto Supergroup. Area covered is approximately 1.75 cm x 1.2 cm.

Figure 14. Photomicrograph of silicified Yamano Rhyolite (sample 820-780, crossed nicols). Sericite may compose the wavy patch in the center of the photograph. The patch is inferred to be a relict feldspar site due to the characteristic rectangular shape. Area covered is approximately 8.7 mm x 6 mm.

Altered Rock. In hand specimen, the effects of silicification and argillization are evident (Figs. 15 and 16). Pyrite is evident within the clay-rich samples. Quartz vugs (1-25 cm) and wide (1.5-2 cm) gypsum veins (Fig. 17) are common in drill core samples. Native sulfur (yellow, characteristic odor) also is present in drill core samples (Fig. 18).

In thin section, the silicified samples are composed mostly of interlocking grains (85-90% of rock) of subhedral to anhedral quartz (Figs. 19a and 19b). The grains are similar in size (0.05-0.15 mm), relief, and color. Opaques (anatase and/or pyrite; 10-15% of rock) appear as aggregates of medium to coarse grains (0.10-0.25 mm) or isolated fine grains (0.02-0.05 mm). Finally, very fine, radiating, colorless to slightly reddish needles (2-5% of rock) were found in samples 100-300 and 100-500. These needles may be tourmaline or rutile.

Discussion of Petrography

A basic understanding of the rocks and alteration minerals present at Tobaru was achieved through the petrographic study. Table 3 is a summary of minerals observed in thin section and detected by the bulk sample X-ray diffraction examination by Mitsui (a listing of results and a copy of the X-ray pattern of 100-300 were released to the author).

A goal of this thesis, again, is to test for the presence of pyrophyllite, tourmaline, and topaz, all of which would be part of the advanced argillic alteration mineral assemblage. However, these minerals are not easily identifiable in thin section. The hydrothermal variety of tourmaline has the form of fine needles (Deer *et al.*, 1962; Kerr, 1977) or "black dust" (Dennis Cox,

Figure 15. Highly altered Yamano Rhyolite, mostly replaced by clay.

Figure 16. Quartz vugs in silicified Yamano Rhyolite.

Figure 17. Gypsum found in drill core.

Figure 18. Drill core sample containing native sulfur and gypsum.

Figure 19a. Photomicrograph of silicified Yamano Rhyolite (sample 600-340, crossed nicols). Area covered is 3.5 mm x 2.4 mm.

Figure 19b. Photomicrograph of silicified Yamano Rhyolite (sample 600-340, plane light). Area covered is 3.5 mm x 2.4 mm.

Table 3. Minerals observed in hand sample, thin section, and X-ray diffraction*

[Y.R.; Yamano Rhyolite; H.A., Hokusatsu Andesite; Key to list of minerals: X > Y, X is present in amounts greater than Y; X >> Y, X is present in much greater amounts than Y; X, Y, X and Y are present in about the same amount]

Sample	Rock unit	Texture	Minerals observed**
100-0	H.A.	Porphyritic	plagioclase >> smectite/sericite, hornblende > chlorite
100-300	Y.R.	Aphanitic	quartz > tourmaline > anatase
100-500	Y.R.	Aphanitic	quartz > tourmaline > rutile
200-630A	H.A.	Porphyritic	plagioclase >> chlorite, hornblende, orthopyroxene
200-630B	H.A.	Porphyritic	plagioclase >> chlorite, hornblende, orthopyroxene
400-450	Y.R.	Aphanitic	quartz >> pyrite, anatase
430-300	Y.R.	Aphanitic	quartz >> anatase
600-340	Y.R.	Aphanitic	quartz >> anatase
700-120	H.A.	Porphyritic	quartz > kaolinite > anatase
700-640	Y.R.	Aphanitic	quartz > anatase
800-400	Y.R.	Aphanitic	quartz >> anatase
800-700	H.A.	Porphyritic	quartz >> anatase > smectite
820-780	Y.R.	Aphanitic	quartz >> sericite > pyrite > anatase
900-700	H.A.	Porphyritic	quartz >> anatase, smectite/sericite
900-780	H.A.	Porphyritic	quartz, pyrite >> smectite, anatase > kaolinite

*Bulk-sample X-ray diffraction performed by Mitsui Mining and Smelting Company, 1991.

