

## SUBSURFACE HIGH TEMPERATURE HYPOGENE ACID ALTERATION AT THE HATCHOBARU GEOTHERMAL FIELD, KYUSHU, JAPAN

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**SUMMARY**-Alteration minerals and their distribution were examined in the northern part of the Hatchobaru geothermal field, especially alunite and other acid alteration minerals. Alunite minerals are widely distributed with more than 300 meters thickness at the northern part of the field. Alunite minerals have a wide range of  $\delta^{34}\text{S}$  from 0 to 24 per mil, indicating several different origins. Newly found acid minerals such as zunyite, topaz, and andalusite suggest that volcanic acid sulfate solutions were once present and was responsible for the formation of the thick acid alteration zone at the Hatchobaru geothermal field.

### 1. INTRODUCTION

The Hatchobaru geothermal field is located on the northwestern **flank** of Kuju volcano, central Kyushu, Japan. The field is water-dominated and located about 4km to the northwest of Kuju Iwo-yama where solfataric activity occurs. The Hatchobaru geothermal power plant at 1,100 m in elevation **has** been operating since 1977 with 55MW; another 55MW was installed in 1990. At 110 MW, it is the largest geothermal power plant in Japan.

The northern part of the field is called Komatsu jigoku and it is the site for reinjection and steaming ground. The jigoku is the most active steaming ground in and around the field, and acid hydrothermal alteration zones (advanced argillic alteration zones) composed of alunite and/or kaolinite **are** dominant. Hayashi (1973) reported that the alunite and the kaolin-pyrophyllite zones at the subsurface of the jigoku area reach depths of 380 m and 1000 m, respectively, with a funnel-like shape. The formation mechanism of such deep acid alteration has not been previously discussed, though modern steam-heated acid sulfate water is believed to play a major role.

The purpose of this report is to present a formation mechanism for this deep acid alteration zone in the northern part of the Hatchobaru geothermal field. We re-examined alteration minerals of the cores and cuttings from wells HT-2, HR-7, and HR-21 through microscopic observation, XRD and EPMA analyses.

Moreover, sulphur isotopes were measured on alunite, anhydrite, and pyrite. The results suggest that at some time in the past, volcanic gas and water emissions discharged and formed a high temperature volcanic acid alteration zone.

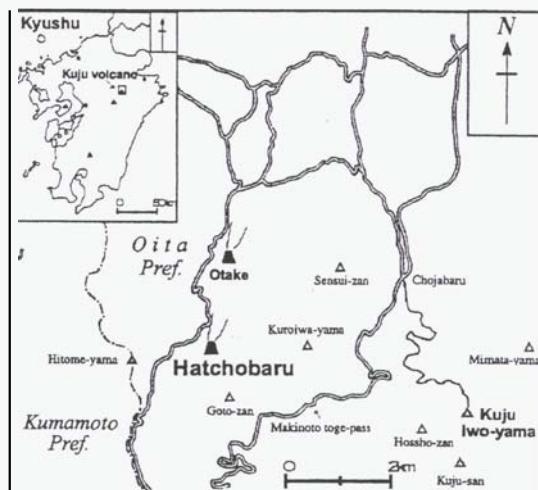


Figure 1 Location of the Hatchobaru geothermal field.

### 2. GEOLOGIC SETTING

The basement rocks, confirmed by drilling to 750 m below sea level, are crystalline schists intruded by Cretaceous granites. The Usa Group of Miocene to Pliocene age, about 700 m thick, unconformably overlies the basement rocks (Manabe and Ejima, 1984). The late Pliocene to early Pleistocene Hohi volcanic rocks composed

of pyroxene andesites (~1,000 m thick) overlie the Usa Group and crop out locally, surrounded by exposures of middle to late Pleistocene Kuju volcanic rocks. The Hohi and Kuju volcanic rocks crop out in the northern part of the Hatchobaru field.

Northwest trending faults dominate the area (Fig. 2) and are accompanied by many active thermal manifestations such as hot springs, fumaroles and advanced argillic alteration. Of these, the Komatsuike fault is the most remarkable. The most active and largest steaming ground in and around the Hatchobaru geothermal field, Komatsu jigoku, is located on this fault which is also a reinjection area for the steam field. At the center of the jigoku, a large lenticular body of alunite occurs along the fault (Hayashi, 1973). This alunite zone reaches to a depth of 380 m in well HT-2, and below this a kaolin-pyrophyllite zone extends to a depth of about 1,000 m, forming a funnel-like shape (Hayashi, 1973). Diaspore also occurs at deeper than 550 m within the kaolin-pyrophyllite zone. These two acid alteration zones are thus conspicuously developed around Well HT-2 (Hayashi, 1973).

