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MANIFESTATION OF HIGH TEMPERATURE HYPOGENE ACID ALTERATION IN STEAMING GROUND AT THE HATCHOBARU GEOTHERMAL FIELD, KYUSHU, JAPAN

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SUMMARY- Steaming ground called Komatsu jigoku is located in the northern part of the Hatchobaru geothermal field where neutral pH deep water has been exploited for power production. Advanced argillic alteration zones composed of alunite, cristobalite, and kaolin are developed in/around the Jigoku due to the alteration by steam-heated waters. The occurrences and sulfur isotopic data of alteration minerals suggest that most of alunite formed under the present fumaloic activities, though some alunite data suggest a different condition and formation in the presence of high temperature volcanic gas.

1. INTRODUCTION

The Hatchobaru geothermal field is located on the northwestern slopes of Kuju volcano (about 1000 to 1100 m above sea level), northern central Kyushu (Figure 1). The geothermal system is a water-dominated type and fluid flow is controlled by fracture. The field is one of the most productive geothermal fields in Japan; Hatchobaru - Unit I (55MW) and Unit II (55MW) geothermal power plants have been operating since 1977 and 1990, respectively.

The northern part of the Hatchobaru geothermal field is a reinjection area, and hypogene acid alteration zones exist beneath the surface (Kiyosaki *et al.*, 2003); the alunite zone is more than **300** m in thick. At the surface above this acid alteration zone, there is steaming ground called Komatsu jigoku. The jigoku is the most active steaming ground in this field, and the total heat discharge by steam, conduction, and hot water from the jigoku was estimated to be 3.02×10^6 cal/sec in 1986 (Yuhara *et al.*, 1988). Acid alteration due to steam heated water is on going. Advanced argillic alteration zone composed of alunite is predominant in and around Komatsu jigoku (Hayashi, 1973). Any high temperature (>100 °C) volcanic gas activities are not observed at the jigoku at present. The nearest site where high temperature solfataras exsist is Kuju Iwo-yama, which is located about **4km** to the southeast of the field (Figure 1). Kuju Iwo-yama erupted in 1995 after 333 years of dormancy. The highest fumarole temperature was 508 °C in 1960 (Mizutani *et al.*, 1986), and before the recent eruption it was 320 °C.

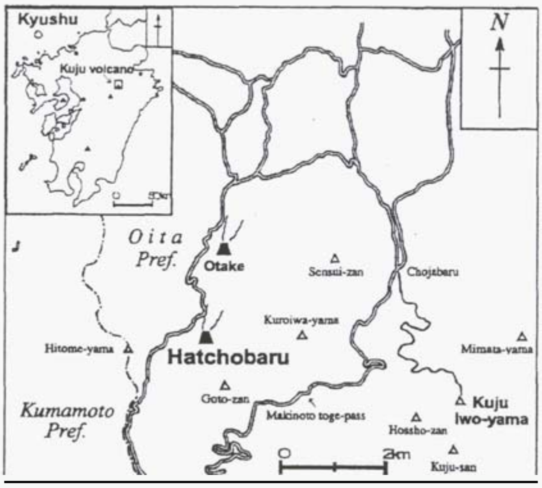

Figure 1 Location of the Hatchobaru geothermal field.

Hot springs in Kuju Iwo-yama area are $Cl\text{-}SO_4$ type with pH<2, and silicification and advanced argillic alteration are predominat (Kohmatsu and Taguchi, **200**1).

In this study, we considered the genesis of hydrothermal alteration at the surface area of Komatsu jigoku based on hydrothermal mineralogy and the geochemistry of minerals and hot springs. Some data indicate there was a contribution of volcanic gas to the formation of acid alteration zone in the past.

2. STEAMING GROUND

Many of steaming ground, hot springs, and hydrothermal alteration zones in this field are associated with northwest trending faults (Hayashi, 1973). One of the areas of steaming ground, Komatsujigoku, is located 300 m to the northeast of the Hatchobaru geothermal power plant. It is one of the most active areas of steaming ground in the field. The area is part of the Hatchobaru geothermal field, and is a good reinjection area for geothermal water.