**Includes whole-rock and complete alteration mineral assemblages.

oral communication, 1991). Topaz has optical properties similar to those of quartz and feldspar and typically is associated with granular vuggy quartz (Deer *et al.*, 1962; Park and MacDiarmid, 1975; Kerr, 1977). Finally, pyrophyllite is difficult to identify unless it is in aggregate form: a radial appearance is evident with crossed nicols. Individual crystals are elongate and colorless (Kerr, 1977). Due to the elusive petrographic characteristics of the minerals in question, a more carefully focused X-ray diffraction examination, presented below, became necessary.

X-Ray Diffraction by Author

Sample Preparation

In order to detect minerals that form minor components of rocks through the use of X-ray diffraction methods, special sample preparation procedures are necessary. Roger Ashley of the U.S. Geological Survey, Menlo Park, California, described a simple method for detecting pyrophyllite and other phyllosilicates (oral communication, 1991), unofficially termed here the "pyrophyllite method". Puru Shresda of BHP-Utah Minerals International, Sunnyvale, California, suggested a preparation method for tourmaline and topaz (oral communication, 1991), the "heavy liquid procedure". Because these three minerals may occur in veins (Deer *et al.*, 1962; Gustafson and Hunt, 1975), a third preparation method was devised by the author, the "vein material method". These methods are designed to separate only certain minerals from a rock sample. The resulting X-ray patterns are not indicative of complete alteration mineral assemblages, but yield *partial* assemblages.

The samples used in this test were selected from T-1 drill core and surface samples suspected to contain pyrophyllite, tourmaline, and/or topaz.

Samples of pyrophyllite (from M. Maddock collection, San Jose State University, California), tourmaline (schorl, hydrothermal variety of tourmaline; Cornelius *et al.*, 1977), topaz (M. Maddock collection), and gypsum (M. Maddock collection) were prepared to function as standards to aid in the resulting X-ray pattern interpretation. The origin of the samples used as standards is unknown except for the topaz and gypsum. The topaz was taken from a rhyolite of central Honshu, Japan. The gypsum came from a lake deposit in Death Valley, California. To avoid contamination of unknowns, the standards were prepared last. These samples were ground to a fine powder, about the texture of talcum powder, placed on a clean glass slide with 3 to 5 drops of water and allowed to dry.

Pyrophyllite Method. This method tends to concentrate clay minerals and to enhance the detection of phyllosilicates. Each sample was ground to a powder in a mortar and pestle. Ten grams of the powder were added to 150 ml of distilled water. To disaggregate the sample, a Sorval ultrasonic blaster was inserted into the mixture and run for 15 minutes. Immediately after turning off the blaster, a portion of the suspension was removed with a dropper and placed on a clean glass slide. The sample was then allowed to dry.

Heavy Liquid Procedure. This method separates minerals with a density greater than 3.0 g/cm^3 , which includes tourmaline (density of 3.10-

3.25 g/cm³; Cornelius *et al.*, 1977) and topaz (density of 3.5-3.6 g/cm³; Cornelius *et al.*, 1977). Samples were ground to a fine powder in an electric rock grinder. Each sample was then each placed in a fluted glass flask containing the heavy liquid lithium metatungstate (density of 3.0 g/cm³, manufactured by BHP-Utah Minerals International, Sunnyvale, California). Once thoroughly mixed, the flasks were placed in a centrifuge at a setting of 610 rpm for 5 minutes. The flasks were then removed and placed over a filter apparatus (a paper filter over a vacuum beaker). After opening the valve at the base of the flask, the mixture was allowed to settle onto the filter paper. To separate the heavy liquid from the minerals, the mixture was washed with distilled water. Once the paper filter reached maximum capacity, the material was rinsed with hot water to remove any remaining heavy liquid. The filter paper was removed and placed below a heat lamp. Once dry, 3 to 5 grams of material were placed on a clean glass slide with 3 to 5 drops of distilled water and set aside to dry.

Vein Material Method. Material was removed from veins in all hand samples. This material was then ground in a mortar and pestle. Acetone was added to the mortar before grinding to prevent the escape of material and also to prohibit the generation of heat during grinding. Heat is of great concern because excess amounts will cause certain minerals to alter and, thus, affect results.

Obtaining and Interpreting Patterns

All runs were performed using a Rigaku Diffractometer, Model No.

150, with Cu-radiation and a goniometer range from 32° to 3° (the topaz standard and sample T-1 284 were re-run with a goniometer range from 50° to 3° to obtain a longer X-ray pattern containing two-theta values greater than 32°), and a scanning range of 2° per minute. Forty-one runs were performed. For interpretation of the resulting X-ray patterns of unknowns, transparent overlays were made of the patterns from the standards. All overlays were placed over unknown patterns to match peaks. A catalog of two-theta values, published by the U.S. Geological Survey (Hauff, 1975) for use with the Rigaku units, was also used.