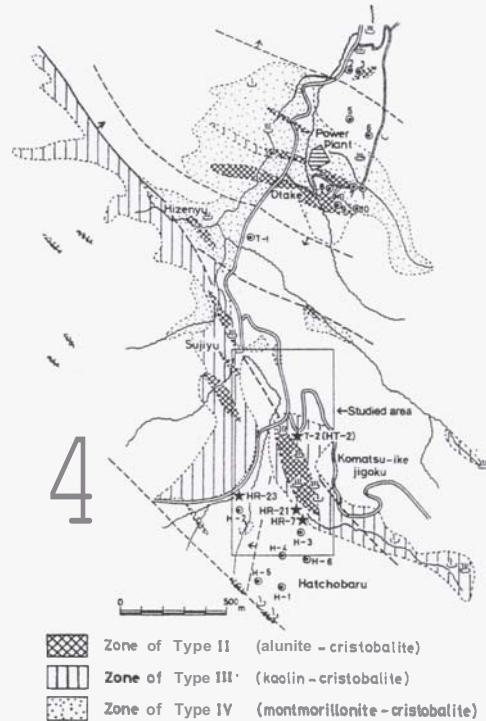


Figure 2. Hydrothermal alteration map of the Hatchobaru geothermal field by Hayashi (1973). Well location studied this time are shown.

### 3. SUBSURFACE ACID ALTERATION

Based on the alteration mineral distributions in more than 100 wells, we selected three wells for detailed study of acid alteration, HT-2, HR-7, and HR-21 from the reinjection area adjacent to the Komatsu jigoku.

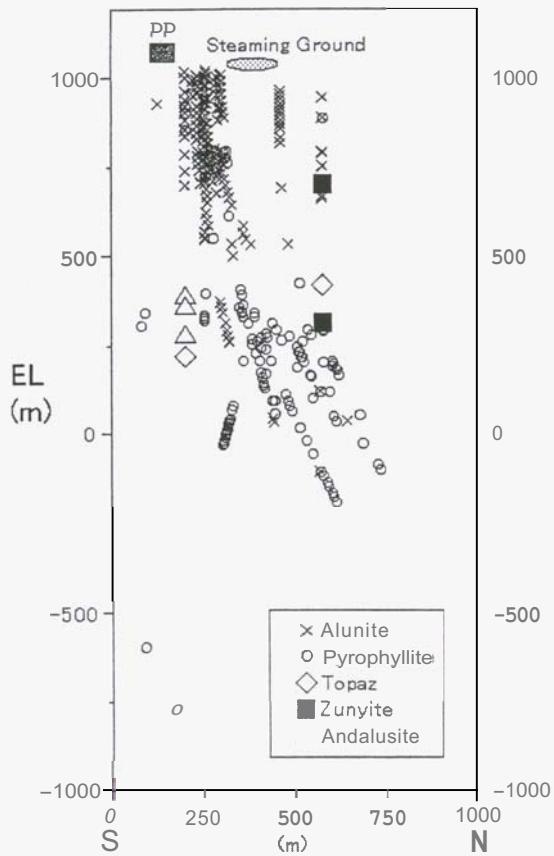


Figure 3. Distribution of hydrothermal alteration minerals on a N-S cross section at the northern part of the Hatchobaru geothermal field.

The occurrences and distributions of alteration minerals were determined by microscope work, XRD and EPMA analysis, and the results are shown in Figures 3 and 4. As shown in Figure 3, the alunite and kaolin-pyrophyllite zones show a wider distribution than that mentioned by Hayashi (1973).

#### 3.1 Alunite zone

As shown in Figure 3, the alunite zone extends from the surface to 350-370 m depth in the three wells, and rarely to deeper level in the other wells. Common minerals at <150 m are alunite, cristobalite, kaolinite, and pyrite. Tridymite, quartz, dickite, pyrophyllite, and anhydrite are associated minerals that sometimes occur. Dickite and pyrophyllite occur at depths of 84.8 m and 140.3 m in well HT-2, respectively. In the deeper zone, alunite, quartz, dickite, and pyrite are common. Often anhydrite, and rarely smectite, kaolinite, pyrophyllite, rutile, native sulfur, and zunyite are associated with these phases. Pyrophyllite occurs at 260.5 and 400.0 m depth in well HR-7. Zunyite is found at depths of 325.7 and 326.5 m in well HT-2.