The elevation of the steaming ground is about 1050m above sea level, extending 350 m long and 70 m width north - south along the Komatsu-gawa stream. Advanced hydrothermal alteration (alunite zone and kaolin zone) are dominant in and around the jigoku showing a lenticular in shape (Figure 2; Hayashi, 1973). Two zones are composed mainly of alunite + cristoballite, and kaolinite + cristobalite, respectively. The former is usually surrounded by the latter.

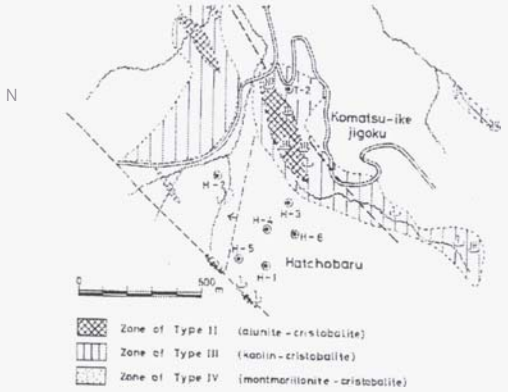

Figure 2 Hydrothermal alteration map of the Hatchobaru geothermal field (Hayashi, 1973).

Steaming ground in Komatsu jigoku is devided into two areas; southern and northern parts (Figure 3). In the southern area, many fumaroles, small mud pools and hot springs are distributed along a tourist trail. Mud pools are mainly found in the northern part of the southern area, while strong fumarolic activity occurs in the southern part. Most of the northern area is covered by landslide debris which occurred at the time of heavy rain during July 9 – 10, 1997. Mud volcanoes of a few meter size are found at the southern edge of the head scars of the landslide.

The Hohi and the Kuju volcanic rocks are the only units exposed in this area. The late Pliocene to early Pleistocene Hohi volcanic rocks composed of pyroxene andesites (1000 m thick) outcrop to the west of the jigoku, and are altered to kaolinite. To the east of Komatsu jigoku, the Kuju volcanic rocks are mainly composed of hornblende andesite. As the rocks of Komatsu jigoku are almost completely altered by acid sulfate waters, it is hard to identify the origin of the altered rock. According to the results of the drilling near the jigoku (HT-2), the top 50 m are composed of the Kuju volcanics (Hayashi, 1973).

3. TEMPERATURE DISTRIBUTION

Suburface temperature was measured at a depth of 50 cm from 170 points (Figure 3). The high temperature zones higher than 80 degree C is almost equivalent to sites of steaming ground. The size and the extent of the high temperature area is similar to that observed in 1986 (Yuhara *et al.*, 1988). In the northern area where there is landslide debris, the high temperature zone is widest at the head scars of the landslide. This zone was not detected in 1980, but firs detected in 1986 (Yuhara *et al.*, 1988). Comparison with an earlier survey (Yuhara *et al.*, 1988) suggests the southern area of Komatsu jigoku has not changed, however the vicinity **of** the head scars of the landslide in the northern area is increasing thermal activity.

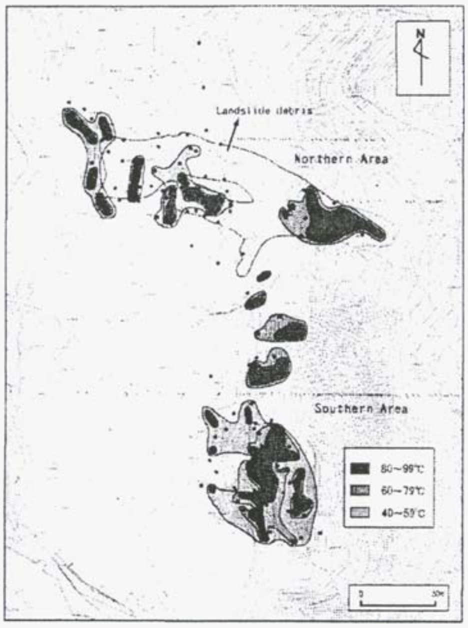

Figure 3 Temperature distribution at Komatsu jigoku. Measured depth is 50 cm.