A summary of minerals detected, or partial mineral assemblages, is listed in Table 4. Patterns obtained from all samples listed in Table 4 are contained in the Appendix. In Figures 20 through 23, comparisons are made between the patterns from standards and some samples. In order to clearly illustrate the presence of pyrophyllite, tourmaline, topaz, and gypsum, only the peaks of these minerals are labeled on the X-ray patterns.

Figure 20 illustrates that pyrophyllite exists at Tobaru. The height of X-ray peaks increases in proportion to population density and ratio of height to width increases in proportion to degree of crystallization (Hauff, 1975). Thus, the high, sharp peaks in Figure 20 show that the samples contain abundant, well crystallized pyrophyllite. Figure 21 illustrates the presence of tourmaline in the variety of schorl (using the catalog of two-theta values; Hauff, 1975). Topaz was not found in the samples collected. Perhaps elemental components to form topaz were not present in the hydrothermal fluid. A comparison between the topaz standard pattern and one sample with a similar, but not identical, pattern is illustrated in Figure 22. A match of only

Table 4. Results of X-ray diffraction¹ by C. Hamilton

[Key to list of minerals: X > Y, X is present in amounts greater than Y; X >> Y, X is present in much greater amounts than Y; X, Y, X and Y are present in about the same amount]

Method ²	Sample	Minerals detected ³
Standard	GM-1	(2 runs) tourmaline >> quartz
	A3087	(2 runs) topaz > quartz
	Pyro-1	pyrophyllite
	Gyp-1	gypsum
Pyrophyllite	T-1 245	quartz > illite > gypsum
	T-1 248-250	quartz > gypsum > illite
	T-1 249	quartz
	T-1 250-251	quartz > gypsum
	T-1 251-253	pyrophyllite, mixed-layer clay > gypsum > quartz, chlorite
	T-1 252	pyrophyllite > gypsum > quartz, chlorite, mixed-layer clay
	T-1 253	gypsum > pyrophyllite > quartz, chlorite, mixed-layer clay
	T-1 253-254	quartz > pyrophyllite, mixed-layer clay > gypsum > chlorite
	T-1 254	quartz > mixed-layer clay
	T-1 284	gypsum > quartz > chlorite
	T-1 286	gypsum > quartz > chlorite
	T-1 290	quartz > illite > gypsum
	T-1 294-294.5	gypsum >> quartz > illite, chlorite
	T-1 307-311	gypsum
	50-550	quartz
	100-740	chlorite > illite > quartz
	200- 90	pyrophyllite > chlorite, quartz, mixed-layer clay
	200-700	illite > chlorite > quartz
	300-700	quartz > illite
	400-220	quartz >> chlorite
Heavy liquid	0-600	quartz > tourmaline
	100-300	quartz > tourmaline
	100-400	quartz
	200-360	quartz >> tourmaline
	450-300	quartz
	500-160	quartz >> tourmaline

¹X-ray patterns can be found in Appendix.

²See section "X-ray Diffraction by Author" in text.

³Partial alteration mineral assemblages.

Table 4. Continued

[Key to list of minerals: X > Y, X is present in amounts greater than Y; X >> Y, X is present in much greater amounts than Y; X, Y, X and Y are present in about the same amount]

Method ²	Sample	Minerals detected ³
Vein material	T-l 251-252v	gypsum > quartz
	T-l 252v	pyrophyllite > gypsum >> quartz
	T-l 253v	gypsum > quartz, pyrophyllite
	T-l 284v	(1st run) tourmaline >> rutile
	T-l 284v	(2nd run) tourmaline > pyrite > rutile
	T-l 286v	gypsum >> chlorite > quartz
	T-l 293.5-294v	gypsum >> quartz
	T-l 294v	gypsum >> quartz
	800-400v	quartz

²See section "X-ray Diffraction by Author" in text.

³Partial alteration mineral assemblages.

Figure 20. Comparison of X-ray patterns from samples containing pyrophyllite. Only the pyrophyllite peaks are labeled for clarity.

Figure 21. Comparison of X-ray patterns from samples containing tourmaline (schorl or schorlite, black tourmaline). Only the tourmaline peaks are labeled for clarity.

Figure 22. Comparison of X-ray pattern from topaz standard and a pattern from a sample originally thought to contain topaz.

Figure 23. Comparison of X-ray pattern from gypsum standard and two patterns from samples containing gypsum.

one large peak (which actually corresponds to a tourmaline peak) is not enough evidence to conclude that topaz is contained in the sample. Finally, the gypsum standard pattern is compared to the patterns of a few samples containing gypsum in Figure 23.