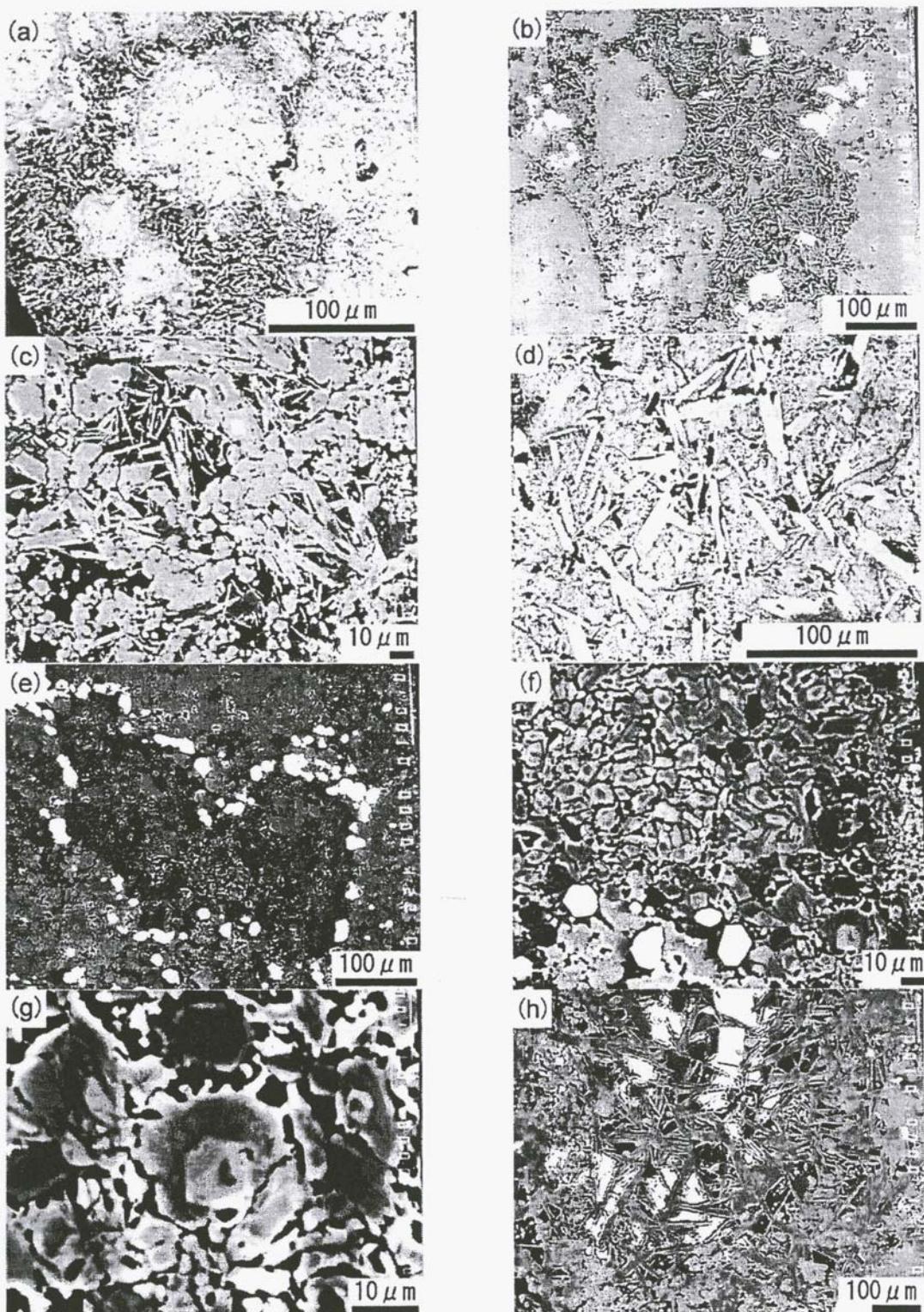


Figure 4. Backscattered electron image of alunite minerals from the alunite zone based on cores and cuttings from the northern part of the Hatchobaru geothermal field. (a). Intergrown tabular alunite, sample **HR-7, 120.4 m**. (b) Intergrown tabular alunite and pyrite (bright minerals), sample **HR-7, 276.0 m**. (c) Tabular and granular alunite, sample **HR-7, 322.0 m**. (d) Tabular alunite, sample **HT-2, 140.3 m**. (e) Granular alunite surrounded by pyrite (bright minerals), sample **HT-2, 326.5 m**. (f) and (g) Enlargement of alunite (from (e)) with potassium-rich cores and potassium-poor rims, suggesting several stage of alunite growth. (h) Minamiite, calcium-rich alunite from **HT-2, 359.8 m**. relatively large tabular crystal intergrowth, bright part is pyrite.