4. ALTERATION MINERALS

4.1 Alteration minerals

About 100 altered rock samples were collected to investigate detail distribution of alteration minerals. Alteration minerals were identified by XRD analysis. According to the mineral assemblages, alteration zones were grouped into next four zones according to the criteria by Hayashi (1973) ; silica, alunite, kaolinite, and smectite zones. The distribution of each alteration zone is shown in Figure 4.

The Silica zone is newly defined, and is subdivided into two groups; Silica I and Silica II. The Silica I sub-zone is mainly composed of cristobalite with minor anatase, sometimes associated with minor tridymite, **quartz,** and pyrite. This sub-zone is usually small and found near the active steaming sites (>80 °C, Figures 3 and 4).

The major mineral of the Silica II sub-zone is quartz, and with small amount of anatase, pyrite, and cristobalite sometimes are found as accessory minerals. The Silica II sub-zone occurs in the southern area, where temperatures are relatively cool (Figures 3 and 4). Silicified rock is largely massive, but in part are vuggy. As quartz is usually formed above **100 °C,** the existence of the Silica **II** sub-zone suggests it formed under conditions different from present steaming activities.

The alunite zone is widely distributed in the steaming ground as mentioned by Hayashi (1973). Alunite is relatively abundant in the southern area as shown in Figure 5. This zone is also subdivided into two groups based on associated minerals. The Alunite I sub-zone is composed of alunite and cristobalite, and lacks kaolinite. Sometimes quartz, tridymite, anatase, pyrite and sulfur are found as minor phases. Compared with the Alunite II sub-zone, pyrite is less abundant and trydimite and sulfur are found. The Alunite I sub-zone usually coincides with steaming ground of > **80** °C. The main minerals of the Alunite II sub-zone are alunite, cristobalite, and kaolinite. Compared with the Alunie I sub-zone, tridymite is less, and pyrite is more **often** appeared. Minor quartz and anatase are sometimes included, and jarosite rarely occurs. The Alunie II sub-zone usually forms a large part of Komatsu jigoku surrounding the Alunite I zone (Figure **4).**

The Kaolinite and smectite zones are found on the periphery of the Alunite II sub-zone (Figure **4).** The main minerals are kaolinite + cristobalite + pyrite, and smectite + cristobalite, respectively.

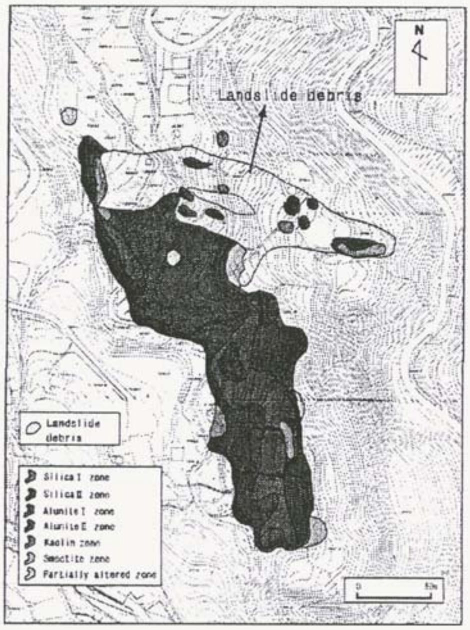

Figure **4** Distribution of alteration zones at Komatsujigoku.

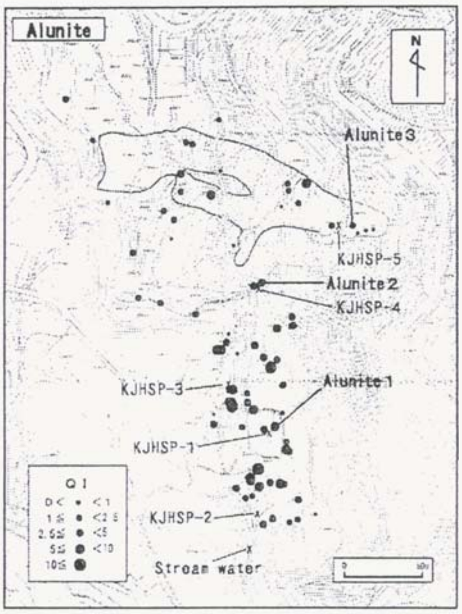

Figure **5** Alunite contents in the altered rocks and sample location of alunite for isotope analysis. QI: quartz index indicating relative contents of minerals (Hayashi, 1979). Locations of hot spring samples (**KJHSP-1** to *5*) are also shown.