Distribution of Altered Rocks

Using the alteration mineralogy confirmed through laboratory methods in combination with field observations, a general alteration map of Tobaru has been compiled (Fig. 24). A silicified zone, including pockets of advanced argillic alteration, is enveloped by a zone of argillic alteration, which is surrounded by propylitized rock. The silicified zone is characterized by quartz, the argillic zone by clays (kaolinite, smectite; as indicated by X-ray diffraction by Mitsui), the advanced argillic areas by pyrophyllite, tourmaline, pyrite, kaolinite, and mixed-layer clay, and the propylitic alteration by chlorite and calcite (based on X-ray diffraction by Mitsui and author). It must be noted that the propylitic alteration and the advanced argillic alteration would not have been apparent by using field observations alone.

At Tobaru, zoning of alteration types is controlled dominantly by steeply-dipping fractures (Fig. 24). Lithology may have a control as well on the zonation pattern. By comparing Figures 6 and 24, silicification appears commonly in the Yamano Rhyolite, whereas argillization and propylitization appear mainly in the Hokusatsu Andesite.

Figure 24. General alteration map of Tobaru study area. (Compiled from field work performed by author and R. Otsubo, 1990, hand sample and thin section analysis, unpublished data from Mitsui Mining and Smelting Company, and results of X-ray diffraction by author.)

INTERPRETATIONS

To develop a better understanding of hydrothermal alteration at Tobaru, the general model of a hydrothermal system and described localities with geology and alteration mineralogy similar to those of Tobaru are briefly examined below. Figure 25 is a location map of areas discussed.

General Model of a Hydrothermal System

A hydrothermal system is created when heat from a deep magmatic body causes convection in groundwater (Seward, 1988). Convection is controlled largely by host rock permeability, typically reflected by the density and orientation of fractures (Hayba *et al.*, 1985; Seward, 1988). Hydrothermal fluid, or heated groundwater, reacts chemically with the surrounding rocks and produces a new mineral suite, referred to as the alteration mineral assemblage (Henley, 1985).

Chemistry in a hydrothermal system is thought to be influenced by the injection of vapor from an underlying magmatic body (Berger and Henley, 1989). The evolution of propylitic alteration is not clearly understood. This may be due to the fact that most research on hydrothermal systems tends to focus on ore deposition and closely associated processes rather than on non-economic processes, such as propylitic alteration. Burbank (1960) suggested that the timing of propylitic alteration may be remotely related to stages of ore deposition and more closely related to the final stage of a volcanic episode. Propylitic alteration is likely the result of the reaction between CO₂-rich fluid and surrounding rock (Burbank, 1960).

Figure 25. Map of Pacific rim showing locations of areas discussed in text.

Advanced argillic alteration occurs in a near-surface environment and results from the condensation of H₂S-bearing vapor from underlying boiling fluids (350° to 400° C; Henley, 1985; White, 1990). An acid component forms in the hydrothermal fluid when the sulfide from the H₂S is oxidized to sulfate (White, 1990). Figure 26 illustrates the possible relationship between advanced argillic alteration and intrusive activity (Berger and Henley, 1989).

Epithermal Ore Deposits

Paradise Peak, Nevada, USA

The gold deposits of southern Kyushu have often been compared to the epithermal deposits of Nevada, particularly the Paradise Peak gold-silver-mercury deposit (Cox, 1980; Bonham, 1989; and John *et al.*, 1989). The lithology and alteration mineralogy of Paradise Peak are similar to those of Tobaru. The host rock consists of 26 to 15 Ma andesitic and felsic volcanic units (John *et al.*, 1989). Vertically-zoned silicic and argillic alteration are superimposed upon pervasive propylitic alteration. The youngest units, which directly overlie this zonation, are unaltered. The heat source is proposed to be a magmatic intrusion (Bonham, 1989; John *et al.*, 1989). From the interpretations made at Paradise Peak, it is inferred that perhaps at Tobaru propylitic alteration came first followed by silicification and argillization, and that both stages of alteration were related to a magmatic body at depth.