Alunite is K-rich with  $K/(K+Na)=0.8\pm0.2$  as determined by the XRD method of Cunningham & Hall (1976). Minamiite (Ca-rich alunite) occurs in well HT-2 at 359.8 and 368.3 m depth. Minamiite is associated with quartz, anhydrite, pyrite and rutile.

Some of alunite minerals from HT-2 and HR-7 are observed under EPMA (Fig. 4). Alunite from shallow to medium depths has a tabular habit and ranges from  $<10$   $\mu\text{m}$  to a few 10s of  $\mu\text{m}$  (Fig. 4). Alunite also shows a granular habit (Fig. 4). Granular alunite cores are bright in BSE image (Fig. 4g) indicating a potassium-rich composition; the dark rims are poor in potassium and rich in calcium according to the semiquantitative EPMA analysis. Minamiaite has a tabular habit (Fig. 4h), and the BSE image indicates the calcium-rich part formed later.

### 3.2 Kaolin-pyrophyllite zone

The kaolin-pyrophyllite zone extends below the alunite zone. Dickite is common with other kaolin minerals in the upper part of this zone whereas pyrophyllite occurs in the lower zone. Kaolinite occurs over a depth range of  $<200$  m to 550 m. Pyrophyllite occurs with quartz, anhydrite and pyrite from 320m to 550m depth. Diaspore is found in this zone with quartz, rutile and anhydrite, at depths below 710 m and 840 m for HT-2 and HR-7, respectively. Andalusite, was also identified in this zone by XRD analysis, at depths of 670-710m and 782 m in HR-7.

Topaz and zunyite, halogen enriched minerals, were identified from XRD analysis (Fig. 3). Topaz occurs at depths of 607.9 m in HT-2 and 842.6 m in HR-7; quartz and dickite, and zunyite appear about 326 and 712 m in depth in HT-2. Zunyite occurs with quartz, alunite, and anhydrite at shallow levels, and pyrophyllite and dickite at deeper levels.

## 4. SULFUR ISOTOPE COMPOSITIONS OF ALTERATION MINERALS

Sulfur isotopic compositions of alunite, anhydrite, pyrite and native sulfur from the acid and neutral alteration zone were obtained, and the data are shown on Figures 5 and 6. Alunite has a wide range of  $\delta^{34}\text{S} = 0$  to 24 per mil. The values are increase with depth, and they are 16 to 24 per mil in the deeper part of alunite zone. Alunite associated with zunyite has 17 per mil of  $\delta^{34}\text{S}$ . On the other hand, alunite minerals forming now in the presence of steam-heated waters are 0 to 3 per mil  $\delta^{34}\text{S}$  (Kiyosaki et al, this volume). Most anhydrite has 18 to 24 per mil  $\delta^{34}\text{S}$ , and the range is almost the same of the present-day geothermal water (Kiyota et al., 1996). Native sulfur, pyrite and  $\text{SO}_4$  in hot spring water are around 0 per mil  $\delta^{34}\text{S}$ .

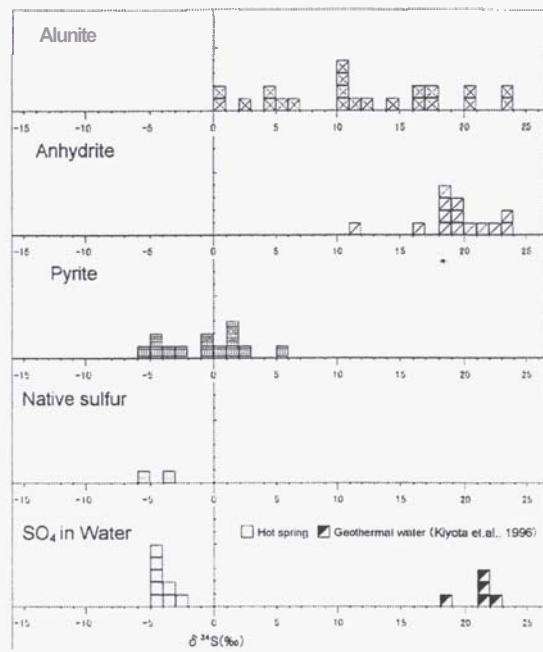


Figure 5. Histograms of  $\delta^{34}\text{S}$  of alteration minerals from the Hatchobaru geothermal field. Data of geothermal waters by Kiyota et al (1996) are also shown.