4.2 Sulfur isotopic analysis of alunite

Three alunite samples for sulfur isotopic analysis were selected (Alunite 1 to **3);** two from the southern area and one is from the northern area (Figure 5). Alunite 1 was collected from the Alunite **I** sub-zone in the central part of the strongest steaming ground in the south area, and Alunite **2** from the Alunite II sub-zone at the northern edge of southern area. Alunite **3** was collected from the Alunite II sub-zone at the head scars of the landslide in the northern area. The obtained $\delta^{34}S$ values and mineral paragenesis of the samples are shown in Table 1. Alunite 1 and 2 show low sulphur isotope ratios indicating oxidation of H_2S. However, Alunite 3 has a high $\delta^{34}S$ of 11.0 per mil.

Table 1 $\delta^{34}S$ of hydrothermal alunite

Sample Name	Sample Number	δ34S (‰□	Alteration minerals(QI)			
			Cri	Qtz	Alu	Kao
Alunite 1	OK02504-13	0.6	7	8	8	0
Alunite 2	OK02050512	2.7	7	3	3	2
Alunite 3	OK-2050524	11	3	5	2	4

5 HOT SPRING CHEMISTRY

Five hot spring waters were collected from the steaming ground; four of them are from the southern area and one from the northern area (Figure *5).* Chemistry of hot springs is shown in Table *2* with isotopic data of water and sulfur. Stream waters near the fumaroles are also shown in the same table. Most of the hot spring waters are acid with pH=2.2 to 3.3. One sample, KJHSP-

2, is neutral pH and comes from the most active steaming part in the south area; here the largest discharges occur at several 10's of liters per minute. Discharges of other four hot springs are a few liters per minute or less. Chloride contents of all samples are low. The sulfate contents show a large variation 47.9 to 1712 **mg/l.** The hot spring water with the highest of sulfate (KJHSP-1) has the lowest pH, and the neutral pH hot spring water (KJHSP-2) has the lowest sulfate. These acid hot spring waters are characteristic of steam heated waters. The neutral one is probably less affected by oxidation of H_2S due to the **high** discharge.

Table 2. Chemistry of hot springs and stream water from the Komatsu jigoku steaming ground.

	KJHSP-1	KJHSP-2	KJHSP-3	KJHSP-4	KJHSP-5	River water
Tem,□	87.7	89.1	95.0	94.2	77.1	18.9
pH	2.24	6.58	2.32	3.25	3.24	7.09
Li	0.00	0.00	0.01	0.01	0.02	0.00
Na	4.87	6.01	11.90	7.86	17.12	4.39
K	6.23	2.77	9.90	4.47	10.32	2.21
Mg	2.71	3.92	9.83	5.01	6.52	1.80
Ca	5.68	12.23	20.51	11.75	27.81	5.94
F	0.11	0.07	0.42	0.11	0.14	0.05
Cl	0.56	1.78	0.88	1.36	1.51	3.10
NO3	0.00	0.08	0.00	0.00	0.00	0.00
HC03			-	-	-	
SO4	1712	47.9	1292	279	265	17.3
δ□	-28.9	-56.1	-37.7	-41.4	-45.7	-56.8
$\delta^{18}O$	-0.3	-8.5	-5.0	-3.6	-4.8	-8.9
$\delta^{34}S(SO_4)$	-4.19	-3.61	-4.09	-3.65	-3.03	

Values in chemistry:mg/l, isotope ratio: **per mil**

The isotopic **data** of the hot spring and stream water are also shown in Table 2. The hot spring with the neutral pH and high discharge in Komatsu jigoku (KJHSP-2) has almost the same values of the δD and $\delta^{18}O$ as that of local meteoric water. However, the data of other acid hot springs have heavier values. This is probably due to evaporation at the surface. $\delta^{34}S$ of sulfate ion in the hot springs are light ranging -3.01 to -4.19 per mil, indicating oxidation H_2S.