Negros, Philippines

Like Japan, the Philippine island arc is tectonically active, as shown by

Figure 26. A possible vertical relationship between advanced argillic alteration (high-temperature minerals) and intrusive activity (after Berger and Henley, 1989). This may be the case at Tobaru.

current volcanic and seismic activity (Celenk *et al.*, 1987). The geology of Negros, an island in the southwest Philippines, is similar to that of Tobaru. Pliocene to Pleistocene andesitic flows are overlain by Holocene sediment. Hydrothermal alteration exists only in the older units, and altered rocks are well exposed on the surface (Fig. 27). The surficial distribution of altered rocks and the minerals in the alteration assemblages are very similar to those of Tobaru. Pockets of advanced argillic alteration (characterized by pyrophyllite, kaolinite, alunite, and opal) are enclosed in silicification zones (quartz, illite), which intersect areas of propylitic alteration (chlorite, epidote) (Celenk *et al.*, 1987). Native sulfur commonly is associated with opal.

The propylitic alteration represents the earliest alteration event, which was later overprinted by silicification and advanced argillic alteration (Celenk *et al.*, 1987). Perhaps this also is the sequence of events at Tobaru. The heat source for hydrothermal activity at Negros is hypothesized to be a magmatic intrusion (Reyes, 1990; Celenk *et al.*, 1987). This additionally suggests that at Tobaru a magmatic body exists at depth.

Geologic History of Tobaru

Utilizing the interpretations of mineralogical relationships discussed above, a geologic history of Tobaru can be hypothesized. First, the Hokusatsu Andesite was deposited over the Cretaceous sedimentary basement from middle to late Miocene time (Urashima and Ikeda, 1980; Inome *et al.*, 1981; Izawa and Urashima, 1989). During a hiatus in volcanic activity, erosion took place and stream valleys developed. In the early Pleistocene, the Yamano Rhyolite was deposited. After this final episode of extrusive activity,

Figure 27. Distribution of alteration zones at Negros, Philippines (after Celenk *et al.*, 1987). As at Tobaru, advanced argillic alteration is enclosed in silicification.

propylitic alteration began (Urashima and Ikeda, 1980; Inome *et al.*, 1981). Later, acidic, high-temperature fluids invaded the rock units producing silicification, including isolated pockets of advanced argillic alteration, and argillic zones superimposed upon propylitized and fresh rock. Eventually, as temperatures of the hydrothermal fluids declined, low-temperature minerals formed (sulfur, gypsum). Hydrothermal activity is not evident at the modern surface. From the studies in Nevada and the Philippines previously discussed, the heat and vapor source of acid-forming components probably was a magmatic body at depth. This possible sequence of events is illustrated in Figure 28.

This interpretation is arguable because the exact timing of the sequence of events is not known. Were the phases of hydrothermal activity abrupt or separated by gradual transitions? The arrangement of alteration zones at Tobaru does not suggest a single, prograding hydrothermal episode. Since propylitic alteration affects rocks on a regional, widespread scale, it is possible that first a widespread, low-temperature event occurred, then a high-temperature, acidic event followed and did not spread beyond the boundaries of the propylitic alteration.

Figure 28. Hypothetical geologic history of the Tobaru area, no scale (based on field observation, X-ray diffraction results, T-1 drill core log; hydrothermal models from Park and MacDiarmid, 1975; Cox, 1980; Hayba *et al.*, 1985; Henley, 1985; and Bonham, 1989; and hydrothermal models of nearby deposits from Hedenquist *et al.*, 1988; Izawa and Cunningham, 1989; Izawa and Urashima, 1983; Izawa *et al.*, 1990)

SUMMARY

At Tobaru, andesite and rhyolite, which overlie a sedimentary basement, are hydrothermally altered. Several types of alteration are evident and some display a zonation pattern controlled by fractures. A widespread zone of propylitic alteration composed of chlorite and some calcite is present. An inner zone of silicification is characterized by quartz veins and pockets of advanced argillic alteration marked by pyrophyllite and tourmaline. The silicification zone is enveloped by a zone of argillic alteration characterized by pyrite, illite, kaolinite, and smectite.

Data collected for this thesis confirm the presence of pyrophyllite, tourmaline, quartz, gypsum, illite, and chlorite. The mineral assemblages and field relations suggest overprinting of a high-temperature, acidic hydrothermal system upon an older low-temperature system. Both systems probably date to the Pleistocene. Propylitic alteration occurred first and belongs to the older system. The high-temperature, acidic system, which occurred after the propylitic alteration, is marked by silicic, argillic, and advanced argillic alteration. The deposition of gypsum and sulfur came last and is evidence of a lowering in the temperature of the hydrothermal system. Presently, the system is not active at the surface.

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*In Japanese with abstract, tables, and figures in English

APPENDIX

Standards

Minerals detected by Pyrophyllite Method

Minerals detected by Heavy Liquid Procedure

Minerals detected by Vein Material Method