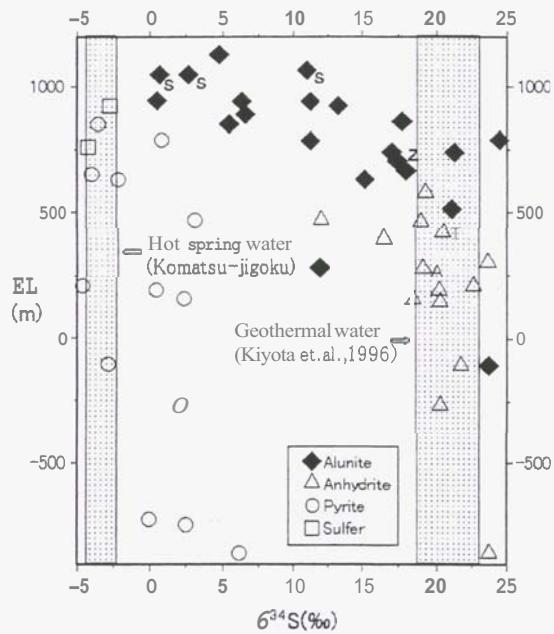


Figure 6. Sulfur isotopic ratios of alteration minerals as a function of depth. S:surface sample, Z:zunyite, T:topaz and A:andalusite.

## 5. DISCUSSION

The formation a thick subsurface alunite zone in and around the Komatsu jigoku steaming ground is believed to have formed under two different conditions. One is related to steam-heated acid sulfate water which forms by oxidation of  $\text{H}_2\text{S}$  in modern steaming ground of Komatsu jigoku. The other involves a direct contribution of  $\text{SO}_2$  in volcanic gas and/or a disproportionation of  $\text{SO}_2$

Some of shallow alunite with less than 5 per mil of  $\delta^{34}\text{S}$  probably originated from steam heated water, because alunite and water with such light sulfur isotope ratio are reported from the Komatsu jigoku (Kiyosaki et al, this volume). On the other hand, the  $\text{SO}_4$  in Hatchobaru geothermal water has 19 to 23 per mil  $\delta^{34}\text{S}$  (Kiyota et al., 1996).

As the  $\delta^{34}\text{S}$  of alunite increases with depth from 0 to 20 per mil (Fig. 6), mixing of light  $\delta^{34}\text{S}$  from steam-heated acid sulfate water and heavy  $\delta^{34}\text{S}$  of present deep geothermal waters are inferred. However, simple mixing of the two waters (steam heated water and present deep geothermal water), does not explain the occurrence of deep zunyite. Zunyite is usually forms under strong acid conditions from halogen-bearing fluids as is commonly associated with volcanic gas. Moreover, topaz, andalusite, and diasporite are found in the kaolin-pyrophyllite zone which lies below the alunite zone. Such mineral occurrence suggests acid alteration formed in the presence of volcanic gas sometime in the past. Although the sulfur isotope of the deep alunite is close to the  $\text{SO}_4$  in the present geothermal water, zunyite occurrence with alunite suggests instead the presence of a strong acid fluid caused by disproportionation of  $\text{SO}_2$  gas.

Alunite which is forming in the high temperature sulfatara area at the surface of Kuju Iwo-yama, 4km to the southeast of the Hatchobaru geothermal area, shows 4 to 12 per mil  $\delta^{34}\text{S}$  in a fracture filling alunite (Taguchi et al., 2002), and the heaviest value was found in a breccia dyke indicating a deep origin. As the most of the  $\text{SO}_2$  gas is in the range of 4 to 10 per mil  $\delta^{34}\text{S}$  (Mizutani et al., 1986), these alunite  $\delta^{34}\text{S}$  values are interpreted as having formed in the presence of volcanic  $\text{SO}_2$  gas. Alunite minerals with about 12 per mil  $\delta^{34}\text{S}$  occur from the surface to 300 m in depth at the Hatchobaru field in a situation that is probably similar to that in the breccia dyke at Kuju Iwo-yama, reflecting a direct contribution of volcanic gas.

Thus, three sulfate-bearing fluids (ie. steam-heated water, direct contribution of  $\text{SO}_2$  volcanic gas, and disproportionation of  $\text{SO}_2$  gas) and their mixtures probably affected the sulfur isotopic composition of alunite in the acid alteration zone in the northern part of the Hatchobaru geothermal field.

The changes in alunite composition, from core to rim (eg. Fig. 4), suggests fluids of different compositions reacted with the minerals.

## 6. ACKNOWLEDGEMENTS

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