6. DISCUSSION

There are two group of alunite with different isotope ratios. One has a light $\delta^{34}S$ (0.6 to 2.7 per mil), and the other is heavy (11.0 per mil). Formation of alunite with light sulphur isotope probably formed under the present conditions where H_2S is oxidized to sulphate in the presence of atmospheric O_2;

$$H_2S + 2O_2 = H_2SO_4$$

Sulphate ions formed **through** this process has **a** light sulphur isotope ratio derived from H_2S. $\delta^{34}S$ of SO_4 ion in hot spring waters from the Komatsu jigoku are light ranging -3.0 to 4.2 per mil, indicating those are formed **through** this process. As shown in Figure *6***,** the origin of these water is derived **from** local meteoric water. Condensates of magmatic steam Kuju Iwo-yama, which contains SO_2 and HCl, plot on a different trend. Local meteoric water and the hot springs with neutral pH plot in almost same position. These suggest that such acid waters are formed by the oxidation of H_2S followed by evaporation at the surface.

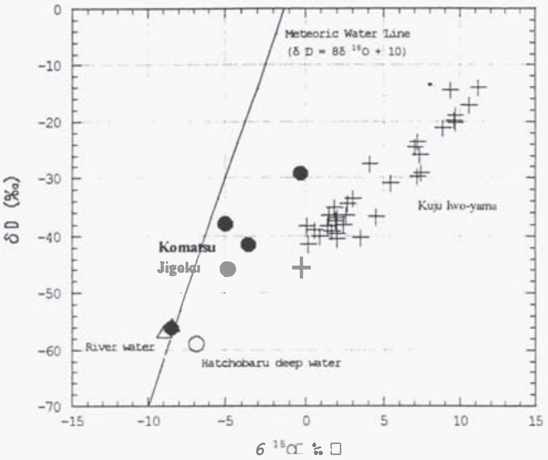

Figure 6 Isotope of waters from the Komatsu jigoku. Data of Kuju Iwo-yama, and Hatchobaru deep water are from Mizutani et al.(1986) and Shimada et al.(1985), respectively.

The alunite with heavy sulphur isotope ratio may have formed from high temperature magmatic steam. Kohmatsu and Taguchi (2001) reported silicification and advanced argillic alteration at Kuju Iwo-yama where high temperature solfataric activity has been active. Alunite from Kuju Iwo-yama has 2 to 12 per **mil** of $\delta^{34}S$ (Taguchi ***et*** *al.* **,** 2002), and the range is almost the same as SO_2 gas **from** Kuju Iwo-yama of 2 to 10 per mil (Mizutani et al., 1986). Taguchi ***et*** *al.* (2002) suggested this **is** introduced by the following process,

$$SO_2 + H_2O = H^+ + HSO_3^-$$

$$2\ HSO_3^- + O_2 = 2H^+ + 2SO_4^{2-}$$

This process leads the isotope ratio equivalent to that of SO_2 in magmatic gases. Usually, **high-**temperature magmatic gas vapour leads to much heavier sulphur isotope of sulfate ion than that of SO_2 through disproportionation; e.g.

$$4SO_2 + 4H_2O = 3H_2SO_4 + H_2S$$

However, the sulphur isotope of alunite from Kuju Iwo-yama suggests that disproportionation did not occur near the surface. ***So,*** the alunite with the heavy sulphur isotope ratio from Komatsu jigoku may have originated in a way similar to Kuju Iwo-yama.

This interpretation is also supported by the presence of Silica II sub-zone at the surface of Komatsu jigoku. This is usally formed under the condition of pH<2 in magmatic hydrothermal environments (Hedenquist ***et*** *al.*, 2000).

These facts suggest that once there was a magmatic gas discharge in this area, during which time the Silica II sub-zone and some of alunite was formed. Later the surface was eroded and succeeded *to* the present thermal activity.

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